



HEAVY MINERAL CATALOGUE FOR THE CHARACTERIZATION AND PROVENANCE OF RED SANDSTONE MACROLITHIC TOOLS FROM HÓDMEZŐVÁSÁRHELY-GORZSA

HÓDMEZŐVÁSÁRHELY-GORZSA LELŐHELYRŐL SZÁRMAZÓ VÖRÖS HOMOKKŐ SZERSZÁMKÖVEK OSZTÁLYOZÁSÁRA ÉS PROVENIENCIA MEGHATÁROZÁSÁRA ALKALMAZOTT NEHÉZÁSVÁNY KATALÓGUSA •

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Jelen tanulmány nem születhetett volna meg Szakmány György iránymutatása és munkássága nélkül

Abstract

Although ground stone tools (GSTs) are among the most abundant stone tools in archaeological sites, they are rarely investigated both from the archaeological and the geological point of view. There are many pieces made of siliciclastic sedimentary raw material. Their fragmentary condition makes it possible to examine them by slightly destructive methods, such as HMA (heavy mineral analysis), which was developed primarily for siliciclastic rocks. Until now, ‘sensu stricto’ (s.str) heavy mineral analysis has not yet been carried out for ground stone tools, neither at the national nor at the international level. By comparing the heavy mineral compositions of the stone tools and their potential raw materials with the help of a newly developed HM descriptive system (including rutile, zircon, tourmaline, apatite and titanite), it is possible to determine the provenance of these archaeological materials.

From the Neolithic tell of Gorzsa, 109 red sandstone macrolithic stone tools are known. These were separated into four, different types (‘Red-1’ – ‘Red-4’), based on the macroscopic and microscopic analysis. 11 fragments were chosen for HMA analysis. In addition, red-coloured sandstones from different geological occurrences of the Carpathian-Pannonian Basin (22 pieces) were also investigated. Based on the heavy mineral analysis, all raw material types of the stone tools could be originated from the Maros River and the Apuseni Mountains (Transylvania). However, the previously determined potential sources (Permotriassic and Miocene sequences from the Mecsek Mountains, Balaton Highlands, pebbles from the Danube River, and Papuk Mountains) cannot be excluded with absolute certainty from the possible raw materials.

Kivonat

A szerszámkövek az ásatások alkalmával feltárt kőzet anyagú régészeti leletek tetemes hányadát teszik ki. Mindezek ellenére ezen lelettípusok régészeti, valamint természettudományos vizsgálata eddig háttérbe szorult. Nyersanyagukat tekintve jelentős többletet képviselnek a törmelékes üledékes eredetű változatok, amelyek gyakori töredékes megjelenése akár a roncsolásos vizsgálatok, mint például a nehézasvány analízis, alkalmazását is lehetővé teszik. Szerszámkövek esetében a hagyományos értelemben vett, azaz dúsítós eljárás eddig mind hazai, mind pedig nemzetközi szinten is kiaknázatlan maradt. Célunk a régészeti, valamint a lehetséges nyersanyagok nehézasvány összetételének összevetésével meghatározni azok provenienciáját, egy erre

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a célra, valamint a vörös homokkő nyersanyagokra kifejlesztett leíró rendszer (rutil, cirkon, turmalin, apatit és titanit) segítségével.

A neolitikus korú gorszai tell településről 109 vörös homokkő anyagi szerszámkő ismert, amelyeket a makroszkópos és mikroszkópos vizsgálatok alapján négy különböző típusba (vörös-1 – vörös-4) soroltunk. 11 szerszámkő töredéket, továbbá 22 vörös homokkő anyagi összehasonlító geológiai mintát választottunk ki nehézasvány vizsgálatra. A nehézasvány eredmények alapján a homokkő anyagi szerszámkövek feltehetően a Maros folyó hordalékanyagából, illetve az Erdélyi-középhegységből származhatnak. Az eddigi eredményeink alapján több, korábban potenciálisnak tekinthető nyersanyagforrás (permotriász és miocén mecseki homokkövek, Balaton-felvidék, a dunai kavicsok, valamint a Papuk-hegységből származó homokkövek) anyagát nem tudjuk teljes mértékben kizárni a lehetséges nyersanyag források közül.

KEYWORDS: HEAVY MINERAL, PROVENANCE, RAW MATERIAL, NEOLITHIC MACROLITHIC TOOLS, RED SANDSTONE

KULCSSZAVAK: NEHÉZÁSVÁNY, PROVENIENCIA, FORRÁSKÖZET, NEOLITIKUS SZERSZÁMKÖVEK, VÖRÖS HOMOKKÖVEK

Introduction

Examination of the pebbles and cobbles of alluvial sediments and tracing suitable raw materials were important activities for prehistoric communities. Their tools were made of 'quality' stones of physically and chemically resistant materials. Finding the origin of the potential raw materials, provides information for understanding past societies and human behaviour and interaction with their environment. For this, the comparison of the different raw materials – archaeological finds and potential geological samples ('sources') – is necessary. The potential sources of the various rock types can be distinguished with different efficiency. For crystalline rocks (e.g. andesite, mica schist, eclogite) it can be easy, because they are commonly composed of, so-called rock-forming (e.g. amphiboles, pyroxenes, olivine) minerals. Based on this, they can be differentiated. In the case of sedimentary rocks (e.g. sandstone), that are stones formed during the sedimentary cycle, they are also influenced by processes acting on the surface (weathering – transport – deposition – diagenesis). Sandstone is a siliciclastic sedimentary rock, its grain size is between 0.063–2 millimetres. Four components are to be considered, namely grains, matrix, cement and pores. The first can have multiple geneses of which detrital ones are classified into two main categories: main components (i.e. 'light minerals', e.g. quartz, feldspar, micas, that have very similar composition, so they can be hardly differentiated) and heavy minerals. Heavy mineral analysis deals with the determination of accessory minerals, with high density but occurring in small quantity (that rarely makes up more than one percent of the whole rock/sediment) and size. Accessories ('heavy minerals'), are zircon, apatite, tourmaline, garnet, rutile etc. The modern geological research on sources of sedimentary rocks started in the 19th century (Ludwig 1874; Meunier 1877; Michel Lévy 1878; Thürach 1884; Dick 1887; Artini 1898). All these works were based on the heavy mineral

analysis (HMA or micromineralogy). Heavy minerals can be studied via two main methods: 1) thin section analysis and 2) 'HMA s.str.' (see 'Materials and methods').

Each heavy mineral (HM) grain carries information about the genetics and lithology of its source rock. Composition of sandstones is determined by the composition of the source rocks and by additional factors (e.g. weathering, mechanical abrasion, hydraulic behaviour, and burial diagenesis) that operate during the sedimentation cycle. Identification and discrimination of provenance is based on the determination of provenance-sensitive features. Mechanical abrasion causes roundness and breakage of minerals, but does not identify the provenance signal, meanwhile weathering and especially burial diagenesis may cause HM loss due to dissolution. For example, the least stable phase under acidic weathering is apatite, whose absence may be the main proxy for acidic weathering. Burial diagenesis causes dissolution, but this depends on many factors, such as mineral stability, pore fluid temperature and composition. Consequently, the heavy mineral diversity decreases with increasing burial depth. HMA is a widely applied, high-resolution approach in provenance studies (Morton 1985, 1991; Morton & Hallsworth 1994, 1999; Lihou & Mange-Rajetzky 1996; Mange & Wright 2007). One of the main advantages of using HMA is that there is a wide variety of detrital heavy minerals in sandstones. For example, over fifty translucent detrital minerals were described by Mange & Maurer (1992). In addition, there are several opaque species as well.

In Hungary, traditional HMA research started in the early 20th century, when Aladár Vendl separated heavy minerals from sand by dense liquid and magnetic separator and examined them by optical microscope (Vendl 1910). Heavy mineral analysis, however, was only used by a few researchers (Sztrókay 1935; Miháltz 1937). Interest increased from the 1950s; the first results were qualitative (e.g. Hermann 1954; Molnár 1964, 1965; Csánk

1969; Gedeon-Rajetzky 1971, 1973). With the spread of grain counting techniques, quantitative, percentage-based compositional data were generated (data collected by Sallay 1984). Modern scientific research on sands and sandstones is still ongoing in present days (Thamóné Bozsó 1991, 1993, 2000; B. Árgyelán & Császár 1998; B. Árgyelán & Horváth 2002; Thamó-Bozsó et al. 2006, 2007, 2014; Obbágy 2017; Pozsgai et al. 2017, Józsa et al. 2020).

Heavy mineral analysis in petro-archaeological studies

The discipline of archaeometry evolved as a result of the interweaving of archaeology and natural sciences. One of the most commonly applied disciplines is petrography, as raw materials of archaeological artefacts are different types of stones or clays. Furthermore, other inorganic, artificial materials, such as ceramics, daub, plaster etc. are also predominantly of geological origin, so they can be best analysed by techniques used in geological and other natural science laboratories.

Pottery sherds are very abundant artefacts at archaeological sites, and they appear in large quantities mostly fragmented. During clay manufacturing, other compounds, mostly sand (as 'non-plastic component' or 'temper') were added to the 'raw material'. Heavy minerals of the non-plastic components provide information about the provenance of the raw material, and manufacturing practices/processes. Archaeometric studies of potteries from different sites may provide information about trade and transport routes in ancient times (Mange & Bezeczky 2006, 2007; Dickinson 2007; Obbágy et al. 2014; Obbágy 2015; Józsa et al. 2016a,b; Szakmány et al. 2017). In most cases these minerals were analysed in thin sections (Kürthy et al. 2015, 2018).

For polished stone tools that are made of mostly crystalline rocks, HMA is not considered as a widespread technique. The 'HMA' method was developed primarily for siliciclastic rocks.

In contrast, the ground stone tools (GSTs) that are more abundant finds in archaeological sites, have less frequently been investigated both from the archaeological and the geological point of view. Most of them were made of siliciclastic sedimentary raw material. Their fragmentary appearance, together with their large amount within the finds, makes it possible to examine them with the help of slightly destructive methods/examinations. Although archaeometric research on ground stone tools has been conducted before, detailed s.str heavy mineral analysis has not yet been carried out, neither at national nor at international level.

The stone tool assemblage of Gorzsa

Hódmezővásárhely-Gorzsa is one of the largest excavated tell site of the Late Neolithic Tisza culture in Hungary. Following some small-scale surveys, a systematic excavation was begun in 1978 until 1996 (Horváth 1987, 2005), where 1061 pieces of macrolithic finds were collected. In the site Bronze, Iron and Sarmatian Age finds were also found. A quarter of the macrolithic finds represents polished tools (i.e. axes, adzes and chisels) and three quarters of them are GSTs (e.g. grinding stones, abraders, whetstones, etc). Ground stone tools of Gorzsa tell were prepared mainly from sandstones (50%), andesite (7%), granitoid-metagranitoid (13%), micaschist-micaceous quartzite (9%), quartzite (10%), limestone and marl (2%) (Starnini et al. 2015; Szakmány et al. 2019; Miklós et al. 2021).

Piros (2010) was the only one, who distinguished the potential raw materials of the red-coloured ground stone tools. Two main types were identified, the 'Red-1' was originated from the Mecsek Mountains (Jakabhegy Sandstone Formation) and the 'Red-2' was from Transylvania (Krassó-Szörény Mts.; Piros 2010, after T. Roth 1888, 1889 and Pálffy 1897), Mecsek Mountains (Jakabhegy Sandstone and the pebbles from the Miocene sequence), Papuk Mountain and pebbles from the drainage of the Danube. In our previous research (Miklós et al. 2021) four different, red-coloured sandstone types ('Red-1', 'Red-2', 'Red-3' and 'Red-4') were identified. We also found that the determination of the potential source of the raw material was made only in the case of Gorzsa type 'Red-3' that was originated from the drainage of the Maros River. The origin of the other three types ('Red-1', 'Red-2' and 'Red-4') having great uncertainties, either the Mecsek Permian series, pebbles from the Danube River, or the Apuseni Mountains (Transylvania) can be considered as potential sources (Miklós et al. 2021).

This paper presents the heavy mineral analysis of some red sandstone fragments of grinding stones performed with the aim of identify their geological source. Our research is based on a previous petrographic analysis (see in Miklós et al. 2021). In addition, we also examined potential raw materials (geological samples), from red sandstone occurrences in Hungary and some territories beyond the border (see latter, '*Heavy mineral content of the examined red sandstones*').

Our goal is to establish a petrological methodology based on sandstone ground stone tools, in which heavy minerals play a key role. Moreover, we have developed a descriptive system that can be used during heavy mineral analysis. It can be applied to different heavy mineral species, such as tourmaline, rutile, zircon, apatite, or titanite, and with the help

of it, HM variants can be separated and characterized within each mineral species. Statistical data on the quantitative determinations can also be gained.

Materials and methods

Sampling strategy

The red sandstone tools, represent the 27% (109 pieces) among the GSTs made of sandstone. The

raw material of the lithic finds from Hódmezővásárhely-Gorzsa was classified on the basis of macroscopic and microscopic characteristics. With the help of thin sections, in total, 11 fragments of representative ground stone tools were chosen from the red sandstone types ('Red-1' – 'Red-4') for heavy mineral analysis (Table 1.). Another 22 geological samples (Table 1.) from different locations of the Pannonian Basin and its surrounding were also investigated.

Table 1.: The sandstone samples of different origin (tools and geological samples) examined by thin section and heavy mineral analysis

1. táblázat: A mikroszkópos petrográfiai és nehézasvány analízis módszerekkel vizsgált, különböző lelőhelyekről származó homokkövek (régészeti és geológiai minták) listája

Sample	Material	Type	Location
GOR-133	Ground stone tool	Type 'Red-1'	Hódmezővásárhely-Gorzsa
GOR-374	Ground stone tool	Type 'Red-1'	Hódmezővásárhely-Gorzsa
GOR-592	Ground stone tool	Type 'Red-1'	Hódmezővásárhely-Gorzsa
GOR-9	Ground stone tool	Type 'Red-2'	Hódmezővásárhely-Gorzsa
GOR-112	Ground stone tool	Type 'Red-2'	Hódmezővásárhely-Gorzsa
GOR-271	Ground stone tool	Type 'Red-2'	Hódmezővásárhely-Gorzsa
GOR-90	Ground stone tool	Type 'Red-3'	Hódmezővásárhely-Gorzsa
GOR-531	Ground stone tool	Type 'Red-3'	Hódmezővásárhely-Gorzsa
GOR-762	Ground stone tool	Type 'Red-3'	Hódmezővásárhely-Gorzsa
GOR-349	Ground stone tool	Type 'Red-4'	Hódmezővásárhely-Gorzsa
GOR-673	Ground stone tool	Type 'Red-4'	Hódmezővásárhely-Gorzsa
Codru-01	Geological	Cod	Codru-Moma Mts.
DVVH/1	Geological	Dan	Danube River (pebble)
DVVH/2	Geological	Dan	Danube River (pebble)
P02 18/a	Geological	Pap	Papuk Mts.
P02 19/b	Geological	Pap	Papuk Mts.
M-1/14	Geological	Mar	Maros River
M-1/18	Geological	Mar	Maros River
M-1/19	Geological	Mar	Maros River
Ba-KFhBaT	Geological	Mecs-Köv	Kővágószőlős F. (Mecsek Mts.)
Ja-JFhJS	Geological	Mecs-Jak	Jakabhegy F. (Mecsek Mts.)
Cs-JFh	Geological	Mecs-Jak	Jakabhegy F. (Mecsek Mts.)
Cs-JFhk	Geological	Mecs-Jak	Jakabhegy F. (Mecsek Mts.)
Kötő-6	Geological	Bal-Hgh	Balaton Highlands
P-1	Geological	Bal-Hgh	Balaton Highlands
P-3	Geological	Bal-Hgh	Balaton Highlands
IHCs	Geological	Mecs-PebI	Mecsek Miocene pebble
HH 98/6	Geological	Mecs-PebI	Mecsek Miocene pebble
LK007A/1	Geological	Mecs-PebII	Mecsek Miocene pebble
PF/1	Geological	Mecs-PebII	Mecsek Miocene pebble
HH 98/4	Geological	Mecs-PebIII	Mecsek Miocene pebble
HH98/5	Geological	Mecs-PebIII	Mecsek Miocene pebble
PF/3	Geological	Mecs-PebIII	Mecsek Miocene pebble

Detailed descriptions about the examined sandstone occurrences can be read in Miklós et al. (2021). Each selected archaeological sample has a representative composition within its sandstone type, the same applies to possible raw materials. For each geological occurrence (potential raw material), materials with a homogeneous composition were selected.

The result and the benefit of heavy mineral analysis depend on the accuracy of sampling; therefore, it should be consequently and thoroughly planned. It is very important to collect fresh (not altered) samples that exhibit fine-to-medium grain size. After Morton & Hallsworth (1994) in most of the cases, the very fine-grained sand fraction (63–125 µm) of the samples was used for the provenance analysis. This fraction is finer than the maximum size of the heavy minerals, so the samples are comparable with each other. The previously mentioned two procedures can be used for heavy mineral testing 1) thin section and/or 2) the 'HMA s.str.'. The most important difference between the two methods is the amount of the examination volume:

1) In the case of thin sections, we only get information from a relatively small surface and the heavy minerals might only appear in small amounts. They most often occur scattered, and they can also be absent. Therefore, in this case we do not get a representative result for the entire sample. Before applying the second method, it is important to prepare a thin section as it can be used to preselect the most suitable samples for the mineral enrichment, based on density rates.

2) For the HMA s.str., heavy minerals need to be separated from non-heavy minerals, using dense liquids (e.g. bromoform: 2.89, tetrabromoethane: 2.96, sodium-polytungstate: ca. 2.89–2.97 g/cm³). The last one is a non-toxic compound with adjustable density. Mineral grains with high-density sink down in these liquids, which permits their complete segregation from the less dense framework components (e.g. quartz, feldspar, carbonate and/or micas) (Mange & Maurer 1992). We can obtain representative information about the heavy mineral composition from a larger, homogenous volume for the entire sample. HMA s.str. were carried out using the technique described by Petelin (1961) as 'immersion method', which was described in detail by Mange & Maurer (1992) and Józsa et al. (2016b). After the sample preparation (Józsa et al. 2016b) about 300 transparent, randomly selected heavy mineral grains ('THM') were counted (ribbon counting) from heavy mineral mounts per sample. Identification was made based on the optical properties of each mineral type described by Mange & Maurer (1992).

For the heavy mineral analysis, we used a Leica DM 2700P polarizing microscope with attached Leica K5C camera and a Nikon Optiphot2-pol microscope with a Nikon CoolPixDS-Fil camera.

Results

Heavy mineral content of the examined red sandstones

Ground stone tools from Gorzsa

Samples of the type 'Red-1' contain a low amount (<0.5%) of heavy minerals, among which a large quantity of opaque minerals were detected. Transparent heavy mineral grains are zircon, rutile, apatite, tourmaline, titanite and a few amphibole and kyanite. Type 'Red-2' samples also contain low amounts, but higher than in the previous group (>1.5%) that are made of lots of opaque and some 'THM'-type (transparent heavy minerals) grains, such as tourmaline, rutile, titanite, zircon, apatite and a few amphibole and garnet. Samples of the type 'Red-3' contain small amounts of heavy minerals (>0.5%); among them there are zircon, tourmaline, rutile, titanite, amphibole and a few apatite, olivine, staurolite and epidote-zoisite/clinozoisite. Type 'Red-4' contains low amounts of heavy minerals (1<red-4<1.5%), out of which garnet, apatite, rutile, zircon, tourmaline, epidote-zoisite/clinozoisite, titanite and a few amphibole and kyanite are present.

Potential raw materials

Out of the many sandstone types present in the Carpathian-Pannonian Region, red is one of the most typical ones. They can originate from different geological localities, such as from primary outcrops: Permian succession of the Mecsek Mountains (Kővágószőlős and Jakabhegy Formations, SW Hungary), the Balaton Highlands (Balatonfelvidék Formation, NW Hungary), the Permian series from the Papuk Mountain (NW Croatia) and the Permian sandstone from the Codru-Moma Mts. (SW Carpathians, Romania).

Secondary sources (e.g. river drainages, terraces) were also studied during the investigation: pebbles from the recent debris of Maros valley (E Hungary, W Romania), from the Miocene siliciclastic sediments in Western Mecsek Mts. (Szászvár Formation, SW Hungary) and from the Pleistocene terraces of the Danube in Dunavarsány, previously belonging to Délpest Pebble Formation (NW Hungary).

Jakabhegy Formation (Mecsek Mts., 'Mecs-Jak') contains lower amounts (0.87%) of heavy minerals; among the transparent heavy minerals there are apatite, zircon/monazite, titanite, rutile and a few amphiboles and pyroxene. Samples from the Kővágószőlős Formation (Mecsek Mts., 'Mecs-Köv') contains relative higher amounts (1%) of

heavy minerals, where among the transparent heavy minerals, rutile, tourmaline, apatite, zircon/monazite, titanite and a few amphiboles were observed. Balaton Highlands ('Bal-Hgh') sandstones contain higher (1.88%) amount of heavy minerals, among which a large quantity of opaque minerals was detected. Transparent heavy mineral grains are zircon/monazite, tourmaline, rutile, titanite and a few amphiboles and garnet. Samples from the Papuk Mts. ('Pap') contain a low amount (0.07%) of heavy minerals. Among the transparent heavy minerals there are zircon/monazite, tourmaline, rutile, titanite and a few amphiboles. Codru-Moma Mts. ('Cod') contain a low amount of heavy minerals; among the transparent heavy minerals there are zircon/monazite, rutile, tourmaline, titanite and a few amphiboles and staurolite.

Pebbles from the Maros River ('Mar') contain high amount (1.55%) of heavy minerals, among the transparent heavy minerals there are zircon/monazite, tourmaline, rutile, titanite and a few apatite, garnet and epidote-zoisite/clinozoisite. Pebbles from the Danube River ('Dan') contain lower amounts (0.52%) of heavy minerals. Among the transparent heavy minerals there are zircon/monazite, tourmaline, and rutile and a few titanite, apatite and garnet. Pebbles from the Miocene siliciclastic sequence of the Mecsek Mts. contain a lot of heavy minerals. Group 'Red-I' ('Mecs-PeBI') contains high amounts (2.13%) of transparent mineral. These are tourmaline, rutile, zircon/monazite, apatite, titanite and a few staurolite, kyanite, garnet and epidote-zoisite/clinozoisite. Group 'Red-II' ('Mecs-PeBII') contains lower amounts (0.48%) of transparent mineral, such as apatite, rutile, tourmaline, titanite, zircon/monazite and a few garnet, epidote-zoisite/clinozoisite and staurolite. Group 'Red-III' ('Mecs-PeBIII') contains the highest amount (1.70%) of transparent minerals. These are apatite, zircon/monazite, rutile, tourmaline and a few titanite, epidote-zoisite/clinozoisite, pyroxene and garnet.

The descriptive system of heavy minerals

Previous investigations dealing with heavy mineral analysis, mainly identify the mineral species, their relative abundance and detect the characteristic mineral associations. Based on them, different types of potential raw materials can be determined/classified, and the provenance of the examined materials can also be identified.

Through the detailed description of the heavy minerals, not only the mineral species can be separated, but different subtypes for each type of mineral can also be defined. These variants may

have different origins and they can arrive from various sources/regions. They are suitable for the characterization and differentiation of the potential raw materials. Therefore, a new methodology was developed for the heavy mineral variants identified in our red sandstone samples:

The descriptive system was created for each mineral species that occurs in higher amounts in the examined sandstones, being either an archaeological or a geological sample. During the previous HMA examination, five main mineral phases were selected: rutile, zircon, tourmaline, apatite and titanite (see above in '*Heavy mineral content of the examined red sandstones*'). Different features were identified in each mineral group, and with their help, different variants could be determined (see below). Each raw material (both for archaeological tools and geological samples) is characterized not only quantitatively, but also qualitatively, so that the gained data sets are also suitable for statistical analysis. The heavy mineral variants of the archaeological and the potential raw materials will also be comparable on a quantitative basis.

Several characteristics formed the basis of the classification for each mineral species, such as colour and/or pleochroism, roundness, the mineral/crystal form and habit, shape, appearance of weathering, alteration, and zoning, etc.

In the case of rutile, the primary classification was based on the mineral's colour and roundness parameters: rutile1 are yellowish-brown coloured, well-, or medium-rounded and rutile2 are also yellowish-brown coloured, but medium-, or poorly rounded types. Rutile3 are reddish brown and rutile4 is dark brown (maybe reddish-brown). The differentiation of the subtypes was aided by the observation of mineral form and shape (**Fig. 1.a-b**).

Zircon: the initial classification was based on the roundness variations: zircon1 is well-, zircon2 is medium- and zircon3 is poorly rounded. Subtypes were defined on the base of the mineral form and shape (**Fig. 2.a-b**).

In the case of tourmaline, the colour and the pleochroism were the defining parameters and the basis for differentiation. Seven different variants were identified within this mineral type (**Fig. 3.a-b**).

Apatite and titanite are less common phases in most of the sandstone samples. The main factor of their classification was roundness and weathering/transformation. Three variants were distinguished for apatite and two for titanite grains (**Fig. 4.a-b** and **Fig. 5.a-b**).

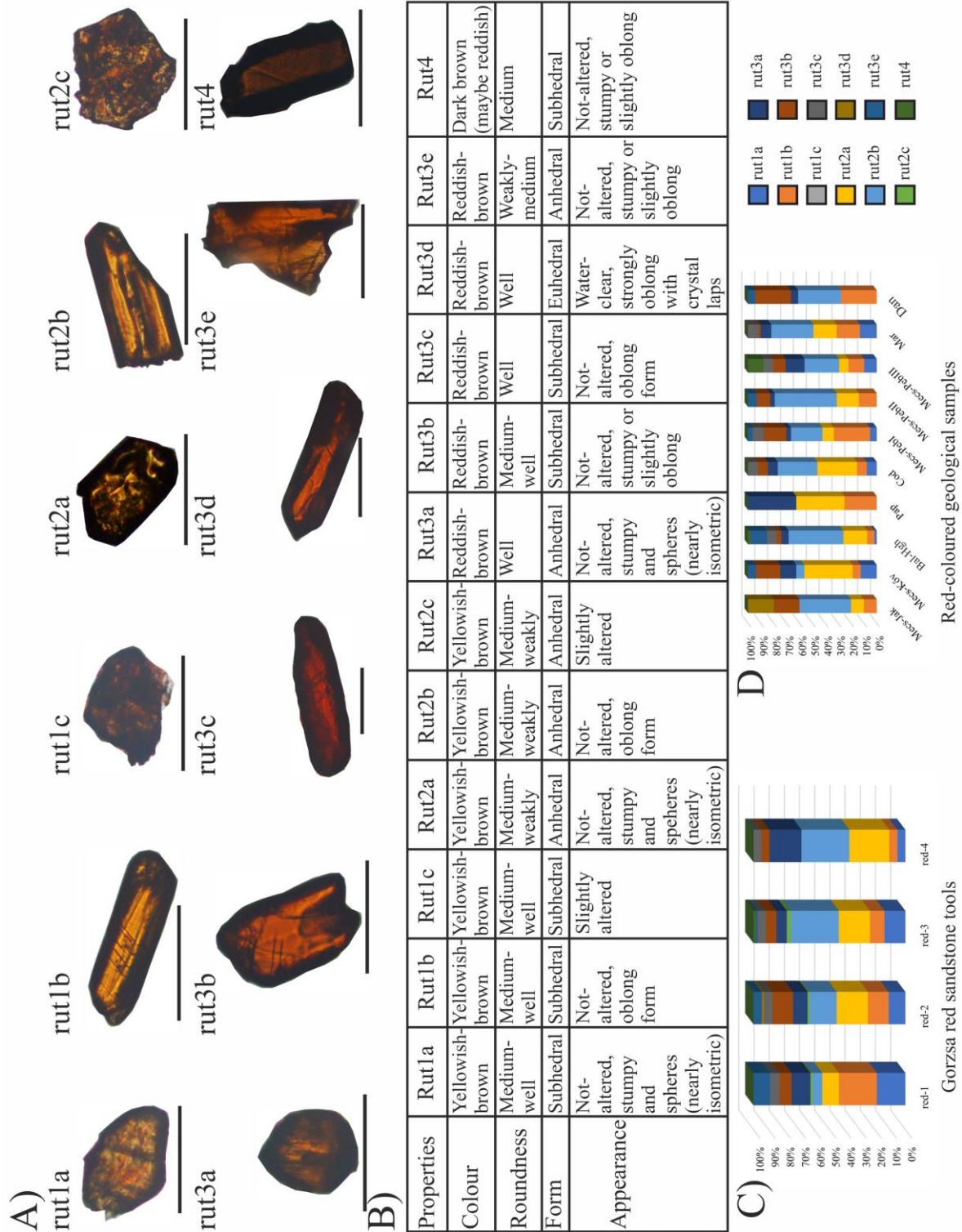


Fig. 1.: Rutile variants. (a) Scales are 100 µm. Rut1a = IHCs ('Mecs-Pebl'); rut1b = GOR-531 ('Red-3' tool type); rut1c = GOR-271 ('Red-2' tool type); rut2a = M-1/14 ('Mar'); rut2b = GOR-762 ('Red-3' tool type); rut2c = HH98/5 ('Mecs-Pebl'); rut3a = PF/1 ('Mecs-PeblII'); rut3b = PF/2 ('Mecs-PeblIII'); rut3c = GOR-90 ('Red-3' tool type); rut3d = GOR-133 ('Red-1' tool type); rut3e = Ja-KFhCseT ('Mecs-Jak'); rut4 = P-3 ('Bal-Hgh'). (b) System showing the rutile variants. (c-d) Column diagrams on quantitative distribution of the rutile variants relative to each other, in the case of Gorzsa and the potential geological samples.

1. ábra: Rutil változatok. (a) A skála minden esetben 100 µm. Rut1a = IHCs ('Mecs-Pebl'); rut1b = GOR-531 ('Red-3' eszköz típus); rut1c = GOR-271 ('Red-2' eszköz típus); rut2a = M-1/14 ('Mar'); rut2b = GOR-762 ('Red-3' eszköz típus); rut2c = HH98/5 ('Mecs-Pebl'); rut3a = PF/1 ('Mecs-PeblII'); rut3b = PF/2 ('Mecs-PeblIII'); rut3c = GOR-90 ('Red-3' eszköz típus); rut3d = GOR-133 ('Red-1' eszköz típus); rut3e = Ja-KFhCseT ('Mecs-Jak'); rut4 = P-3 ('Bal-Hgh'). (b) A rutil változatokat bemutató rendszer. (c-d) A rutil változatok egymáshoz viszonyított mennyiségi eloszlását bemutató oszlop diagramok.

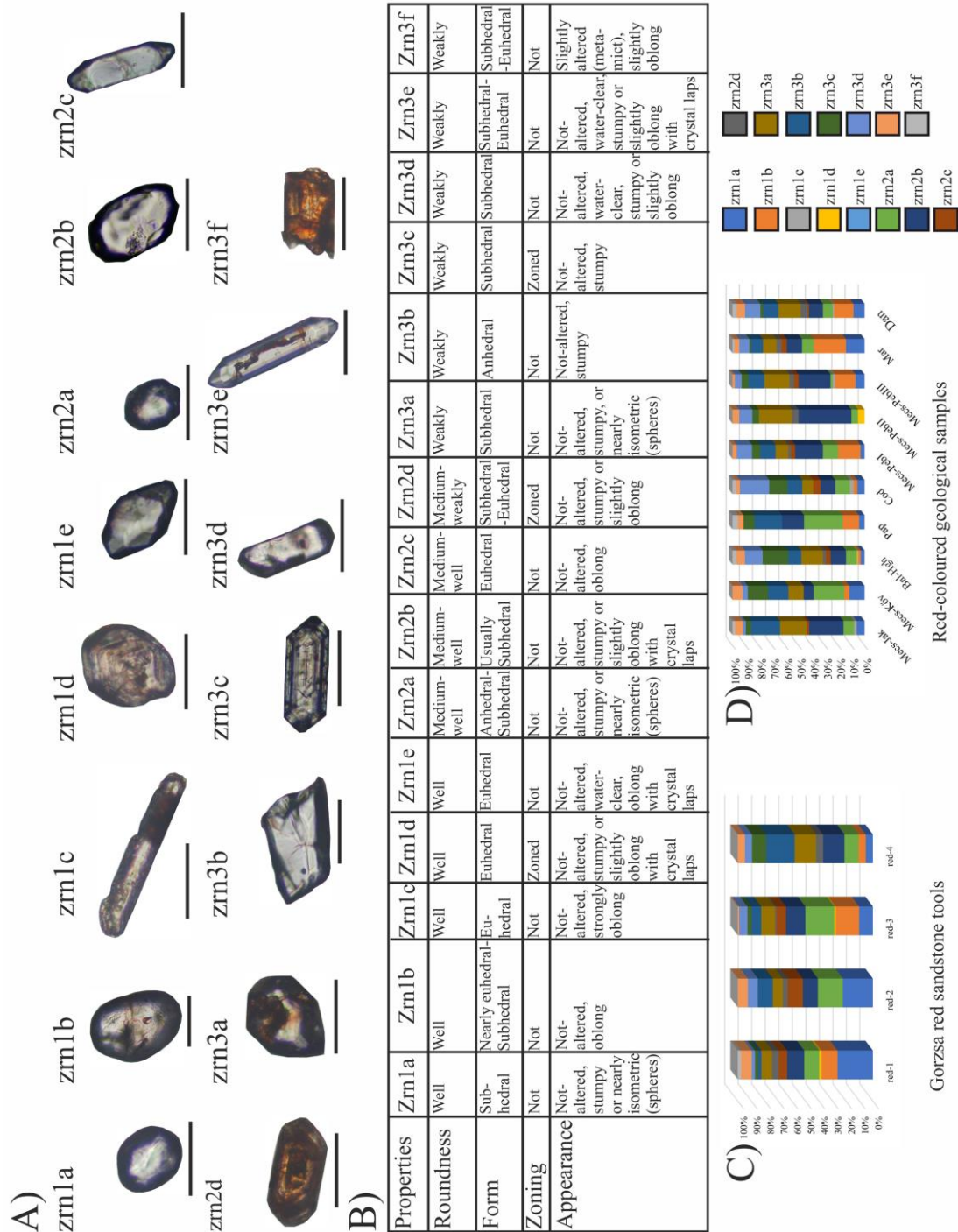


Fig. 2.: Zircon variants. (a) Scales are 100 µm. Zm1a = GOR-592 ('Red-1' tool type); zm1b = GOR-531 ('Red-3' tool type); zm1c = HCs/40 ('Mecs-PebIII'); zm1d = IIHCs ('Mecs-PebI'); zm1e = DVVH/1 ('Dan'); zm2a = P-1 ('Bal-Hgh'); zm2b = Codru-01 ('Cod'); zm2c = DVVH/1 ('Dan'); zm2d = GOR-90 ('Red-3' tool type); zm3a = GOR-133 ('Red-1' tool type); zm3b = Káfü-1 ('Bal-Hgh'); zm3c = P-3 ('Bal-Hgh'); zm3d = GOR-531 ('Red-3' tool type); zm3e = IHCs ('Mecs-PebI'); zm3f = GOR-374 ('Red-1' tool type). (b) System showing the zircon variants. (c-d) Column diagrams on quantitative distribution of the zircon variants relative to each other, in the case of Gorzsa and the potential geological samples.

2. ábra: Cirkon változatok. (a) A skála minden esetben 100 µm. Zm1a = GOR-592 ('Red-1' eszköz típus); zm1b = GOR-531 ('Red-3' eszköz típus); zm1c = HCs/40 ('Mecs-PebIII'); zm1d = IIHCs ('Mecs-PebI'); zm1e = DVVH/1 ('Dan'); zm2a = P-1 ('Bal-Hgh'); zm2b = Codru-01 ('Cod'); zm2c = DVVH/1 ('Dan'); zm2d = GOR-90 ('Red-3' eszköz típus); zm3a = GOR-133 ('Red-1' eszköz típus); zm3b = Káfü-1 ('Bal-Hgh'); zm3c = P-3 ('Bal-Hgh'); zm3d = GOR-531 ('Red-3' eszköz típus); zm3e = IHCs ('Mecs-PebI'); zm3f = GOR-374 ('Red-1' eszköz típus). B) A cirkon változatokat bemutató rendszer. C-D) A cirkon változatok egymáshoz viszonyított mennyiségi eloszlását bemutató oszlop diagramok.

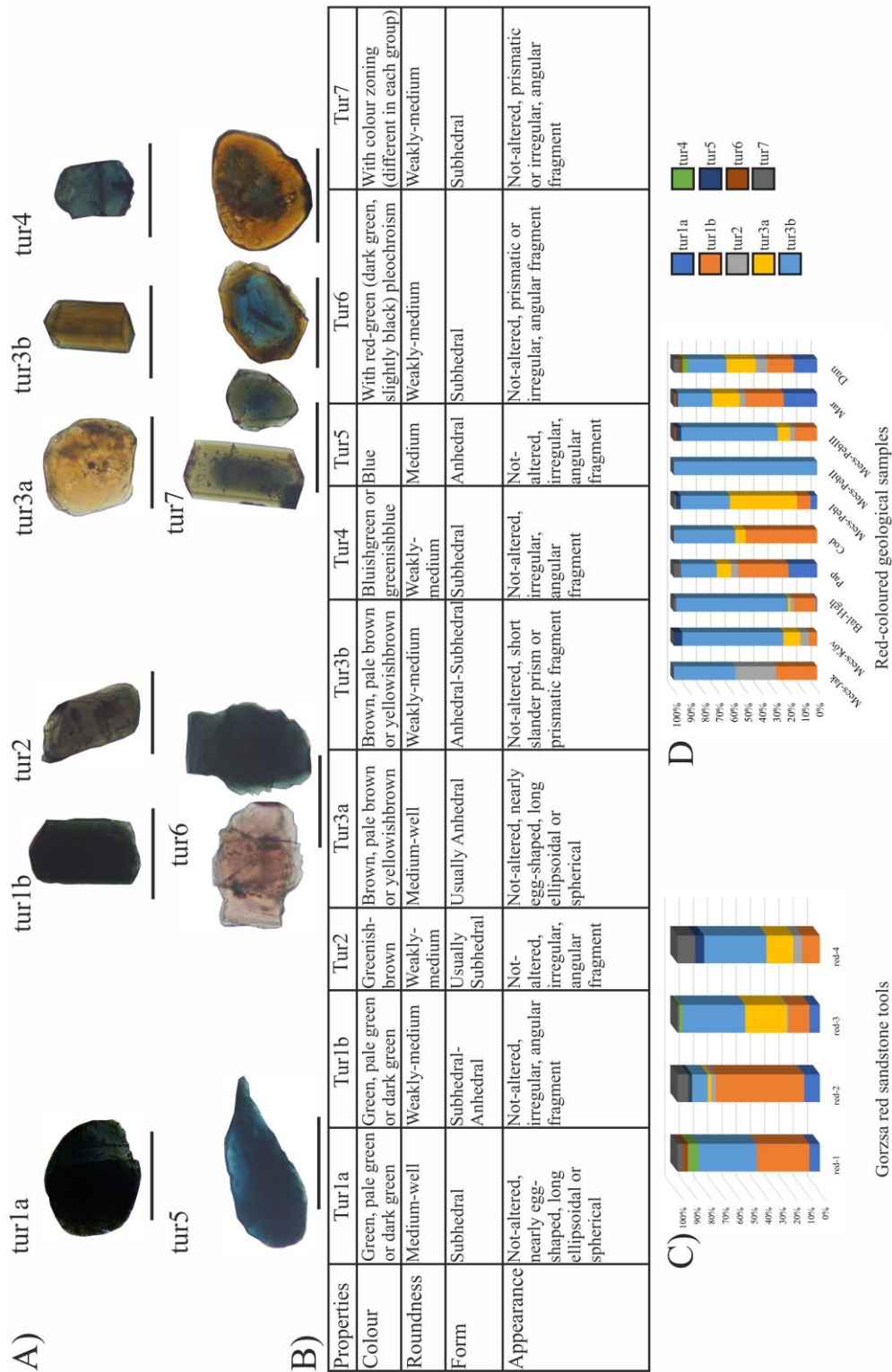


Fig. 3.: Tourmaline variants. (a) Scales are 100 µm. Tur1a = M-1/14 ('Mar'); tur1b = HH98/4 ('Mecs-PebIII'); tur2 = PF/2 ('Mecs-PebIII'); tur3a = PF/3 ('Mecs-PebIII'); tur3b = Alö-5 ('Bal-Hgh'); tur4 = Bare-1 ('Bal-Hgh'); tur5 = IHCs ('Mecs-Pebi'); tur6 = Ba-KFhBaT ('Mecs-Köv'); tur7 = HH98/5 ('Mecs-Pebi), GOR-112 ('Red-2' tool type), GOR-349 ('Red-4' tool type) and GOR-531 ('Red-3' tool type). (b) System showing the tourmaline variants. (c-d) Column diagrams on quantitative distribution of the tourmaline variants relative to each other, in the case of Gorzsa and the potential geological samples.

3. ábra: Turmalin változatok. (a) A skála minden esetben 100 µm. Tur1a = M-1/14 ('Mar'); tur1b = HH98/4 ('Mecs-PebIII'); tur2 = PF/2 ('Mecs-PebIII'); tur3a = PF/3 ('Mecs-PebIII'); tur3b = Alö-5 ('Bal-Hgh'); tur4 = Bare-1 ('Bal-Hgh'); tur5 = IHCs ('Mecs-Pebi'); tur6 = Ba-KFhBaT ('Mecs-Köv'); tur7 = HH98/5 ('Mecs-Pebi), GOR-112 ('Red-2' eszköz típus), GOR-349 ('Red-4' eszköz típus) and GOR-531 ('Red-3' eszköz típus). (b) A turmalin változatokat bemutató rendszer. (c-d) A turmalin változatok egymáshoz viszonyított mennyiségi eloszlását bemutató oszlop diagramok.

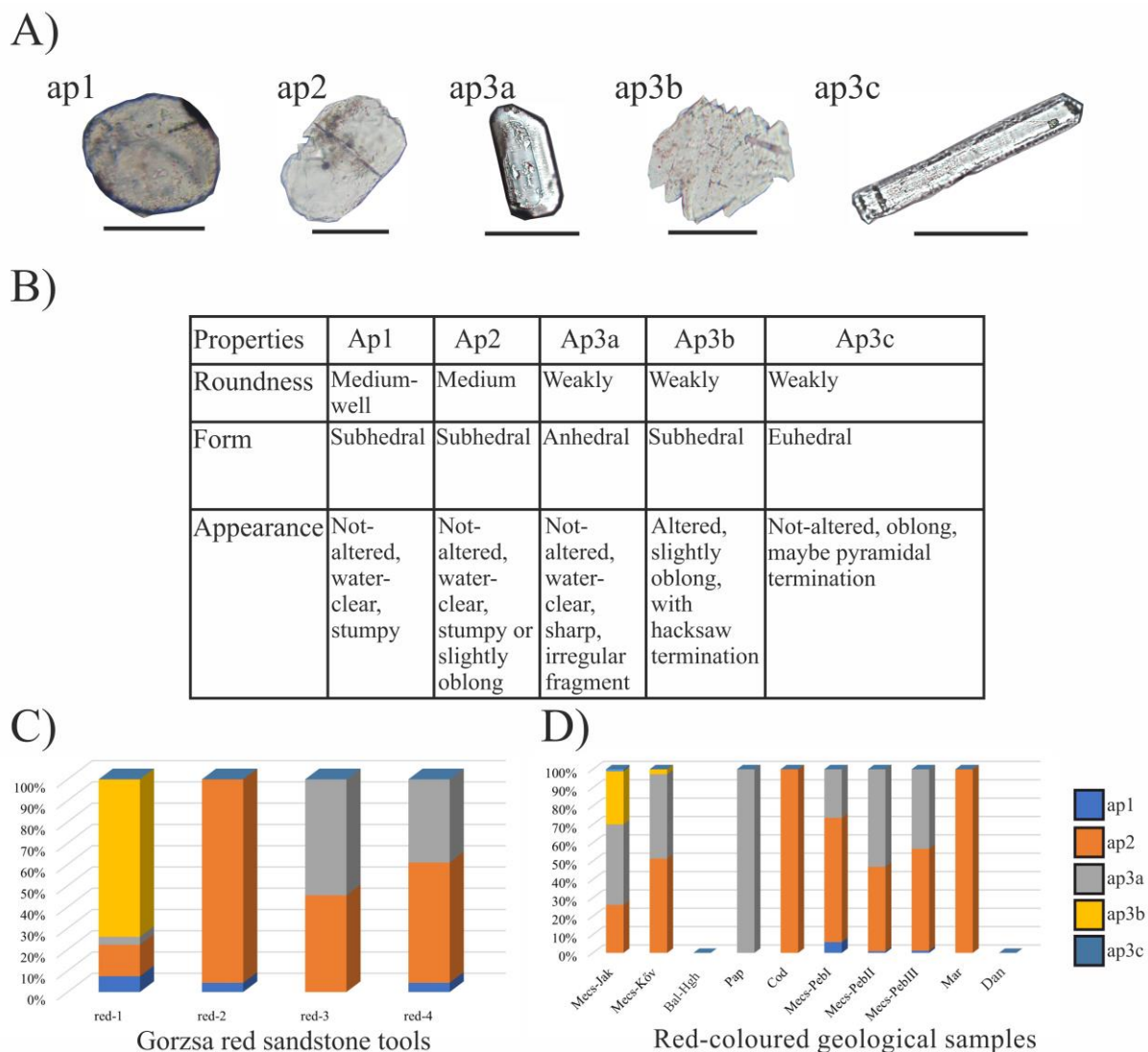


Fig. 4.: Apatite variants. (a) Scales are 100 µm. Ap1 = Ja-JFh_JS ('Mecs-Jak'); ap2 = GOR-592 ('Red-1' tool type); ap3a = IHCs ('Mecs-PebI'); ap3b = GOR-592 ('Red-1' tool type); ap3c = Ja-JFh_JS ('Mecs-Jak'). (b) Table showing the apatite variants. (c-d) Column diagrams on quantitative distribution of the apatite variants relative to each other, in the case of Gorzsa and the potential geological samples.

4. ábra: Apatit változatok. (a) A skála minden esetben 100 µm. Ap1 = Ja-JFh_JS ('Mecs-Jak'); ap2 = GOR-592 ('Red-1' eszköz típus); ap3a = IHCs ('Mecs-PebI'); ap3b = GOR-592 ('Red-1' eszköz típus); ap3c = Ja-JFh_JS ('Mecs-Jak'). (b) Az apatit változatokat bemutató táblázat. (c-d) Az apatit változatok egymáshoz viszonyított mennyiségi eloszlását bemutató oszlop diagramok.

Abundance of heavy mineral variants in the examined red sandstone samples

Ground stone tools from Gorzsa

In the case of the ground stone tools from Gorzsa, yellowish brown coloured, well-, or medium rounded rutile is the most common heavy mineral type. Different variants may be distinguished in each red sandstone tool type (**Appendix – Table 1.**). Three of four types of tool samples contain well-, or medium-rounded zircon grains. The type

'Red-4', contains the highest amount of poorly rounded, almost euhedral crystals (**Appendix – Table 2.**).

Tourmalines have brown colour in most of the samples, but green and other types also appear (**Appendix – Table 3.**).

Apatite grains are mainly medium-, or poorly rounded, but in type 'Red-1' there is a special variant with 'hacksaw' terminations (**Appendix – Table 4.**).

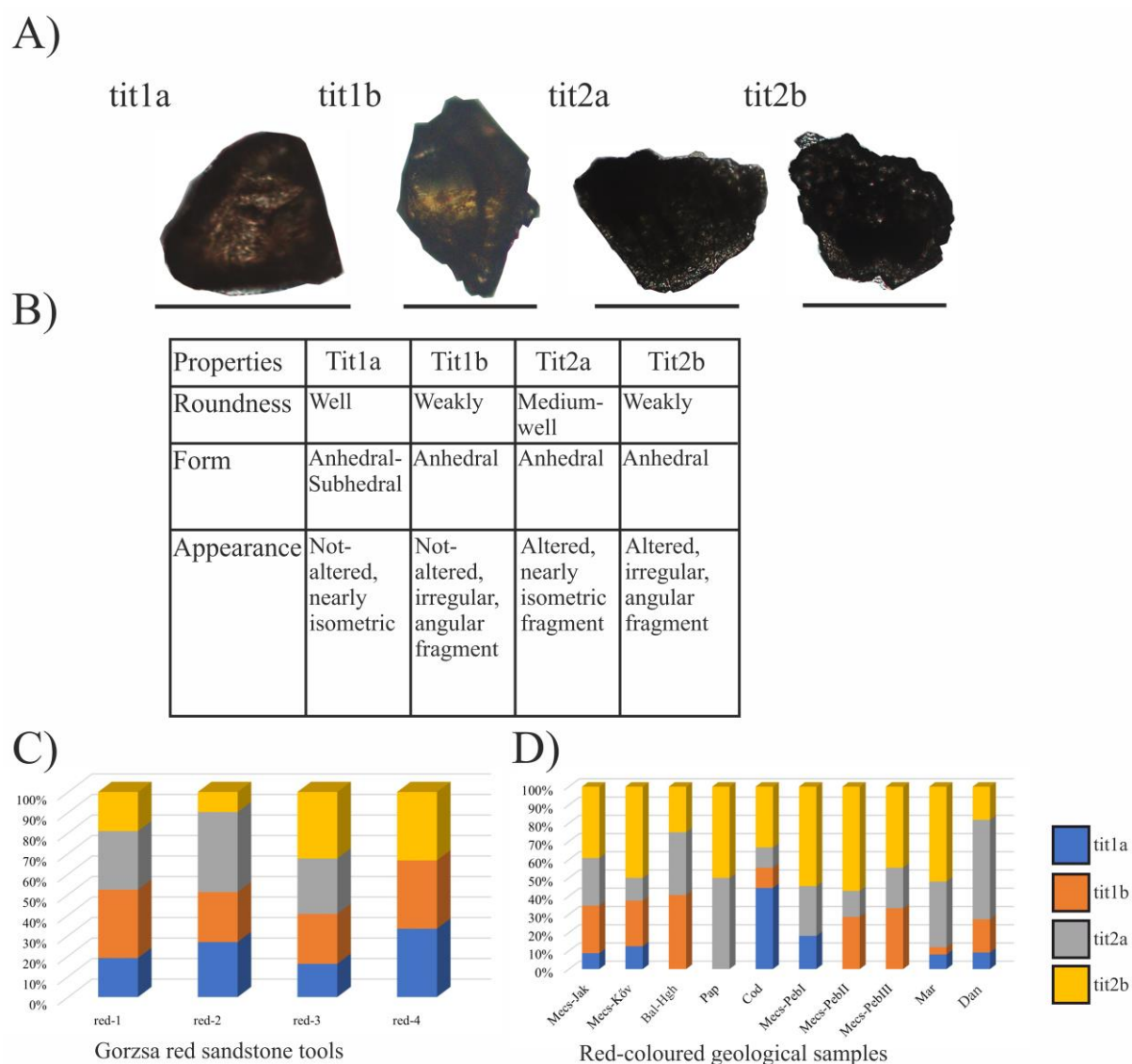


Fig. 5.: Titanite variants. (a) Scales are 100 µm. Tit1a = Codru-01 ('Cod'); tit1b = Káfű-1 ('Bal-Hgh'); tit2a = M-1/19 ('Mar'); tit2b = Ja-JFh_JS ('Mecs-Jak'). (b) Table showing the titanite variants. (c-d) Column diagrams on quantitative distribution of the titanite variants relative to each other, in the case of Gorzsa and the potential geological samples.

5. ábra: Titanit változatok. (a) A skála minden esetben 100 µm. Tit1a = Codru-01 ('Cod'); tit1b = Káfű-1 ('Bal-Hgh'); tit2a = M-1/19 ('Mar'); tit2b = Ja-JFh_JS ('Mecs-Jak'). (b) A titanit változatokat bemutató táblázat. (c-d) A titanit változatok egymáshoz viszonyított mennyiségi eloszlását bemutató oszlop diagramok.

Titanite grains occur in altered and fresh (not altered) forms. 'Red-1' and 'Red-2' contain them in the same amount, but 'Red-3' and 'Red-4' are different from them (**Appendix – Table 5**).

Potential raw materials

Rutile

In the samples from the Permian-Triassic sequence in the Mecsek Mountains (Jakabhegy Sandstone = 'Mecs-Jak' and Kővágószőlős Sandstone = 'Mecs-Köv'), yellowish-brown coloured, medium-, or poorly rounded rutile appear with the highest

amount. They make up more than 50% of the rutile grains. Reddish-brown versions also appear in smaller quantities. Pebbles from the Miocene sequence from the Mecsek Mts. contain higher amounts (more than 60%) of yellowish-brown coloured rutile, than the Permian-Triassic sandstones. Reddish brown coloured ones usually occurred in smaller amounts (less than 30%) (**Appendix – Table 6**).

Balaton Highlands ('Bal-Hgh') samples contain yellowish-brown coloured, medium-, or poorly rounded rutile and a few reddish-brown variants. Maros River ('Mar') samples have a lot of

yellowish brown, medium-, or poorly rounded rutile and a few reddish-brown coloured grains. Pebbles from the Danube River ('Dan') exhibit a lot of well- and medium rounded, yellowish-brown rutile and a small amount of reddish-brown coloured ones are also present. In the Papuk samples, yellowish-, and reddish-brown coloured grains appear in high quantities. In the Codru ('Cod') samples, there are lots of medium-, or poorly rounded, yellowish-brown coloured grains (more than 70%) and there is also a small amount of reddish-brown rutile (**Appendix – Table 6.**).

Zircon

Samples of the 'Mecs–Jak' and 'Mecs–Köv' contain poorly rounded, nearly isometric (stumpy) or isometric zircon grains. Pebbles from the Miocene sequence of the Mecsek Mts. ('Mecs–PebI – III') contain a lot of medium and poorly rounded grains that are stumpy, or nearly isometric. Among them there are zircons, which have oblong forms and some of them show zonation (**Appendix – Table 7.**).

Zircon grains from the 'Bal–Hgh' samples are poorly rounded crystals; they can be isometric or have oblong form. They are present in similar amount. Grains with zoning are also a common type. The 'Mar' samples contain a lot of well and poorly rounded types. These are stumpy and isometric in most of the cases. Pebbles from the Danube River contain a lot of poorly- and well-, or medium rounded grains. These can be isometric or stumpy and there are crystals with zoning as well. In the Papuk samples, there are medium-, or poorly rounded grains, with nearly isometric, stumpy forms and a few grains with zoning also appear. In the Codru samples, there are lots of poorly-, or medium rounded, oblong or nearly isometric grains and a few crystals with zoning are also present (**Appendix – Table 7.**).

Tourmaline

Samples of the 'Mecs–Jak' and 'Mecs–Köv' contain brown, greenish-brown and some green-coloured tourmalines. Tourmaline grains that come from the pebbles of the Miocene sequence of the Mecsek Mts. are brown and green coloured ones (**Appendix – Table 8.**).

Tourmaline grains from the 'Bal–Hgh' samples are mainly brown- and sometimes green-coloured ones. The 'Mar' samples contain brown and green ones with a similar amount. Pebbles from the Danube River contain brown- and green-coloured tourmalines (similar amounts). In the Papuk samples, there are green- and brown-coloured tourmalines, where the green ones are more common types. In the Codru samples, there are green and brown grains, with similar distributions (**Appendix – Table 8.**).

Apatite

Samples of the 'Mecs–Jak' contain poorly-rounded apatite grains that in some of the cases have 'hacksaw' terminations. There are also a few strongly oblong ones. The 'Mecs–Köv' samples contain medium- and poorly-rounded grains in similar amounts. Apatite grains come from the pebbles of the Miocene sequence of the Mecsek Mts. They are medium or poorly-rounded (**Appendix – Table 9.**).

From the Balaton Highlands and the Danube River samples, apatite is missing. In the 'Mar' and 'Cod' samples, apatite grains are medium-rounded, meanwhile in the 'Pap' samples they are poorly-rounded (**Appendix – Table 9.**).

Titanite

In the 'Mecs–Jak' and 'Mecs–Köv' samples, titanite grains occur dominantly in weathered forms. Those coming from the pebbles of Miocene sequences of the Mecsek Mts. occur in both altered and fresh forms (**Appendix – Table 10.**).

In the Balaton Highlands samples, altered and fresh variants occur with an equal amount. Titanite grains of the Danube River and Papuk samples are mostly weathered. In the Codru sandstones, titanite grains occur in both forms with the same amounts (**Appendix – Table 10.**).

Discussion

Heavy mineral composition of the red-coloured ground stone tools

Ground stone tool types, 'Red–1', 'Red–2' and 'Red–3' have similar heavy mineral composition, but the relative abundance of each HM phase is different. Type 'Red–4' has a special HM composition, with lots of garnet and a smaller amount of apatite, epidote, zircon, rutile and tourmaline.

Heavy mineral variants distributions per mineral species

Rutile

Yellowish-brown coloured, well-, or medium-rounded ones are the most common types, in the archaeological samples from Gorzsa tell. The different rutile variants show a slightly different distribution. The most common phases are rutile1b, 2b, 2a and 1a (**Appendix – Table 1.; Fig. 1.c.**).

Types 'Red–1' and 'Red–3' from Gorzsa show similar composition to 'Mecs–PebI', Codru and Maros. Type 'Red–2' shows the most similar composition to 'Mecs–Köv', 'Bal–Hgh', Codru and Maros. 'Red–4' shows similarities to 'Mar', 'Mecs–PebIII', 'Bal–Hgh' and 'Cod'. There are geological samples, which can certainly be excluded from the

possible source rocks, such as Papuk, Jakabhegy Sandstone and 'Mecs-PebII'. On the other hand, samples from the Balaton Highlands and Mecs-PebI cannot be excluded with absolute certainty (**Fig. 1.d**).

Zircon

Type 'Red-1' from Gorzsa mainly includes well-, or medium-, types 'Red-2' and 'Red-3' medium-, or well-rounded zircon grains. Type 'Red-4' from Gorzsa contains the highest amount of poorly rounded, slightly euhedral crystals. Most of the grains have isometric shape/appearance, but the 'Red-4' ones are more oblong and among them crystals with zoning are more common than in the other types (**Appendix – Table 2.; Fig. 2.c**).

Type 'Red-1' and 'Red-3' from Gorzsa show similar composition as than samples from the Maros River and 'Mecs-PebI'. Tool type 'Red-2' is not similar to any of the geological samples. Type 'Red-4' from Gorzsa is similar to 'Bal-Hgh' and 'Cod'. There are geological samples, which ones can certainly be excluded from the possible source rocks, such as Papuk, Jakabhegy and Kővágószőlős Sandstones, and pebbles from the Miocene sequence from the Mecsek Mountains ('Mecs-PebII and III'). Samples from the group 'Mecs-PebI' cannot be excluded from the possible raw materials with absolute certainty (**Fig. 2.d**).

Tourmaline

Type 'Red-1' from Gorzsa contains brown- and green-coloured tourmalines in similar amounts. Tool type 'Red-3' contains the highest number of brown-variants. Tourmalines of the 'Red-4' tool type are mostly brown but show high variation. 'Red-2' exhibits the highest number of green-coloured tourmalines. Type 'Tur-7' grains show colour zoning; they are common phases in Gorzsa type 'Red-4' and 'Red-2' (**Appendix – Table 3.; Fig. 3.c**).

Gorzsa types 'Red-1' and 'Red-2' are not similar to any of the examined geological samples. Type 'Red-3' from Gorzsa shows similar composition to the samples from the Maros and Danube rivers, and also to the Papuk Mountains. Tool type 'Red-4' is similar to 'Mecs-Köv' and 'Mecs-PebIII'. There are geological samples, which can certainly be excluded from the possible source rocks, such as Jakabhegy Sandstone, Codru, and 'Mecs-PebII'. Samples from the 'Mecs-PebI and III' groups, Kővágószőlős Sandstone and Balaton Highlands cannot be excluded with absolute certainty from the possible raw materials (**Fig. 3.d**).

Apatite

Apatite grains in type 'Red-1' from Gorzsa are poorly rounded crystals with 'hacksaw' terminations. In tool type 'Red-2' they are medium-rounded, while in 'Red-3' type from Gorzsa they are medium-, or poorly-rounded with similar frequency. In 'Red-4' tool type there are medium, and some poorly rounded grains (**Appendix – Table 4.; Fig. 4.c**).

'Red-1' and 'Red-2' types from Gorzsa are not similar to any of the geological samples. Type 'Red-3' from Gorzsa show similar composition to pebbles from the Miocene sequence of the Mecsek Mountains ('Mecs-PebII and III') and Kővágószőlős Sandstone. 'Red-4' tool type is similar to 'Mecs-PebI'. There are geological samples, which can certainly be excluded from the possible source rocks, such as Jakabhegy Sandstone, Codru, Papuk and Maros River. Samples from the 'Mecs-PebI' group cannot be excluded with absolute certainty from the possible raw materials (**Fig. 4.d**).

Titanite

Titanite grains in 'Red-1' and 'Red-2' types from Gorzsa have similar appearance. In 'Red-3' tool type grains are weathered in most of the cases. In type 'Red-4' from Gorzsa, not-altered grains are the most dominant variants (**Appendix – Table 5.; Fig. 5.c**).

'Red-1' and 'Red-4' are not similar to any of the examined geological samples. 'Red-2' is similar to 'Dan'. Type 'Red-3' from Gorzsa shows similar composition to Jakabhegy Sandstone. There are geological samples, which can certainly be excluded from the possible source rocks, such as Papuk, Kővágószőlős Sandstone, Balaton Highlands, Codru, and Maros River (**Fig. 5.d**).

Heavy mineral variants distributions per ground stone tool types

Type 'Red-1'

Based on the HM observations and counting, rutile and zircon grains from the 'Red-1' type were useful for the identification of its provenance. Rutile grains of 'Red-1' tools show similarities with those of 'Red-I' pebbles from the Mecsek ('Mecs-PebI'), Maros River pebbles ('Mar') and sandstone from the Codru-Moma Mts. ('Cod'). Zircon variants match with Maros and 'Mecs-PebI' grains. Tourmaline, apatite and titanite turned out to be less useful in the provenance analysis (**Figs. 1–5.c-d**).

Type 'Red-2'

Only two minerals showed similarities with some of the potential sources, these were rutile and titanite. The other three phases – zircon, tourmaline and apatite – were less useful in our research. Rutile

grains were similar to grains from Codru, Maros, Balaton Highlands ('Bal-Hgh') and Kővágószőlős Sandstone ('Mecs-Köv'). Titanite grains were similar to Danube samples ('Dan') (Figs. 1–5.c-d).

Type 'Red-3'

In the case of type 'Red-3', all five mineral phases proved to be suitable for the source identification. Rutile grains show similarities with 'Mecs-PebI', 'Cod' and 'Mar'; zircon variants with 'Mecs-PebI' and 'Mar'; tourmalines with 'Mar', 'Dan' and 'Pap'; apatites with 'Mecs-PebII and III' and 'Mecs-Köv' and titanites with 'Mecs-Jak' samples (Figs. 1–5.c-d).

Type 'Red-4'

Almost all types of mineral species are suitable for provenance analysis, except for titanite. Rutile grains are similar to 'Mecs-PebIII', 'Cod', 'Mar' and 'Bal-Hgh'; zircon to 'Bal-Hgh' and 'Cod'; tourmaline to 'Mecs-Köv' and 'Mecs-PebIII' and apatite to 'Mecs-PebI' (Figs. 1–5.c-d).

Conclusions

Based on the HM observations, ground stone tool types 'Red-1' – 'Red-4' could be distinguished. 'Red-1', 'Red-2' and 'Red-3' show similarities in their heavy mineral composition, their main components are zircon, tourmaline, rutile, titanite and maybe apatite. But the relative abundance of each heavy mineral phase is different. Type 'Red-4' from Gorzsa has a special heavy mineral composition, with lots of garnet and a lower amount of apatite, epidote, zircon, rutile and tourmaline. For the most common mineral phases, rutile, zircon, tourmaline, apatite and titanite a descriptive system was developed, where different variants were identified for each mineral species.

Rutile, zircon and tourmaline grains were the most suitable mineral phases, apatite and titanite turned out to be less indicative as provenance markers. The main results are summarized across the ground stone tool types:

-Type 'Red-1' from Gorzsa shows similar heavy mineral composition (zircon and rutile) to pebbles of the Maros River and of the Mecsek Mts. (Type-I, 'Mecs-PebI').

- Type 'Red-2' has a very uncertain raw material source; pebbles from the Maros River have the nearest/ most similar composition, this result was determined by the variants of rutile grains.

-Type 'Red-3' has a similar composition – referring to zircon, rutile and tourmaline grains – to pebbles, group 'Red-1', of the Mecsek Mountains ('Mecs-PebI') and pebbles of the Maros River ('Mar').

-Type 'Red-4' is similar to pebbles, group 'Red-III' from the Mecsek ('Mecs-PebIII'), Balaton Highlands ('Bal-Hgh') and possibly to samples from the Codru-Moma Mountains ('Cod').

There are raw materials, which were excluded from the potential sources, based on their heavy mineral and mineral variants compositions, such as Papuk ('Pap'), Jakabhegy Sandstone of the Mecsek ('Mecs-Jak') and pebble group 'Red-II' ('Mecs-PebII'). The latest result is in contrast with the statement of Piros (2010), who originated the Gorzsa type 'Red-1' from the Jakabhegy Sandstone from the Permotriassic sequence of the Mecsek Mountains and tool type 'Red-2' from the Papuk and pebbles from the Mecsek.

Based on the previous petrographic (see in Miklós et al. 2021) and current heavy mineral examinations, ground stone tools can be originated from the Maros River and the Apuseni Mountains (Transylvania). Samples from the Mecsek Miocene, ('Mecs-PebI and III' groups), Kővágószőlős Sandstone ('Mecs-Köv'), Balaton Highlands ('Bal-Hgh') and pebbles from the drainage of the Danube River ('Dan') cannot be excluded with absolute certainty from the possible raw materials. However, according to the previous results of the petrographic examination (Miklós et al. 2021), in the choice of raw materials the above mentioned, four or five sources might have been of less significance than the Transylvanian ones.

In the future, in order to clarify the results, additional possible sources of Transylvanian origin must be involved in our research.

Contribution of authors

Miklós Dóra Georgina Methodology, Writing – Original draft. **Józsa Sándor** Writing – Original draft.

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