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RESEARCH

Provenance analysis of red sandstone ground stone tools from the tell site of Hódmezővásárhely-Gorzsa (SE Hungary)

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Abstract

Hódmezővásárhely–Gorzsa is a multi-period tell settlement in South Hungary in the centre of the Great Hungarian Plain, about 15 km southwest of the city of Hódmezővásárhely. The thickest section of the settlement belongs to the Late Neolithic Tisza Culture period. In total, 1061 macrolithic artefacts were unearthed, a quarter of which was polished, and three quarter of which were ground stone tools. Half of the ground stones were made of different types of sandstone, including (1) red-, (2) grey micaceous-, (3) calcareous-, (4) white meta sandstones, and (5) other sandstones and metasandstones were identified. The red sandstones are further categorised into four subgroups based on optical microscopy. This examination is the first systematic multi-analytical investigation (i.e. optical microscopy, whole-rock geochemistry and mineral chemistry), carried out on these ground stone tool types. The goal is to identify and precisely locate the raw material types, in which heavy minerals and the tourmaline mineral chemistry play the key role. To determine the provenance of each of these subgroups, samples were collected from seven geological localities (i.e. primary outcrops and secondary presences, such as river drainages or terraces) for a comparative study. Based on our results, the alluvium of the Maros River can be considered as a possible source for the 'Red – 3' type of Gorzsa, while the results for the rest red sandstone types ('Red – 1', 'Red – 2' and 'Red – 4') are inconclusive in terms of provenance.

Keywords Neolithic sandstone tools · Tisza culture · Sandstone geochemistry · Heavy mineral · Tourmaline mineral chemistry · Provenance

Introduction

Ground stone tools (GSTs), also termed macro-lithic tools, are non-chipped and non-polished tools, which are used for grinding, pounding, abrading, pecking and polishing of vegetal, animal and mineral materials (Adams et al. 2009; de Beaune 2004; Adams 2014; Dubreuil and Savage 2014; Dubreuil et al. 2015), and are generally characterized by long functional histories (Dubreuil and Savage 2014; Dubreuil et al. 2015). Over the last decade, research on GSTs showed an exponential growth by the development of the instrumental techniques. A large variety of qualitative and quantitative methods, including use-wear analysis, mechanical tests, 3D modelling, surface morphometrics, spatial and residue analyses, as well as experimental frameworks have been actively implemented focusing on the archaeological perspective (Procopiou et al. 2002; Delgado-Raack et al. 2009; Caruana et al. 2014; Benito-Calvo et al. 2015, 2018; Hayes et al. 2017; Caricola et al. 2018; Hayes and Rots 2019;

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Zupancich et al. 2019; Arroyo and de la Torre 2020; Cristiani and Zupancich 2021). The archaeometric investigation, including petrography and geochemistry, of such artefacts to determine their raw materials have not been conducted routinely. Collection of ground stone tools of archaeological contexts only became part of the protocol in the last 20 – 30 years. Most of them are made from sedimentary rocks, such as sandstone. Throughout human history sandstones were frequently used for making ground stone tools (e.g. grinding stones, mill stones, abrasive tools, hammerstones) or moulds for casting metal artefacts, but they were also utilized as building stones (Wright 1992; Adams et al. 2009; Dubreuil and Savage 2014; Dubreuil et al. 2015; Caricola et al. 2018; Cristiani and Zupancich 2021).

Sandstones are siliciclastic sedimentary rocks composed of mineral grains and rock fragments, fine-grained particles (e.g. clay, silt) called matrix, and pores (Pettijohn et al. 1973). In addition, cement of calcareous or siliceous composition is present binding the various grains together. These characteristics usually result in quite massive, resistant, sturdy, and durable rocks (Pettijohn et al. 1973).

In the Carpathian-Pannonian Region, sandstones, especially red in colour are very common rock types, and they were widely used as raw materials in the past (Péterdi 2012, 2020). These rocks often exhibit similar main detrital components, including dominantly quartz, minor feldspar and micas, and occasionally different types of rock fragments (Pettijohn et al. 1973). Specific detrital accessory minerals (or heavy minerals, HMs) in the sandstones, may assist to determine their provenance (Morton 1985; Morton and Hallworth 1999). Their mineralogy and abundance may be characteristic for sandstone and thus indicative of the source area of the raw material implemented in tool production (Dickinson 2007; Mange and Bezeczky 2007; Józsa et al. 2016). Furthermore, the majority of the HMs are highly sensitive to transport and to the environmental processes and changes (e.g. burial diagenesis, mineral solution, cementation) during the whole sedimentary cycle which impact the sandstone raw materials. At the same time, the more resistant heavy minerals are suitable for indicating the source area.

Sandstones are perfect raw materials, because the fine-grained matrix, the diagenetic cement, and the presence and size distribution of the pores also influence the physical and chemical resistance of the sandstone (Pettijohn et al. 1973). Based on these aspects, different sandstone types can be determined and distinguished (Pettijohn et al. 1973; Csernussi 1984; Péterdi 2012, 2020). The formation of the sandstones and the secondary sedimentary processes affecting them make the provenance study of the raw materials used for ground stone tool production problematic needing thorough approach and investigation (Thomas 1909; Lovell 1971; Arribas et al. 2003; Szakmány and Nagy 2005; Phillips 2007; Adams 2014; Chima et al. 2018; Baiyegunhi et al. 2020; Chen et al. 2020; Martínez-Sevilla et al. 2020; Petrounias et al. 2020; Péterdi 2020; Stergiou et al. 2021; Critelli and Criniti 2022). In the Carpathian-Pannonian Region there are not too many published detailed petrographic-geochemical analyses referring to sandstone formations (Csernussi 1984; Fazekas 1987, 1989; Wéber 1990; Varga et al. 2001; Varga 2009; Szőcs et al. 2015). Therefore, during archaeometric investigations both the archaeological tools and their potential source rocks need to be studied, as no available database currently exists.

Stone tools represent a very significant amount of finds in the archaeological assemblages, but in general most of them bear little aesthetic value; therefore, in many cases it is possible to conduct destructive analyses on them. Detailed petrographic and geochemical analysis, especially heavy minerals have never been employed before for sandstone macrolithic tools to assess the geological provenance of sandstones, with particular regard to the Carpathian-Pannonian region. The results are important to define the territorial network of cultural connections activated by the inhabitants for the procurement of stone raw materials necessary for their domestic activities.

Archaeological and archaeometric background

Following the small-scale surveys, systematic excavations were carried out between 1978 and 1996 at the tell (i.e. after Arabic for “settlement mounds”, Horváth 2009) site of Gorzsa (formerly marked as Keleti- and Czukor-farmstead) that eventually grew into a long-term research program (Horváth 1987, 2005). The settlement lies in the environs of Hódmezővásárhely city (N 46° 25′ 49″, E 20° 19′ 08″) at the confluence of the Tisza and Maros Rivers in the middle of the Great Hungarian Plain (Fig. 1). In the surrounding environment there are floodplain meadows, back swamps, drainage channels and natural loess-covered landforms, while the tell rises 3 to 4 m higher than its surroundings. The immediate to the tell area is covered by Holocene clayey silt and Pleistocene loessic sand. Local sediments near the tell settlement are composed of fine-grained sand, silt or loam. Bedrock outcrops are absent from the immediate area, therefore any types of raw materials for stone tool production had to be collected and transported to the site from various distances which exceed 60 km. The raw materials were gathered and transported completely untouched, as a rough-out, or even as a ready-made artefact (Starnini et al. 2015; Szakmány et al. 2019). The excavated area of the Gorzsa tell represents a nearly complete sequence of the Late Neolithic Tisza Culture with remnants of the later periods (i.e. Bronze and Iron Ages, Sarmatian) on the top. A total of 1061 macrolithic artefacts were collected. A quarter of

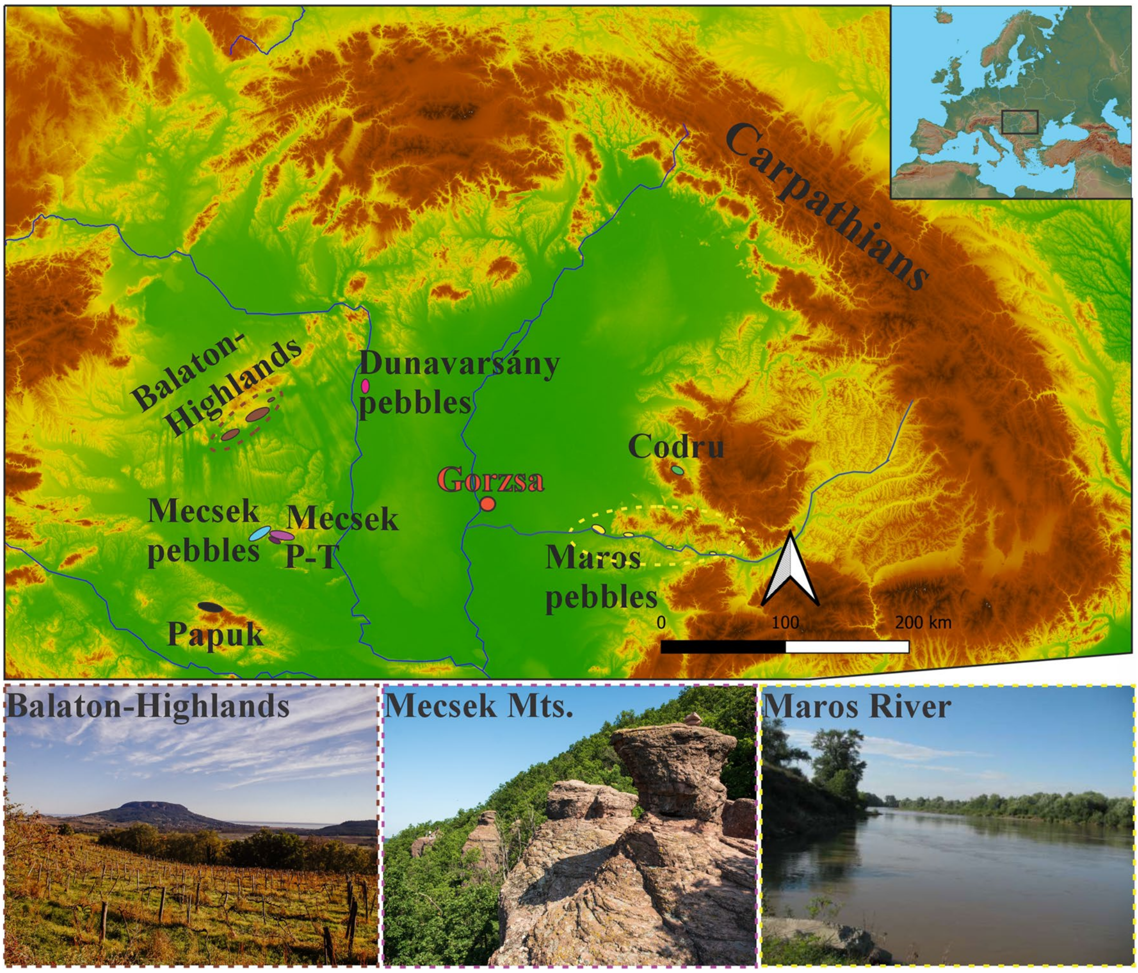

Fig. 1 Topographic map of the Carpathian Basin showing the tell site of Gorzsa and red sandstone occurrences investigated in this study. Mecsek P-T means the Jakabhegy Sandstone (with light purple ellipse) and Kővágószőlős Sandstone (with dark purple ellipse) formations. Mecsek pebbles occurs four terrains in the Mecsek Mountains, due to the scale of the map, it is only marked by a blue ellipse. Maros pebbles were collected from four different places, therefore similar to the Balaton Highlands samples, they were marked by ellipses (with yellow colour)

them represents polished stones (i.e. axes, adzes and chisels) and three quarters are GSTs (i.e. grinding stones, abraders, whetstones, hammerstones, pestles, hand stones etc.). Half of the GSTs are made of sandstone (Szakmány et al. 2008, Miklós et al. 2021).

The layer sequence containing the Late Neolithic Tisza Culture extends over 6 ha (Horváth 1982, 2003). The total area of the excavated and the connected registered site is 1000 m^2. The lifetime of the Neolithic village was divided into four settlement phases (D, C, B – A, phase D being the oldest one) based on the changes of the occupation patterns and the material findings (Horváth 2009). The thickness of the cultural layer is 2.6 to 3 m, with 1.8 to 2 m of it representing the Late Neolithic Tisza II-IV periods (Horváth 1987, 2003, 2005, 2014). The internal chronology of the Gorzsa settlement can be well defined based on a series of AMS radiocarbon dates between 4905 – 4810 and 4540 – 4440 cal BC. Several archaeological and archaeometric studies have been previously published, focusing on chipped stones, polished stones, ceramics (Vanicsek et al. 2013; Szakmány et al. 2019) and on ground stones (Biró 1998; Starnini et al. 2007, 2015; Szakmány et al. 2008, 2009, 2010). In contrary, only preliminary investigations were made on the archaeometric study of the ground stone tools made of sandstone (Szakmány et al. 2008, 2010; Piros 2010; Starnini et al. 2015; Miklós et al. 2021).

Szakmány et al. (2008, 2010) and Piros (2010) carried out macroscopic and polarizing microscopic analysis on thin sections and differentiated six sandstone types (Table 1). During a more recent re-examination of the sandstone tools of Gorzsa, five main types of sandstone could be distinguished, in which grey (32%), and red (27%) ones are the most common types (Table 1; Fig. 2a) (Miklós et al. 2021). Different subtypes were distinguished within the red sandstone tools

Table 1 Different classifications of sandstone implements of Gorzsa

New classification by Miklós et al. (this paper)		Szakmány et al. (2008)	Szakmány et al. (2010)	Piros (2010)
Grey	Grey-1	1) Dark grey, well or medium sorted orthosandstone with muscovite	1) Well sorted grey sandstone	3) Grey sandstone
	Grey-2	2) Polimict, weakly sorted, usually dark grey coloured orthosandstone	2) Poorly sorted grey sandstone	
Red	Red-1 (volcanic)	3) Red or lilac sandstone or siltstone, it can be layered with volcanic grains, sometimes foliated	3) Red-lilac sandstone	(1) Red-1 sandstone with volcanic grains (2) Red-2 sandstone
	Red-2 (fine-grained)			
	Red-3 (mature)			
	Red-4 (metamorphic)			
White meta	White meta	4) White-coloured, silicified metasandstone	4) White metasandstone	4) White metasandstone
Calcareous	Calcareous	5) Well sorted, grey or dark grey coloured sandstone, pebbly sandstone with well rounded grains and carbonate cement	5) Sandstone with sparite cement	5) Sandstone with sparite cement
Other sandstone, metasandstone	Other sandstone, metasandstones, siltstones, conglomerates and breccias	6) Wacke, grey coloured	6) Wacke	6) Other metasandstone

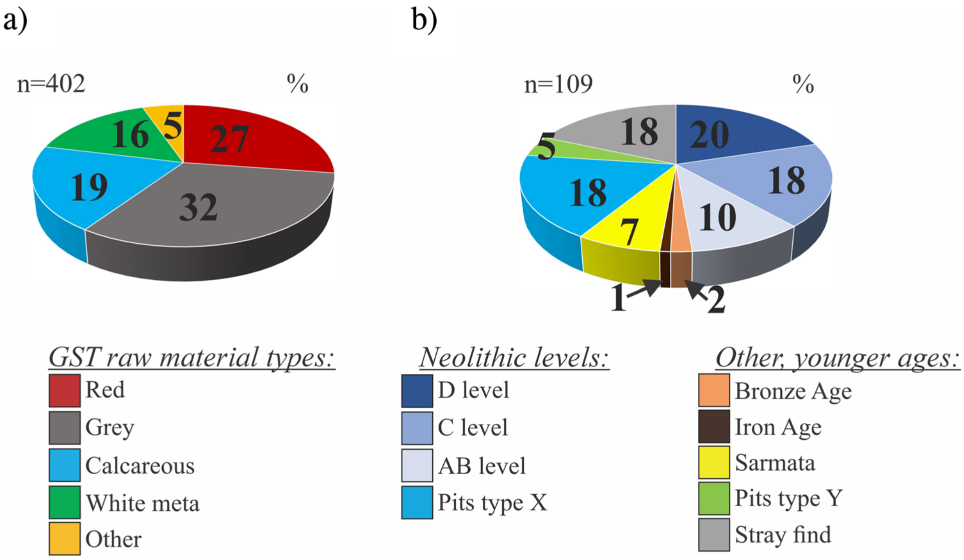

Fig. 2 (**a**) Pie chart showing the quantitative distribution of the sandstone types from Gorzsa site, (**b**) Distribution of ground stone tools from Gorzsa in relation to their archaeological context

(Table 1) by macroscopic (red-, or lilac sandstones with a homogenous composition by Szakmány et al. 2008, 2010) and/or microscopic petrographic examinations (separate two different variants (type – 1 and – 2 by Piros 2010). The potential raw material of the type – 1 was originated from the Permo-Triassic succession of the Mecsek Mountains (Jakabhegy Sandstone Formation; Fazekas 1987, 1989; Varga 2009; Piros 2010; Péterdi 2012, 2020; Miklós et al. 2021), whilst type – 2 may have originated from the Carpații Banatului, or the Gilău Mountains in Transylvania (Central Romania, Roth 1888, 1889; Pálfy 1897; Piros 2010). Other suggested possible sources for both red types, included the Miocene conglomerate sequence of the Mecsek Mountains, Permian sequence of the Papuk Mountain, and the Pleistocene pebble material of the Danube terraces (Szakmány et al. 2003; Józsa et al. 2009; Szakmány et al. 2009, 2010; Piros 2010; Miklós et al. 2021).

The aim of this work is to identify and describe the red sandstone types with petrography and geochemistry, and to make a comparative investigation of the archaeological samples of the tell and the red sandstone samples collected from potential sources. HMs have not been studied in case of macrolithic tools yet.

Regional geology

The Pannonian Basin located in Central Europe, surrounded by the Alps, Carpathians and Dinarides, is the largest intermontane basin in Europe (Royden et al. 1983; Nádor et al. 2003; Haas (ed.) 2013, Horváth et al. 2015; Fig. 1). The Pannonian Basin was composed of three megastructural facies units (terranes): the ALCAPA (ALps, CArpathians, and PAnnonian Basin, Csontos et al. 1992), the Tisza-Dacia Mega-units and the Mid-Hungarian Zone. Red sandstone formations occurred among the Palaeozoic – Mesozoic basement of the above-mentioned mega structures.

One of the most researched red sandstone occurrences in Hungary is situated in the Mecsek Mountains. A large-scale of fluvial, red-coloured siliciclastic Permian – Early-Triassic assemblage with variable composition can be detected on the surface. The above-mentioned succession has six different members (with different origin, composition, and lithology) that consist of conglomerate, sandstone, and siltstone materials in general (Barabás and Barabásné Stuhl 1998, Török 1998; Konrád and Barabásné Stuhl 2023, Konrád 2023). Another red sandstone occurrence, examined in similar detail, can be found in the Balaton Highlands (i.e. the Balatonfelvidék Formation, Fig. 1). The Formation begins with a polymictic conglomerate, pebbly sandstone that consisting of an alternation of red-coloured sandstone and siltstone, and in the upper part intraformational conglomerate layers settle, developed in fluvial and flood plains (Majoros 1983; Csernussi 1984).

Other red sandstone occurrences in the Carpathian basin, from longer distances to the archaeological site were included in our research, such as the Permo-Triassic siliciclastic succession of the Papuk Mountains (Croatia, Szakmány et al. 2003). The Permo-Triassic sediments of the Slavonian Mountains are divided into two units (the lower and the upper part; Jamičić 1989; Jamičić and Brkić 1987; Jamičić et al. 1987, 1989). The lower part consists of Paleozoic phyllite and plutonic pebbles with fluvial and lacustrine environments. The upper part shows a continuous transition towards the Lower Triassic sediments (Jamičić 1989). It is built by quartz sandstones and pebbly sandstones that is also characterized by a significant number of carbonates, which indicates the strengthening influence of the marine environment (transgression, Szakmány et al. 2003). Red, purplish-red Permian sediments can often be observed in the Codru nappe system within the area of the Apuseni Mountains (Transylvania, Romania, Fig. 1). Four different Carboniferous-Permian clastic and/or volcanic sedimentary formations were separated. (1) the Laminated Conglomerate Formation, which consists of oligomictic metaconglomerate and associated laminated metasandstone and purple metapelites, (2) the Vermicular Sandstone Formation, which consists of red biotrace sandstone and consists of interbedded shales, sandy shales, (3) the Rhyolitic Formation, which consists mainly of ignimbrites and interbedded tuffs and tuffaceous sandstones, and (4) the Feldspatic Formation, which consists of feldspatic sandstones (Vozárová 2009, Nicolae et al. 2014).

Among the examined secondary sources, the Maros River and its gravelly sediment, which can be observed east of Arad in recent times (Fig. 1). Detailed lithological examination has not been realized yet.

In the Mecsek Mountains, unconsolidated siliciclastic sequence (pebbles, sands and sandstones) of early-middle Miocene with fluvial origin comes to the surface in a large area, up to 100 m thick (Szászvár Formation; Jámbor and Szabó 1961; Hámor and Jámbor 1964; Hámor 1970; Ravaszné-Baranyai 1973, Chikán 1991; Barabás 2010; Józsa et al. 2009; Miklós 2018; Sebe 2023; Fig. 1). Approximately 6% of the pebble material consists of red siltstone and sandstone pebbles with varied composition and appearance (Miklós 2018). Their preliminary petrographic microscopic examination and classification were taken by Varga et al. (2002) and Tóth (2014). The origin of these red-coloured sandstone pebbles was not clarified until now.

During the Pleistocene, in the section above Dunaújváros, the Danube deposited its polymictic pebble sediments on the Pannonian sediments in a wide strip and in some places with a thickness exceeding 100 m (Pécsi 1959; Hahn 1975, Rónai 1985; Jaskó and Kordos 1990; Gábris and Nádor 2007). The assemblage, rich in coarse debris (consisting of pebble and pebbly sand, to a lesser extent sand, silt and clay) is exposed in many places in two main areas, the Kisalföld and the Southern Pest plain (Fig. 1). The pebble material is described as the Pestvidék Pebble Formation by Jaskó and Kordos (1990) that is a Pleistocene Formation. Tóthné Makk et al. (2023) created a new system in which this material is presented as a Quaternary fluvial sediment, not an independent Formation. Almost no attention has been paid to the red sandstone pebbles until now, apart from a few older publications (Horusitzky 1917; Kriván 1973; Jaskó and Kordos 1990, Bors and Vörös 2008; Micsinai and Molnár 2010; Biró et al. 2013; Spránitz et al. 2017).

Materials and methods

Sampling

A total of 234 pieces of red-coloured sandstone samples were examined. Out of these 110 fragments represented ground stone tools from Gorzsa (Suppl. Table 1). More than half of the archaeological samples were found in a well-defined archaeological context, along a distinct layer. About 82% of them are dated to the Late Neolithic period and the rest belong to younger periods (e.g. Bronze Age, Iron Age, and Sarmatian period, Fig. 2b) (Horváth 1982). The 18% of the red-coloured sandstone GSTs have been found in Neolithic pits that cut through 2 – 3 or even more Neolithic layers ('Pits type X'). Only few analysed sandstone artefacts cannot be connected to a precise archaeological context, as they were either found inside multi-period pits ('Pits type Y'), which cut through some Neolithic and 'younger' layers (5%), or were stray finds scattered throughout the archaeological site or found in an undatable context (stray find, 18%) (Fig. 2b). The analysed archaeological samples belong to the collections of the János Tornyai Museum (Hódmezővásárhely, Hungary).

The archaeometric analysis allowed us to define the rock types constituting the sampled archaeological finds and their provenance at various degrees of resolution. For the more precise provenance analysis simultaneous petrographic and geochemical investigations were carried out on the archaeological finds and even on the geological samples. The latter ones are comparative sandstone samples (124 geological samples, Suppl. Table 2) from different locations of the Pannonian Basin and its surrounding (Fig. 1). They can be originated from primary and also from secondary occurrences. Primary occurrences of Permo-Triassic red sandstones were analysed from five different formations, Kővágószőlős- and Jakabhegy formations (Mecsek Mountains, SW Hungary), Balatonfelvidék Formation (from Balaton Highlands, NW Hungary), and additional samples from the Permo-Triassic sequences of the Papuk- (the upper part of the succession, Northwestern Croatia) and the Codru-Moma Mountains (Apuseni Mountains, SW Carpathians, Romania, Suppl. Table 3). In case of the Papuk samples the sampling process was carried out based on the results of Szakmány et al. (2003). Additionally, three different, secondary occurrences were also investigated, pebbles from the recent debris of the Maros-valley (E Hungary, W Romania), pebbles from Miocene siliciclastic sediments of the Western Mecsek Mts. (Szászvár Formation, SW Hungary) and pebbles from the Pleistocene terraces of the Danube from around Dunavarsány, that previously belonged to the Délpest Pebble Formation (Central Hungary, Suppl. Table 3). Red-coloured sandstone pebbles from the Miocene sequence (Szászvár Formation) of Mecsek Mountains are called as 'Mecsek pebbles' in this paper. In the Maros pebble category, there are different types of pebbles, which were collected from the recent drainage of the Maros River. Out of these, we only deal with red-coloured sandstone pebble variants in detail in this paper. Red-coloured sandstone pebbles of the Pleistocene terrace of the Danube River from Dunavarsány are named 'Danube pebbles' in our article.

Petrography

The raw material of the red-coloured sandstone GSTs found at Hódmezővásárhely-Gorzsa were classified on the basis of macroscopic observations. In the case of each geological sandstone samples the same petrographic methods were used. Fragmented stone tools (52 pieces) and geological samples (121 pieces) were selected for the thin section study. In the case of the archaeological finds, sampling was carried out with the permission of the Tornyai János and the Móra Ferenc Museums. The fractured surfaces were used for sampling and the prepared thin sections were studied under a polarize microscope (Leica DM 2700P couple with Leica K5C camera and a Nikon Optiphot2-pol couple with a Nikon CoolPixDS-Fil camera). Both the main components and the accessory heavy minerals of the analysed samples were investigated (e.g. Garzanti and Vezzoli 2003; Whitney and Evans 2010).

Heavy minerals are detrital grains that have high density but occur in small quantities (their total quantity rarely makes up more than one percent of the whole rock/sediment) and size (63 to 250 micrometres, Garzanti and Andò 2007, 2019). Heavy mineral preparation and optical microscopy play a key role in these works, as they yield information on the genetics and lithology of the source rock. However, this information can be changed by additional factors (e.g. weathering, mechanical abrasion, hydraulic behaviour, and burial diagenesis) that operate during the sedimentation cycle (Mange and Maurer 1992; Morton and Hallworth 1994, 1999; Garzanti 2016). In our case there is a good opportunity of using heavy minerals as indicators for provenance as there is a wide variety of detrital heavy minerals in sandstones (e.g. over 50 translucent detrital minerals were described by Mange and Maurer 1992). Furthermore, these accessory components are more informative for the provenance determination than the main components ('light minerals' have very similar composition, so they can be hardly differentiated) in the case of the siliciclastic rocks, such as sandstones. HMs need to be separated from the main components using dense liquid, such as bromoform (2.89 g/cm^3) and sodium-polytungstate. The latter is a non-toxic compound with adjustable density (ca. 2.89–2.97 g/cm^3). Mineral grains with high-density sink down in these liquids, which permits their complete segregation from the less dense framework components ('light minerals', Mange and Maurer 1992; Andò 2020). Therefore, HMs can be studied in higher concentrations, using the 'immersion method' of Petelin (1961). For HM mounts it was required to include approximately 1000 – 1200 pieces of HMs. After the sample preparation about 300 transparent, randomly selected (ribbon counting) heavy mineral grains were counted from heavy mineral mounts per sample (on 11 GSTs and 33 geological samples; Mange and Maurer 1992; Józsa et al. 2016). Identification was made based on optical properties of each mineral type described by Mange and Maurer (1992). The results are more representative of the entire sample.

Classification of the sandstones was carried out by determining and measuring the quantitative proportions of the sand sized (0.063 to 2.0 mm in width) detrital fragments. Based on the ratio of other components (i.e. matrix, cement and pores) and detrital grains, arenites and wacke can also be distinguished (Pettijohn et al. 1973; Ingersoll et al. 1984; Tucker 2001). In this study, a complex volumetric point-counting method was applied. Detrital grains were identified based on its principals of the 'traditional' and the 'Gazzi-Dickinson' methods (e.g. Dickinson 1970; Gazzi 1966; Ingersoll et al. 1984). In case of coarse-grained composite detrital grains, such as granite, gneiss, and/or mica schist, detrital grains were described as feldspar, quartz (mono-, or polycrystalline), and/or micas in plutonic and/or in medium grade metamorphic rocks. (Pettijohn triangle (QFL) diagrams (Pettijohn et al. 1973) are used to present the results.

Whole-rock geochemistry

Bulk-rock geochemical examinations were carried out by prompt-gamma activation analysis (PGAA) and neutron activation analysis (NAA). PGAA measurements were performed on a selection of 41 selected sandstone samples (11 sandstone archaeological finds and 30 geological samples) at the PGAA instrument of the Budapest Neutron Centre (i.e. BNC, Suppl. Table 4). In the case of PGAA measurement, in most cases entire samples were measured. The samples were placed into the guided external horizontal beam of cold neutrons (with 7.75×10^7 $cm^{-2}s^{-1}$ thermal equivalent intensity, Szentmiklósi et al. 2010) and irradiated for 1700 – 52.000 s. The prompt-gamma photons were detected with a high purity germanium detector-bismuth germanate scintillation detector system (i.e. HPGe-BGO) and the spectra were evaluated with the Hyperlab (Simonits et al. 2003) and ProSpeRo in-house softwares (Révay et al. 2005).

NAA measurements of 40 selected samples (11 archaeological finds and 29 geological samples) were carried out at the NAA laboratory of the BNC (Suppl. Table 4). Samples were weighed (150 – 180 mg) and sealed in high-purity quartz ampoules. The samples were irradiated in the rotating, well-thermalized channel for 4 h, together with a set of monitor foils: Zr, and Au 0.1% in Al (IRMM-530) to get the neutron flux parameters. The thermal neutron flux density has been 2.2×10^{13} $cm^{-2}s^{-1}$, with $f = 45$ (thermal/epithermal ratio) during irradiation. After the irradiation the gamma spectra of the samples were collected on two detectors, a Canberra HPGe and an Ortec HPGe p-type detector and connected to a dual-input ORTEC DSPEC 502 digital gamma spectrometer, which is controlled by the ORTEC Maestro 7 software. For spectrum evaluation, HyperLab 2013.1 software was used (Simonits et al. 2003). For the identification of radioactive isotopes and element concentration calculations, the KayZero program (De Corte et al. 2001) was applied.

Heavy minerals and tourmaline mineral chemistry

The chemical composition of select heavy minerals, including tourmaline, apatite, garnet, amphibole, and pyroxene, was determined using SEM-EDX on polished heavy mineral separates obtained from 37 sandstone samples (11 archaeological and 26 geological) (see Suppl. Table 5). Tourmaline played a particularly prominent role due to its exceptional stability and ubiquitous presence that was present in significant quantities and in various colours in all the investigated samples. Notably, tourmaline exhibits a wide range of colours (brown, green, blue, etc.) and geochemical compositions, making it a valuable tool for differentiating source materials. Therefore, a detailed mineralogical and mineral chemical examination was performed on them.

The analyses were conducted at the HUN-REN Centre for Energy Research, Budapest. Primarily, an Oxford Ultimax 40 EDX detector mounted on a Zeiss LEO 1540 XB SEM was utilized with the following operating conditions: 21 kV accelerating voltage, 3 nA beam current, and 30 s signal acquisition time. Additionally, a ThermoScientific Scios 2 equipped with an Oxford Xmax 20 EDX detector was employed, operating at 20 kV accelerating voltage, 1.6 nA beam current, and 30 s signal acquisition time.

In the case of tourmalines, electron microprobe measurements can only be considered as partial chemical analyses, since some of the essential components of the tourmaline (i.e. H, Li, B) cannot be measured by conventional EDS method. Furthermore, data cannot be received on the valence ratios of the transition metals (i.e. Fe, Mn), therefore a normalization procedure needs to be used to calculate the formula of these minerals (Clark 2007). Tourmaline compositions were calculated with the Excel spreadsheet of Selway and Xiong (Selway 2002), normalizing the analyses to 31 anions and assuming B stoichiometric value of 3 apfu (atoms per formula, apfu) and $OH + F = 4$ apfu. The proportions of B_2O_3, H_2O and Li_2O were calculated by stoichiometry, where the Fe_{total} was assumed to be all Fe^{2+}. Tourmaline can be classified into several groups based on the dominant occupancy of the X site. Tourmalines have been described containing dominant Na^+, Ca^{2+}, $^X\square$ (i.e. vacancy of the X site), and, rarely, K^+ (Henry et al. 2011). However, due to the relatively rare occurrence of K-rich tourmalines, it is practical to combine the content of the Na^+ and K^+, into an alkali group. This way, alkali-, calcic-, and X vacant-tourmaline groups can be separated. X-site occupancy generally reflects the paragenesis of the rock in which these tourmalines crystallize. In order to group tourmalines, there is an another diagram type, $Fe/(Fe + Mg)$ vs. $^X\square/(^X\square + Na^+ + K^+)$ (Fehér 2022).

Results

Macroscopic study

Macroscopic features of the archaeological finds from Hódmezővásárhely-Gorzsa

Based on macroscopic observations four groups of archaeological finds were distinguished, including 'Red – 1',

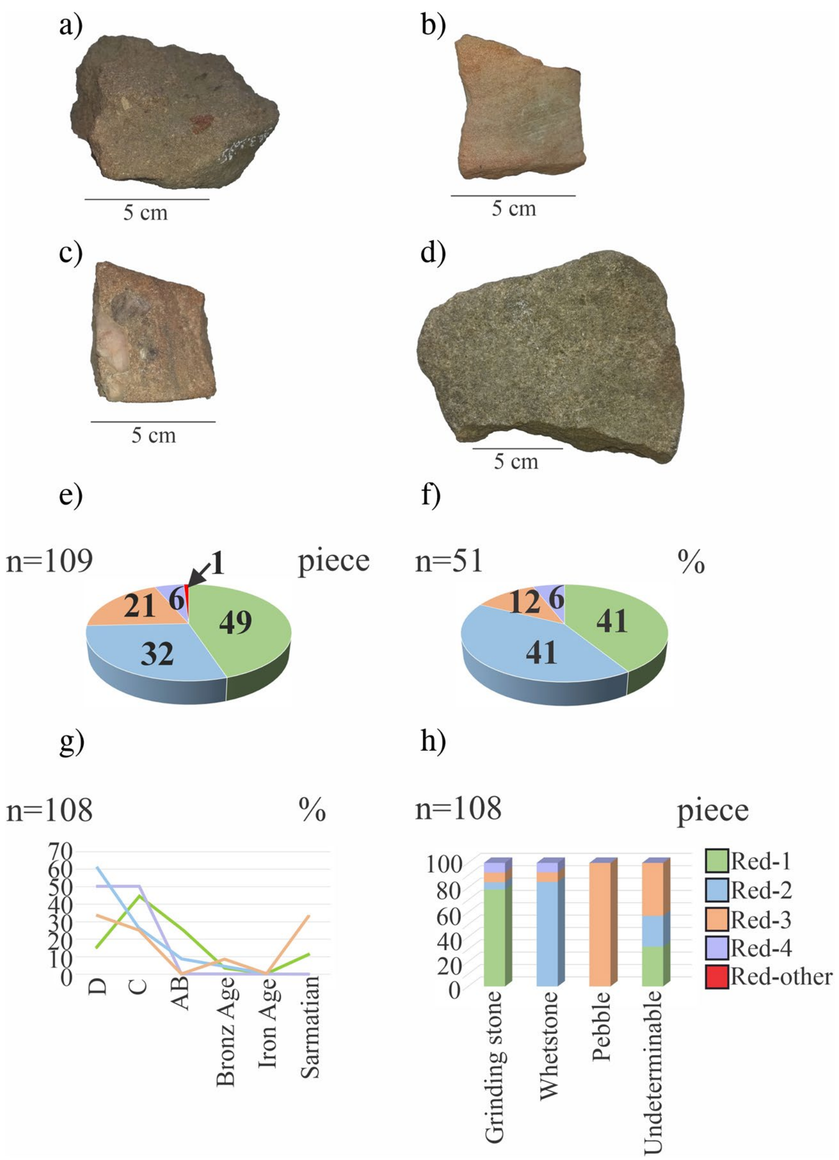

Fig. 3 Macroscopic characteristics (**a** – **d**), age and functional distributions of the red sandstone tools from Gorzsa. (**a**) GOR-592 (Red – 1), (**b**) GOR-854 (Red – 2), (**c**) GOR-534 (Red – 3), (**d**) GOR-349 (Red – 4), (**e**) Pie chart showing the quantitative distribution of the red sandstone types of the whole site, (**f**) Pie chart showing the quantitative distribution (percentage) of the red sandstone types from the Neolithic layers, (**g**) The archaeological age distribution of red sandstone types of Gorzsa, (**h**) The function distribution of the four main types of red sandstones from Gorzsa

'Red – 2', 'Red – 3' and 'Red – 4' (Fig. 3a-d). Samples of 'Red – 1' group (49 pieces, 45% of the assemblage) show a homogeneous composition. They are described as red-, purple-purplish red, grey-greyish red coloured, poorly sorted, coarse – very coarse, or rarely fine – medium-grained sandstones incorporating large quantities of quartz and volcanic rock fragments (Fig. 3a). Samples of the 'Red – 2' group (32 pieces, 29% of the assemblage) red-, pale red, brown-brownish red, purple-purplish red, grey, yellow-yellowish brown, brownish yellow coloured, well sorted, fine – medium grained, very porous sandstones were present (Fig. 3b). Sandstones of 'Red – 3' group (21 pieces, 19% of the assemblage) are red, purple-purplish red and grey-greyish red, well – medium sorted, compact variants with a fine – medium to large – coarse grain size (Fig. 3c). Such artefacts have been manufactured from pebbles. Reddish-grey or greyish-red coloured, coarse – very coarse or rarely medium-grained, medium sorted, compact sandstones (Fig. 3d) formed an independent group, being 'Red – 4' type (6 pieces, 6% of the assemblage).

Based on the petrographic analyses of the archaeological samples, four red sandstone groups of the GSTs and an other red sandstone sample with a special composition (GOR-970) could be distinguished. The latter one could be separated by its quartz, feldspar and matrix content. Among the ground stone tools examined from the settlement, 'Red – 1' and 'Red – 2' type sandstones occurred in the largest number, followed by 'Red – 3' and 'Red – 4' in decreasing order (Fig. 3e). Most of the tools from the Neolithic layers (51 pieces) are made of sandstone 'Red – 1' and 'Red – 2' (Fig. 3f).

The age distribution of the archaeological finds is as follows (Fig. 3g): from the Neolithic settlement phases (D – AB) all red sandstone types are present, but several differences were noticed. In the oldest phase (‘D’), the dominant sandstone type is ‘Red – 2’ followed by the ‘Red – 3’ type. In the D – C phases, half of the tools are of ‘Red – 4’. At the border of the ‘D’ and ‘C’ phases, changes to the opposite direction can be observed; ‘Red – 2’ and ‘Red – 3’ items show decreasing, while ‘Red – 1’ show increasing tendencies. Moreover, regarding the transition of ‘C’ and ‘AB’ phases, in the case of the ‘Red – 1’ and ‘Red – 3’ types a stronger, while in the case of ‘Red – 2’ type a more moderate decrease could be observed (Fig. 3g). Only a few red sandstone ground stone tools were found related to the younger periods (Bronze Age, Iron Age and Sarmatian period) (Fig. 3g). In the Bronze Age contexts, from the red-coloured sandstone variants, ‘Red – 3’ type was the dominant, followed by the ‘Red – 2’ and the ‘Red – 1’ types. The number of the sandstone tools dated to the Iron Ages show a strong decline. In the Sarmatian layers, ‘Red – 2’ is not present and ‘Red – 3’ shows an intensive increase, whilst ‘Red – 1’ exhibits a moderate growth. These patterns are most probably the results of the different strategies of raw material choices during the different periods.

Regarding the use/function of the various sandstone types, the following conclusions can be drawn (Suppl. Table 1, Fig. 3h): most of the grinding stones are made of red sandstone, ‘Red – 1’ type, whetstones were dominantly made of ‘Red – 2’ and all the red sandstone pebbles are of ‘Red – 3’. Among the tool fragments, there are lots of ‘Red – 3’, some ‘Red – 1’ and ‘Red – 2’. The fragmented state of these finds does not allow the precise determination of their typology and possible function. The ‘uncertain’ category in Suppl. Table 1 implies that the find cannot be specifically categorised and/or its multifunctional use cannot be securely excluded. The expression ‘undeterminable fragment’ in Fig. 3h and Suppl. Table 1 indicates that due to missing typological features, proper identification cannot be done. Among ‘Red – 4’ and ‘Red – 3’, both grinding stones and whetstones were present.

Macroscopic features of the potential source rocks

The macroscopic petrographic features of each sandstone occurrences from different geographical areas show significant macroscopic and compositional variability (Fig. 4a-f). Samples of the Kővágószőlős and Jakabhegy Sandstone formations are red-purplish red coloured, weakly sorted, coarse – very coarse grained, sometimes pebbly sandstones, with sometimes layered and even cross-layered versions (Fig. 4a-b). Balatonfelvidék Sandstone is a dark red, red, sometimes brownish-red coloured, weakly sorted, medium-coarse grained sandstones with some pebbles or very-fine – fine grained sandstone and/or siltstones

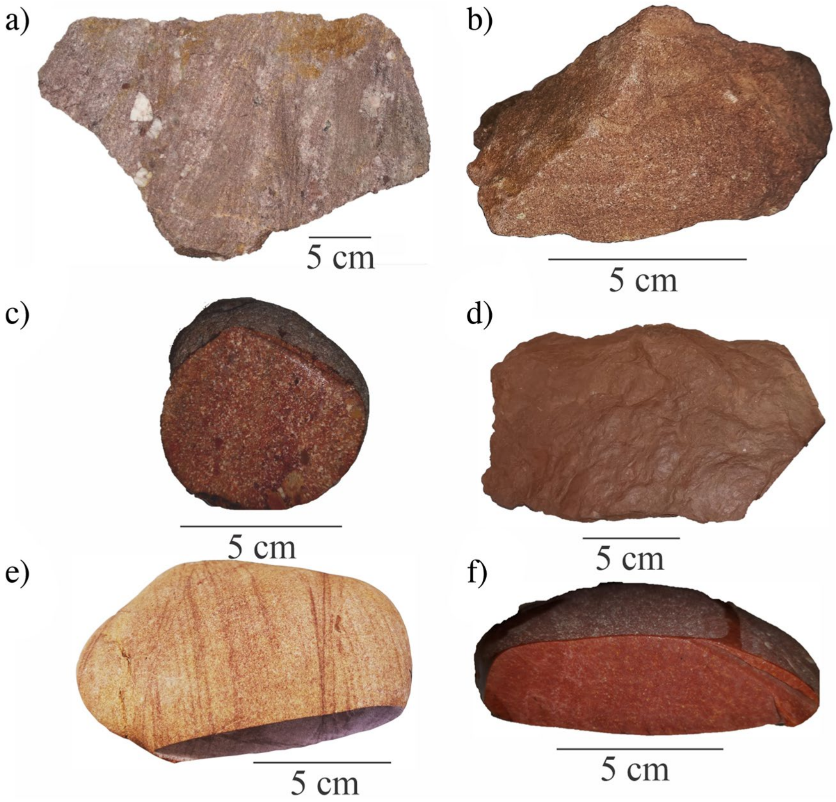

Fig. 4 Macroscopic photographs of the geological samples. (**a**) Jakabhegy Sandstone, (**b**) Kővágószőlős Sandstone, (**c**) Pebble from the Miocene sequence of the Mecsek Mts, (**d**) Balaton Highlands, (**e**) Pebble from Dunavarsány exhibiting cross-lamination, (**f**) Pebble from the Maros River. All scales are 5 cm in length

(Fig. 4d). Papuk samples are reddish-grey – yellowish-grey coloured, weakly sorted fine-medium grained and Codru sample is a purplish red-coloured, medium sorted medium-grained sandstone. Among the Maros pebbles, there are also red types with a variety of colours and shades (e.g. red, purplish-red, brown-brownish red, grey-greyish red) that were described as medium – well sorted coarse-very coarse and/or fine-medium grained sandstones (Fig. 4f). The red-coloured sandstone pebbles of Mecsek (Fig. 4c) show a varied appearance in terms of colour, sorting and appearance. There are a lot of colours and shade among these pebbles, their classification cannot be taken about the macroscopic properties. Pebbles of the Danube are red, pale red and purplish-red, weakly sorted, fine – medium grained sandstones (Fig. 4e) with sometimes layered and even cross-layered versions. Based on the macroscopic observations certain raw material groups could be distinguished. However, neither the classification of sandstone types, nor the correlation with archaeological materials, and the determination of provenance can be based only on macroscopic petrographic analysis.

Optical microscopic investigations

Microscopic features and heavy mineral composition of the archaeological finds from Hódmezővásárhely-Gorzsa

Sandstones of 'Red – 1' type can be distinguished based on the large quantities of quartz and volcanic rock fragments (Fig. 5c), with small to medium amounts of feldspar (subtypes 1a (< 15%), and 1b (> 15%) (Miklós et al. 2021). The grains are originally well-rounded, with well-developed syntaxial siliceous cement. There are sericite pseudomatrix and a few pores (Table 2). 'Red – 2' group can be identified based on the large quantities of quartz, micas and pores (Fig. 5f). Samples of this

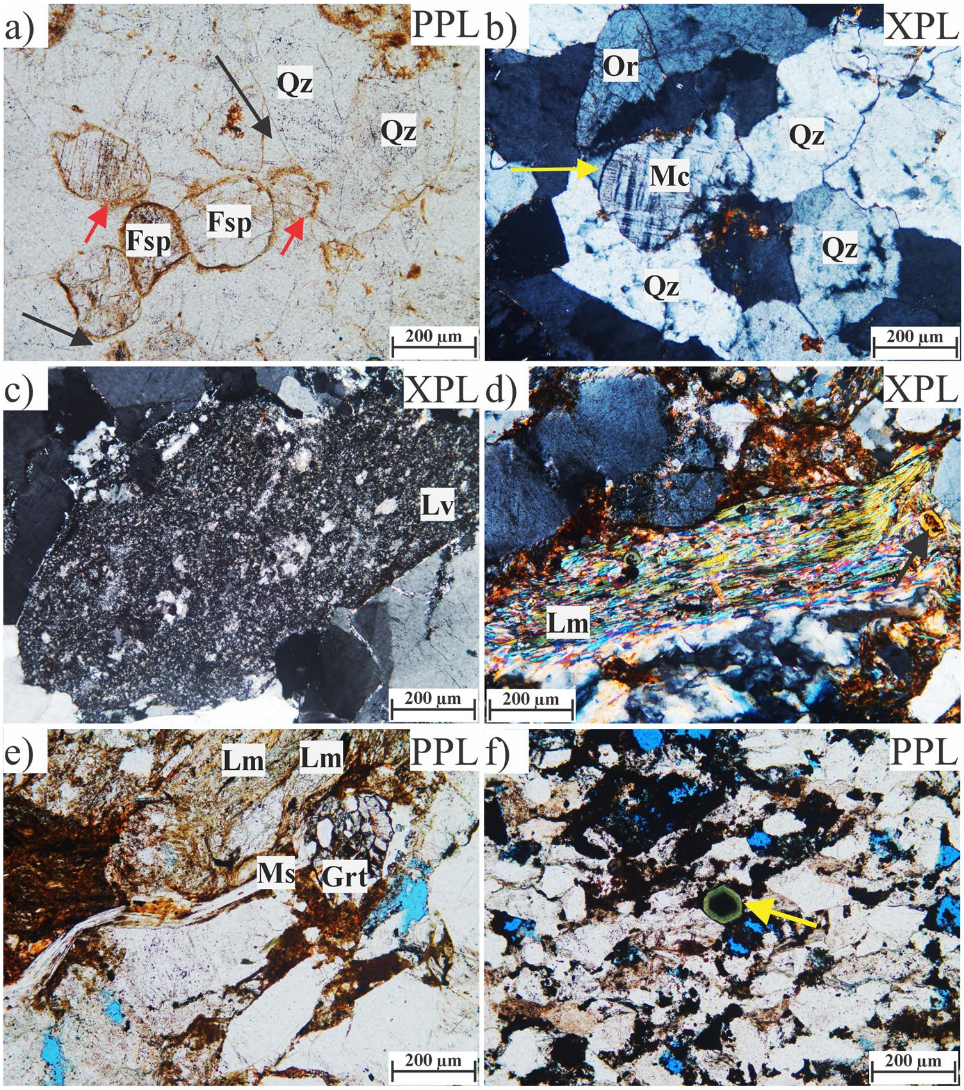

Fig. 5 Photomicrographs of the red sandstone tools from Gorzsa. (**a**) Mature sandstone, with some feldspar (with red arrows) and quartz cement (with black arrows) (GOR-90, Red – 3); (**b**) Medium-, or well-rounded microcline with quartz cement (with yellow arrow) (GOR-90, Red – 3); (**c**) Felsic volcanic rock fragments (GOR-76, Red – 1); (**d**) Micaschist rock fragment with brown-coloured tourmalines (GOR-673, Red – 4); (**e**) Garnet, muscovite and grains of metamorphic origin (GOR-673, Red – 4); (**f**) Green-coloured, poorly-rounded tourmaline with colour zoning (dark green core and pale green rim, with yellow arrow) in fine-grained sandstone (GOR-92, Red – 2). Abbreviations: *Fsp* feldspar, *Grt* garnet, *Lm* metamorphic lithics, *Lp* plutonic lithics, *Lv* volcanic lithics, *Mc* microcline, *Ms* muscovite, *Or* orthoclase, *Qz* quartz (after Whitney and Evans 2010), *PPL* plan polarized light, *XPL* cross polarized light

Table 2 Microscopic features of the Gorzsa red sandstone tools

Detrital components, fabric	'Red – 1'	'Red – 2'	'Red – 3'	'Red – 4'
Grain size	coarse – very coarse	very fine – medium	medium – coarse	coarse – very coarse
Sorting	weak – medium	well	medium – well	medium
Roundness	well	weakly	well	medium
Cement	siliceous (sericite)	siliceous, sericite, goethite, carbonate	siliceous, limonite (sericite-nontronite)	siliceous, albite, limonite (sericite-nontronite)
Quartz	Qp > > Qm	Qm > > Qp	Qm > > Qp	Qm > > Qp
Feldspar	Kfs > > Pl	Pl > > Kfs	Kfs > > Pl	Pl > > Kfs
Mica	+	+++	+	+
Volcanic fragments	+++	+	+	+
Metamorphic rock fragments		+	+	+++
Plutonic rock fragments				++
Heavy minerals	**'Red – 1'**	**'Red – 2'**	**'Red – 3'**	**'Red – 4'**
Tourmaline	+ (yellowish brown)	+++ (green and brown)	++ (greenish brown)	++ (brown and green)
Zircon	+	++	+	
Apatite		+		
Rutile	+	+	+	++
Titanite	+	+	+	++
Garnet				++

(+) = very rare, + = rare, ++ = common, +++ = very common

Abbreviations: *Qm* monocrystalline quartz, *Qp* polycrystalline quartz, *Kfs* kalifeldspar, *Pl* plagioclase

group contain smaller amounts of feldspar and rock fragments as well. The grains are weakly rounded with a considerable amount of syntaxial siliceous, carbonate and goethite cement. There can be seen some sericite as pseudomatrix (Table 2). Samples of 'Red – 3' group are mature sandstones, which are mainly composed of quartz, less feldspar (dominantly K-feldspar) and micas (dominantly muscovite). The grains are well-rounded with syntaxial siliceous and/or sometimes albite (feldspar) cement (Fig. 5a-b; Table 2). The samples of the fourth group ('Red – 4') are composed mainly of quartz and metamorphic-metasedimentary rock fragments (phyllite, metasiltstone-metasandstone, Fig. 5d-e). They also contain feldspar and muscovite. The grains are originally medium rounded mainly with syntaxial siliceous cement (Table 2). GOR-970 sample is a red-coloured sandstone sample, with a unique/special composition, which distinguishes it from the materials of the above-mentioned four groups. This kind of sandstone was a quartzarenite with monocrystalline and polycrystalline quartz grains in similar ratios, a lot of matrix and very low amounts of feldspar.

In the case of zircon/monazite and apatite, the composition of the phases was clarified with SEM-EDX measurements that were written in parenthesis. Archaeological samples belonging to the 'Red – 1' group contain the smallest amount (0.16%) of heavy minerals, there are opaque minerals (titanomagnetite and ilmenite) in large quantities among them. Transparent heavy mineral grains are as follows: zircon/monazite (zircon, monazite > > xenotime; Fig. 7g), rutile, apatite (fluor-chloro-apatite > > fluorapatite), tourmaline, titanite (Fig. 6) and amphiboles, kyanites, Cr-spinels, hollandites, staurolites and cassiterites in traces. Samples of 'Red – 2' type contain the highest amount (1.70%) of heavy minerals among the red-coloured raw material types with lots of opaque grains (ulvospinel, titanomagnetite and ilmenite). The observed transparent heavy minerals were tourmaline, rutile, titanite, zircon/monazite (zircon, xenotime and florencite in the same amounts), apatite (only fluorapatite could be identified) and amphiboles, garnet (Fig. 6) and olivine in traces. Small amount (0.60%) of heavy minerals could be observed in the third red-coloured raw material type, 'Red – 3'. There are some opaque minerals (only titanomagnetite), but these are occurred in smaller amounts, than in case of the above-mentioned groups. Among the transparent heavy minerals, zircon/monazite (zircon > > monazite), tourmaline, rutile, titanite, amphibole and a few fluorapatite, staurolite, epidote-group (Fig. 6) and olivine and cassiterite in traces could be observed. Sandstones of 'Red – 4' type contain a high amount (1.42%) of heavy minerals and a few opaque mineral phases, such as sphalerite. Among the transparent fraction were garnet (Fig. 7f), apatite (fluorapatite > > fluor-chloro-apatite), rutile, zircon/monazite (zircon and monazite in almost the same proportion), tourmaline, epidote-group, titanite and amphibole and kyanite in traces (Fig. 6).

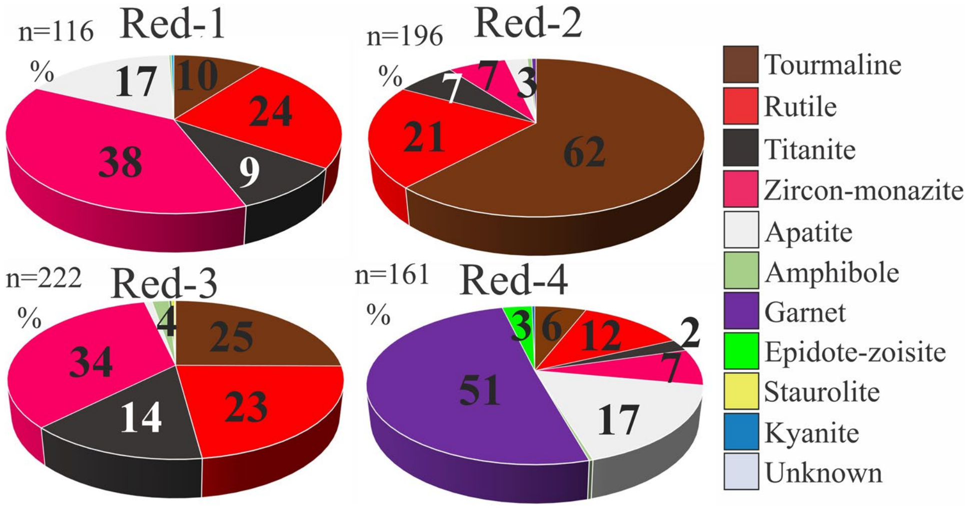

Fig. 6 Quantitative distribution of detrital transparent heavy mineral species of the ground stone tools of Gorzsa. The ‘unknown’ category mainly refers to slightly weathered, fragmented grains, probably zircon or titanite grains

Microscopic features and heavy minerals of potential source rocks

The samples of the Jakabhegy Sandstone Formation contain medium – well rounded K-feldspar (orthoclase > microcline, Fig. 8b), granitoid, and acidic volcanic rock fragments and between them a thick, syntaxial siliceous overgrowth can be observed (Table 3). The Kővágószőlős Sandstone samples are medium – well rounded sandstones that contains K-feldspar and plagioclase in similar quantities, moreover acidic and intermediate volcanic and granitoid rock fragments (Fig. 8c). Siliceous overgrowth sometimes can also be observed, but in a very thin form (Table 3). Samples of the Balatonfelvidék Sandstone Formation are weakly-rounded sandstones with a lot of acidic volcanic rock fragments (Fig. 8f), quartz and clay minerals, a small amount of feldspar and micas (Table 3). Papuk samples are well-rounded sandstones with a lot of quartz and quartzite, a few feldspar grains. The samples were cemented with siliceous material and a high amount of sericite can also be described (pseudomatrix, Table 3). The Codru sample is a weakly-rounded sandstone with a lot of quartz, feldspar, some mica, and intergranular sericite (pseudomatrix) and original matrix. In addition, small amounts of acidic volcanic (Fig. 8d), granitoid, and even metasedimentary rock fragments were also described (Table 3). Maros pebbles are well-rounded sandstones with a lot of quartz, quartzite, varying amounts of feldspar, and small amount of mica, metasedimentary grains (Fig. 8e). A thicker siliceous overgrowth can be observed between the grains (Table 3). The red-coloured sandstone pebbles of the Mecsek show a varied appearance by polarizing microscope, three groups were distinguished. The first group consists of quartz, quartzite, less K-feldspar, mica, acidic volcanic and granitoid rock fragments, and a thick syntaxial siliceous cement (Fig. 8a; Table 3). The second group consists of poorly sorted quartz, K-feldspar, and plagioclase in nearly equal proportions, mica, and small amount of acidic and mafic volcanic and granitoid rock fragments. In the third group, there are well-sorted, very fine-grained sandstone pebbles, which consist of quartz, plagioclase and less amount of K-feldspar, moreover large number of micas, and acidic and mafic volcanic and granitoid rock fragments can also be seen. The Dunavarsány pebbles are medium-rounded sandstones with a lot of quartz, quartzite, feldspar, and muscovite. Metamorphic and metasedimentary (e.g. phyllite, metasandstone, etc.) rock fragments were also described. Siliceous, albite, and sometimes carbonate cement can also be identified between the grains.

Based on the heavy mineral composition, the possible raw materials can be classified into four groups (‘character groups’: ‘Group – CI’, ‘Group – CII’, ‘Group – CIII’ and ‘Group – CIV’) (Fig. 9). The first one (‘Group – CI’) includes the Jakabhegy Sandstone (‘Mecs – Jak’) and the Mecsek pebbles II and III (‘Mecs – PebII and III’). These sandstones have a similar transparent heavy mineral composition. Their dominant component is apatite (Fig. 7k); it is present with an average amount of 64%. Tourmaline, rutile (Fig. 7e), titanite and zircon (Fig. 7h) are also visible with roughly the same amounts. There may be minor differences regarding the rare components, such as epidote-group, amphibole, kyanite (Fig. 7l) and/or garnet. The second character group (‘Group – CII’) includes the Kővágószőlős Sandstone (‘Mecs – Kőv’) and the Mecsek pebbles I (‘Mecs – PebI’). No dominant heavy mineral phase could be identified in the examined samples. Four mineral types, namely tourmaline (Fig. 7b), rutile, apatite, and zircon appear in almost equal amounts (almost 20 – 25% for each phase). A fifth phase, titanite, could also be observed with significant quantities in this group. Minor differences could be registered regarding the rare components, such as epidote-group (Fig. 7c), amphibole, garnet, staurolite (Fig. 7i) and/or kyanite. In the third

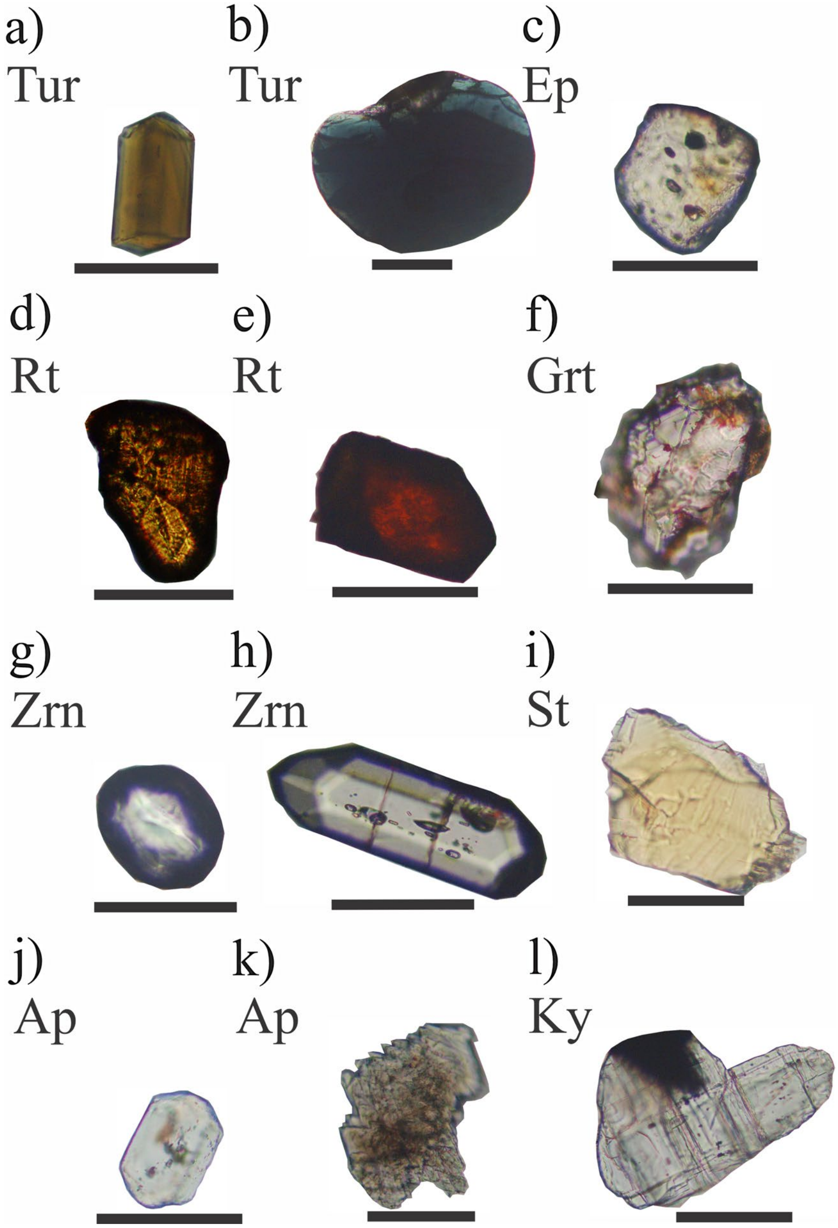

Fig. 7 Microphotographs of different heavy mineral phases of the investigated archaeological and geological samples. (**a**) Brown-coloured tourmaline grain from the sample id. Alö-5 (Balaton Highlands), (**b**) Blue-coloured tourmaline grain from the sample id. II HCs (Mecsek pebble I), (**c**) Epidote from sample id. I HCs (Mecsek pebble I), (**d**) rutile grain from the sample id. M-1/14 (Maros pebble), (**e**) Rutile grain from the sample id. Cs-JFh (Jakabhegy Sandstone Formation), (**f**) Garnet from the GST sample of id. GOR-673 (Gorzsa tool, 'Red – 4'), (**g**) Zircon from the ground stone sample of id. GOR-592 (Gorzsa tool, 'Red – 1'), (**h**) Zircon from the sample of id. Ja-JFhJS (Jakabhegy Sandstone Formation), (**i**) Staurolite grain from the sample id. I HCs (Mecsek pebble I), (**j**) Apatite grain from the sample id. Ká-fü-1 (Balaton Highlands), (**k**) Apatite from the sample id. Ja-JFhJS (Jakabhegy Sandstone Formation), (**l**) Kyanite grain from the sample id. Ja-JFhJS (Jakabhegy Sandstone Formation). Abbreviations: *Tur* tourmaline, *Ep* epidote, *Rt* rutile, *Grt* garnet, *Zrn* zircon, *St* staurolite, *Ap* apatite, *Ky* kyanite

group ('Group – CIII'), there are the Codru ('Cod') and the Danube ('Dan') pebble samples. These sandstones have a similar transparent heavy mineral composition. Their dominant component is zircon, which occurs in an average amount of 52%. In addition, rutile, and tourmaline, as well as titanite, are also common components. There are minor differences in rare components, such as amphibole, staurolite, garnet and/or apatite. The fourth group ('Group – CIV') includes samples from the Balaton Highlands ('Bal – Hgh'), the Maros River pebbles ('Mar') and the Papuk Mts. ('Pap'). Within this group, zircon and tourmaline (Fig. 7a) represent two thirds of total heavy minerals. The combined amount of rutile (Fig. 7d) and titanite equals to 25%. There are minor differences in the observed rare components, such as apatite (Fig. 7j), garnet and/or amphibole.

Whole-rock geochemistry

Bulk-rock geochemical data of the archaeological finds from Hódmezővásárhely-Gorzsa

The tools are characterized by high SiO_2 content ranging from 75.34 to 96.58 wt%. The maximum value was reached in 'Red – 3' and minimum values in 'Red – 2' and 'Red – 4' groups. TiO_2, Al_2O_3 and Fe_2O_{3total} showed similar characteristics; they reached the highest values in 'Red – 2', while the lowest values were registered in

Table 3 Microscopic features of the potential raw material sources

Detrital components, fabric	'Mecs – Kőv'	'Mecs – Jak'	'Bal – Hgh'	'Cod'	'Pap'	'Mar'	'Mecs – PebI'	'Mecs – PebII'	'Mecs – PebIII'	'Dan'
Grain size	siltstone-, very fined – medium grained	coarse – very coarse	siltstone-fine grained and medium – very coarse	fine-medium	fine – medium grained	fine – very coarse grained with fine pebbles	fine – coarse grained	medium – coarse grained with fine pebbles	very fine – medium grained	medium – coarse grained with fine pebbles
Sorting	medium	medium	weak – medium	weak – medium	medium	medium – well	medium	weak	well	well, or weak – medium
Roundness	medium	medium	weak	weak – medium	well	well	well	medium – well	medium	well
Cement	siliceous, sericite	siliceous (sericite)	siliceous, clay minerals	sericite	sericite	siliceous (sericite)	siliceous (sericite)	siliceous (sericite)	siliceous (sericite, carbonate, biotite-nontronite)	siliceous, sericite, carbonate, biotite-nontronite)
Quartz	Qm > > Qp	Qm ~ Qp	Qp > > Qm	Qm > > Qp	Qp > > Qm	Qm > > Qp	Qm > > Qp	Qp > > Qm	Qm > > Qp	Qm > > Qp
Feldspar	Kfs > > Pl	Kfs ~ Pl	Kfs > > Pl	Kfs > > Pl	Kfs > > Pl	Kfs > > Pl	Kfs > > Pl	Kfs ~ Pl	Pl > Kfs	Kfs > > Pl
Mica	+	+	+	+	+	+	+	+	+	+
Volcanic fragments	++	+	+++	+	+	+	+	+	+	+
Metamorph rock fragments		+	+	+	+	+	+	(+)	+(+)	+
Plutonic rock fragments	+	+		+			(+)	+	+(+)	
Heavy minerals	**'Mecs – Kőv'**	**'Mecs – Jak'**	**'Bal – Hgh'**	**'Cod'**	**'Pap'**	**'Mar'**	**'Mecs – PebI'**	**'Mecs – PebII'**	**'Mecs – PebIII'**	**'Dan'**
Tourmaline	+ (yellowish brown)	+ (yellowish green)	+ brown (yellowish) and green	+ brown (yellowish)	(+) brown (yellowish)	++ green and yellowish green	++ brown (greenish)		(+) brown (yellowish)	++ yellowish brown and green
Zircon		+	+	++		++	+	(+)	(+)	++
Apatite	+(+)	++					(+)	+(+)	++	++
Rutile		+	+	+	++	+	++	+	+	++
Titanite			(+)	+	++	+	(+)	+	+	++

(+) = very rare, + = rare, +(+) = medium, ++ = common, +++ = very common

Abbreviations: Qm monocrystalline quartz, *Qp* polycrystalline quartz, *Kfs* kalifeldspar, *Pl* plagioclase, *Mecs – Kőv* Kővágószőlős Sandstone, *Mecs – Jak* Jakabhegy Sandstone, *Bal – Hgh* Balaton-Highlands, *Cod* Codru, *Pap* Papuk, *Mar* Maros pebble, *Mecs – PebI* Mecsek pebble Type – I, *Mecs – PebII* Mecsek pebble Type – II, *Mecs – PebIII* Mecsek pebble Type – III, *Dan* Danube pebble

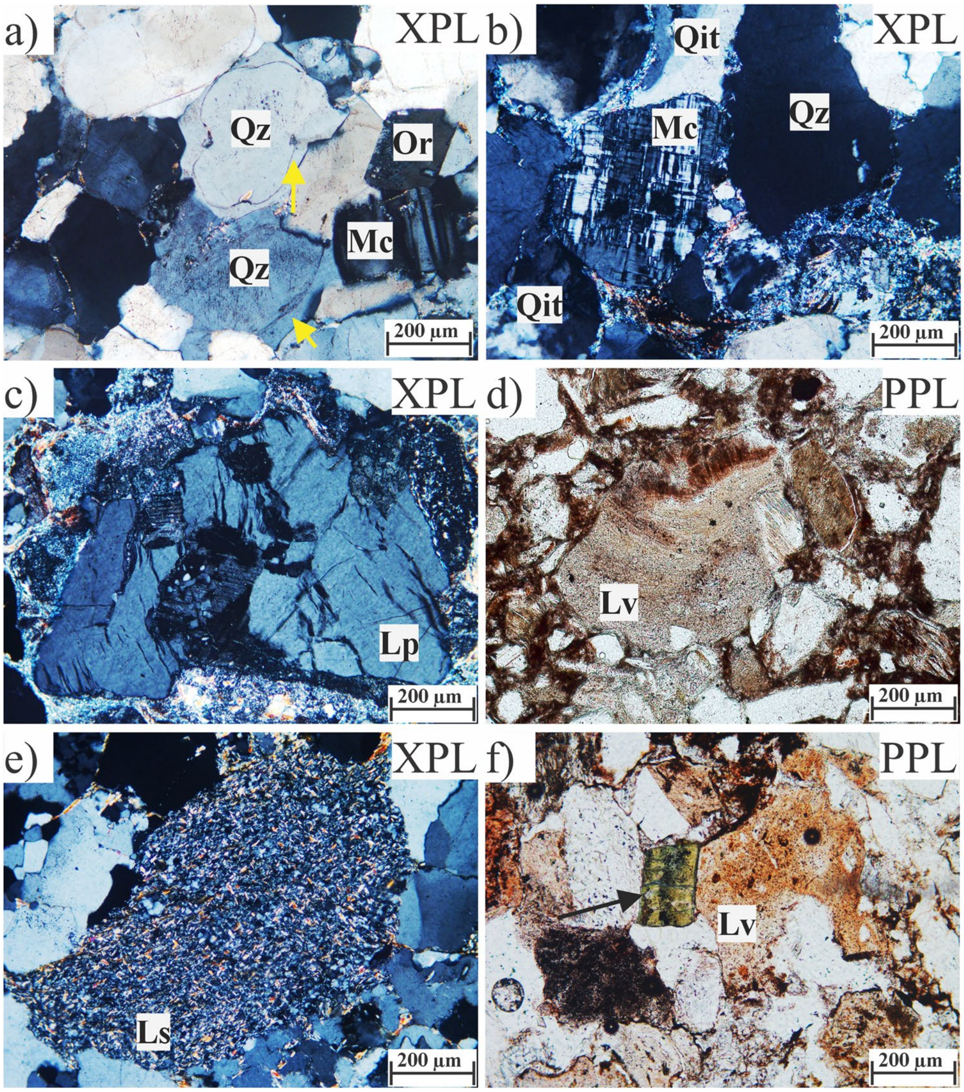

Fig. 8 Photomicrographs of the geological rock samples. (**a**) Monocrystalline quartz (Qz) and microcline (Mc) grains with syntaxial quartz cement (with yellow arrows) (HCs/30, Mecsek pebble I); (**b**) Medium-, or well-rounded microcline and quartzite grains (Ja-JFh, Jakabhegy Sandstone); (**c**) Granitoid rock fragment (Lp) with K-feldspar and plagioclase (Ba-KFhBaT, Kővágószőlős Sandstone); (**d**) Poorly-rounded felsic volcanic rock fragment (Lv), monocrystalline quartz and K-feldspar grains (Codru-01, Codru); (**e**) Medium-rounded metasiltstone rock fragment (Ls) (M-1/19, Maros pebble); (**f**) Poorly rounded brown-coloured tourmaline (with black arrow) and felsic volcanic grains (Lv) from the Balatonfelvidék Sandstone (Káfü-1, Balaton Highlands). *PPL* plan polarized light, *XPL* cross polarized light

'Red – 1' and 'Red – 3' groups. Minor elements of the GSTs are discussed in groups based on their similar characteristics and elemental distributions: Ba and Rb can be found in the highest amount in 'Red – 1' (GOR-133). Rubidium reaches another peak in the case of 'Red – 2' group. The minimum values of Rb can be seen in the case of 'Red – 3' group. Zr, Hf, Th and U have extreme high values in 'Red – 2' group, whilst in the case of 'Red–3', 'Red–4' and some samples of 'Red – 1' groups have lower, but similar quantities. The Ta, Sc, Cr and Co show high values in 'Red – 2' and 'Red – 4' groups and low ratios in 'Red – 1' and 'Red – 3' variants. Boron shows the highest values in 'Red – 1' group and it has similar, but lower values in 'Red – 2', 'Red – 3' and 'Red – 4' groups (Suppl. Table 4). Rare-earth element distribution (REEs) of all the analysed tool samples showed similarities to each other, but in the case of the REE content, differences could be observed among the archaeological groups. The highest REE values were measured in 'Red – 2' and 'Red – 4', and the lowest in 'Red – 1' and 'Red – 3' groups (Suppl. Table 4).

Whole-rock geochemistry of potential sources

The geological samples are characterized by high SiO_2 content ranging from 61.00 to 97.72 wt%. SiO_2 content reaches the highest values in the Maros, the Danube and the Mecsek pebbles I samples, and also in the case of the Papuk samples. Lower SiO_2 quantities were detected in the Jakabhegy and Kővágószőlős Sandstone formations and Mecsek pebble II, whilst the lowest values were observed in the Mecsek pebble III, Codru and the Balaton Highlands samples. TiO_2 and FeO_{total} content varies 0.03 to 0.30 wt% in the case of TiO_2 and 0.41 to 2.15 wt% of FeO_{tot} in the Maros, the Danube and the Mecsek pebbles I and II, moreover in the case of the Papuk, the Kővágószőlős and Jakbahegy Sandstone

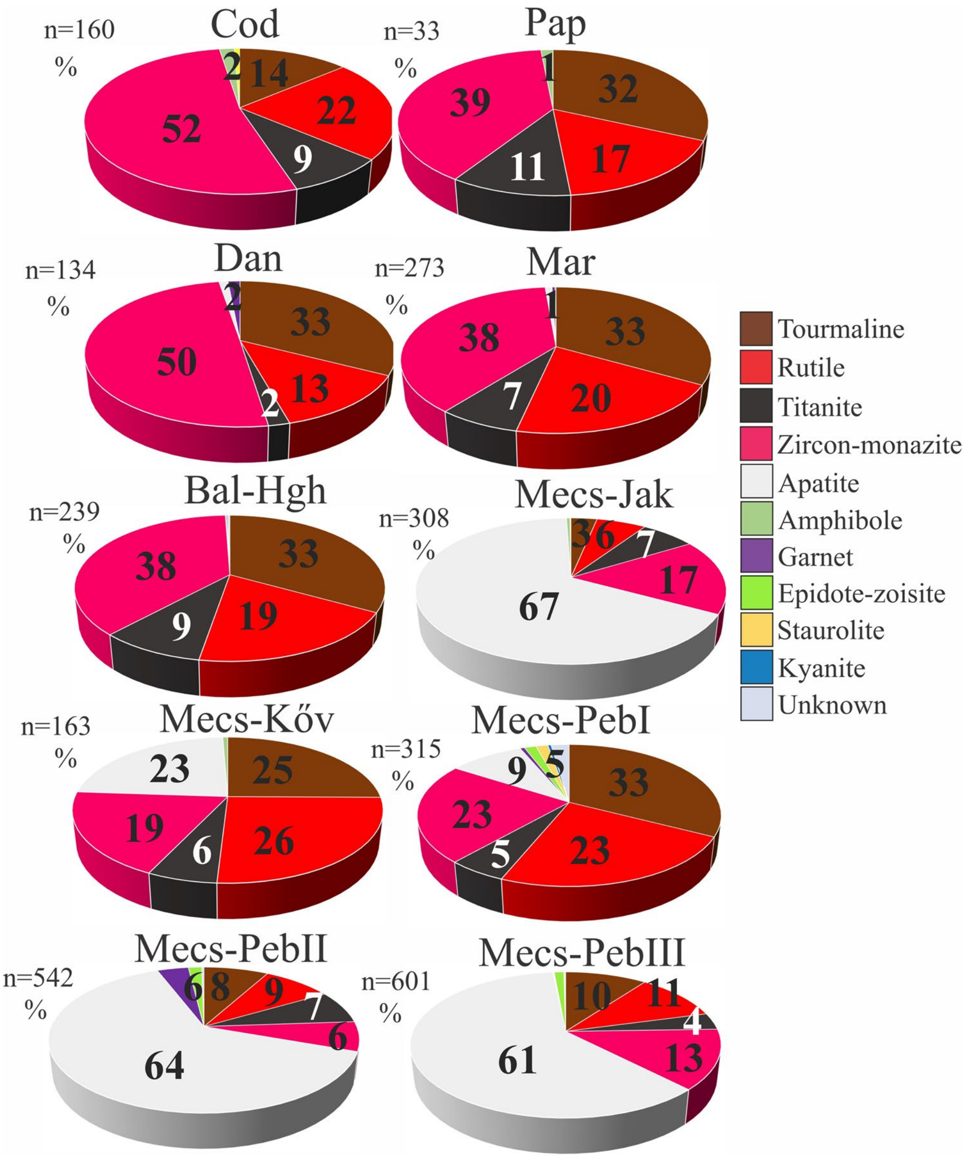

Fig. 9 Quantitative distribution of the detrital transparent heavy mineral phases of the examined geological samples. The 'unknown' category mainly refers to slightly weathered, fragmentary grains, which can usually be zircon or titanite grains. Abbreviations: *Mecs – Kőv* Kővágószőlős Sandstone, *Mecs – Jak* Jakabhegy Sandstone, *Bal – Hgh* Balaton Highlands, *Cod* Codru, *Pap* Papuk, *Mar* Maros pebble, *Mecs – PebI* Mecsek pebble Type – I, *Mecs – PebII* Mecsek pebble Type – II, *Mecs – PebIII* Mecsek pebble Type – III, *Dan* Danube pebble

formations and in the Balaton Highlands samples. Barium and Rb reach the highest amounts in the samples of the Balaton Highlands, Codru, the Jakabhegy Sandstone Formation and in the Mecsek pebbles II and III. The lowest values occur in the Maros, the Danube and the Mecsek pebble I, furthermore in the case of the Papuk and the Kővágószőlős Sandstone Formation samples. The Zr, Hf, Th and U show similar behaviour, so they reach high values in Mecsek pebble III, the Codru and the Balaton Highland samples. The lowest values can be observed in the Kővágószőlős and Jakabhegy Sandstone formations, Papuk and the Danube, Maros and Mecsek pebbles I and II. The Ta, Sc, Co and Cr show high values in the samples of the Balaton Highlands, and Codru, moreover in the Mecsek pebble III. The lowest values can be seen in the Maros, Danube and Mecsek pebbles I and II, moreover in the case of the Papuk, the Kővágószőlős and Jakabhegy Sandstone formations. The last one is B that shows higher values in the Danube, Maros and Mecsek pebbles I and II, furthermore in the case of Papuk samples. In contrast, lower ratios were present in Codru, Kővágószőlős and Jakabhegy Sandstone formations and Mecsek pebble III samples. Rare-earth element distribution of the geological samples shows relative similar behaviours: La, Ce, Nd and Sm show high values in the Balaton Highlands, Codru and Kővágószőlős and Jakabhegy Sandstone formations, Mecsek pebbles II and III. In contrast, in the case of the REE contents, differences could be identified among the raw materials. Their lowest values could be found in the Maros, Danube and the Mecsek pebble I group, furthermore in the case of the Papuk samples. Eu, Tb, Yb and Lu show higher values in the Balaton Highlands, Mecsek pebble III and Codru; lower values in the Maros, the Danube and the Mecsek pebbles I and II, moreover the Papuk and the Jakabhegy Sandstone Formation samples (Supplementary Table 4).

Tourmaline mineral chemistry

Tourmaline chemical data of the archaeological finds from Hódmezővásárhely-Gorzsa

Archaeological samples of ‘Red – 1’, ‘Red – 2’ and ‘Red – 4’ types from Gorzsa contain green- and brown-coloured tourmalines, which have very similar major elemental compositions and are classified as alkali tourmalines, dravite (in all raw material types) and schorl (only in the ‘Red – 4’ type, Figs. 10 and 11, Suppl. Table 5). The data is somewhat scattered, most of the points shift towards the vacancy peak in the case of ‘Red – 1’ and ‘Red – 4’ and the Na^+ peak in the case of ‘Red – 2’ type (Fig. 10). Blue-coloured tourmalines have a higher Na^+ content. Tourmalines of Type ‘Red – 3’ are alkali tourmalines too, but the green and brown variants have different major element compositions: the green ones are richer in calcium than the brown-coloured ones (Fig. 10). Tourmalines with dravite and schorl composition could be originated from medium- and/or low-grade metamorphic rocks (Fig. 11; Supplementary Table 5).

Tourmaline chemistry data of potential sources

Most of the examined tourmalines from the geological samples have alkali composition similar to those of the GSTs. Exceptions are some of the green tourmalines from the Balaton Highlands (‘Bal – Hgh’), the Jakabhegy Sandstone (‘Mecs – Jak’) and the Papuk Mountains (‘Pap’), and some of the brown- and blue-coloured grains of the Danube River pebbles (‘Dan’). In addition, some blue-coloured tourmalines found in Mecsek pebble I (‘Mecs – PebI’) with the highest calcium-component are classified as Ca-tourmalines (Fig. 12). The majority of the potential sources exhibit at least two types of tourmaline compositions. The most common ones being dravite and schorl, but occasionally foitites can also be observed. An exception to this is Mecsek pebble II (‘Mecs – PebII’), where only dravites were identified. Among the tourmalines of the Jakabhegy Sandstone (‘Mecs – Jak’) samples, grains with brown colour were observed that show a transition towards the diagenetic range (see in Fig. 13). Moreover, there are also brown- and green-coloured grains from Mecsek pebbles III and probably I (‘Mecs – PebIII’ and probably ‘Mecs – PebI’) showing a high metamorphic grade (see in Fig. 13, Supplementary Table 5).

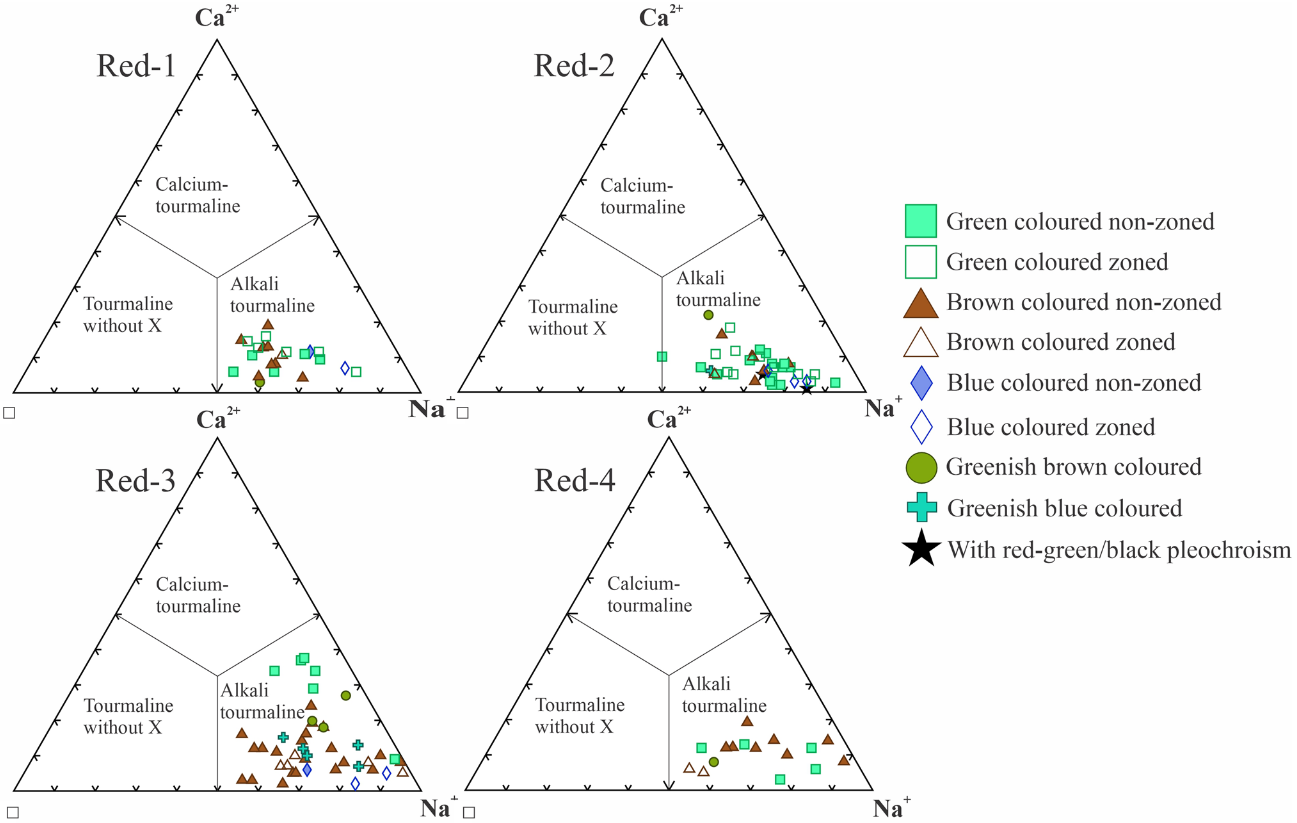

Fig. 10 The X-cation content of the Gorzsa tourmalines of the main tourmaline groups (identified in Red – 1 to – 4 sandstones (triangular diagrams after Henry et al. 2011)

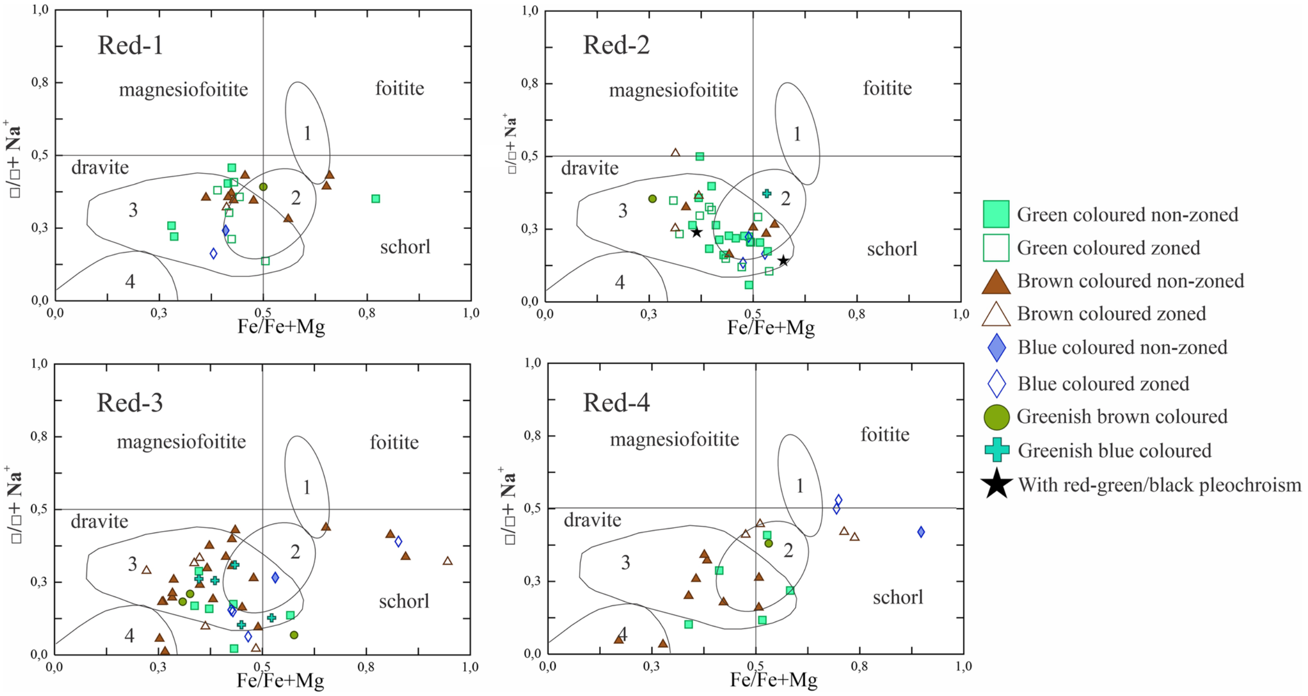

Fig. 11 Chemical composition of the tourmalines of the Gorzsa tools represented in the Fe/(Fe + Mg)^{Y} – □/(□ + Na)^{X} diagram (□ is vacancy). Fields marked with numbers show the typical compositional ranges of tourmalines of different metamorphic grades based on Henry and Dutrow (1996): 1 = diagenetic tourmalines; 2 = low grade tourmalines; 3 = medium grade tourmalines; 4 = high grade tourmalines

Discussion

The diverse tool types show a variety difference in terms of their macroscopic and microscopic features. 'Red – 1', 'Red – 3' and 'Red – 4' have coarse-, or very coarse-grained raw materials. Red-coloured sandstone types can be distinguished based on their sorting, because 'Red – 1' is the least sorted, whilst 'Red – 2' is a well-sorted raw material type with the finest grain size and the highest degree of porosity. The various tool types had quite similar heavy mineral compositions, but slight differences could be spotted among them. Samples of the 'Red – 1', 'Red – 2' and 'Red – 3' were similar in this regard, principally resistant, stable minerals, such as zircon, tourmaline and rutile are enriched in them. Moreover, some other phases, such as opaque minerals (i.e. titanomagnetite and ilmenite), titanite and apatite were also identified; garnet and other minerals with metamorphic origin (e.g. epidote-group, kyanite and staurolite) are rare. On the other hand, 'Red – 4' contained a lot of garnet besides the above-mentioned stable mineral phases. The major-, trace- and REE-element patterns of the finds have been compared with the data of all the above-mentioned formations (see Suppl. Table 4). Figures 15, 16 and 17 show the major-, minor- and trace-elemental compositions of the sandstone formations compared with the artefacts. The high SiO_2 content of the Gorzsa tools is related to their high quartz content that appears in the form of quartz and quartzite grains and/or siliceous cement and the high degree of maturity of the sandstone (e.g. in the case of 'Red – 3' type). TiO_2 and Fe_2O_{3total} values show correlation with the opaque mineral content of the samples (e.g. 'Red – 2', Suppl. Table 4). Comparison of the major and the minor elemental data showed that Rb correlates with K_2O and hence with the K-feldspar content and partly with the maturity of the sandstone, such as 'Red – 1b' and 'Red – 3' types. In the case of Ba, correlation with the K_2O was not observed. The relationship between the Ba and the total feldspar (plagioclase and K-feldspar) content is not clear. Among minor elements: Zr, Hf, Th and U show similar behaviour. In many cases, the measurement of Zr failed; meanwhile Hf had similar properties, and often substitutes Zr in zircon grains. Th and U can also correlate with the quantity of zircon grains, as replacements of Zr happens in the same way as of Hf. Therefore, Zr, Hf, Th, U were related to the heavy mineral content of sandstones and indirectly to the amount of zircon grains, which reach the highest values in the Type 'Red-2' of the Gorzsa finds. Ta, Sc, Cr and Co showed mafic character which, like Ti, were related to the amount of opaque minerals (e.g. in the case of 'Red – 2' type; titanomagnetite, ilmenite and ulvispinel). B content was correlated with the amount of tourmaline grains, such as in the case of 'Red – 2' type, because tourmaline is a heavy mineral with a significant B content. This result agrees with

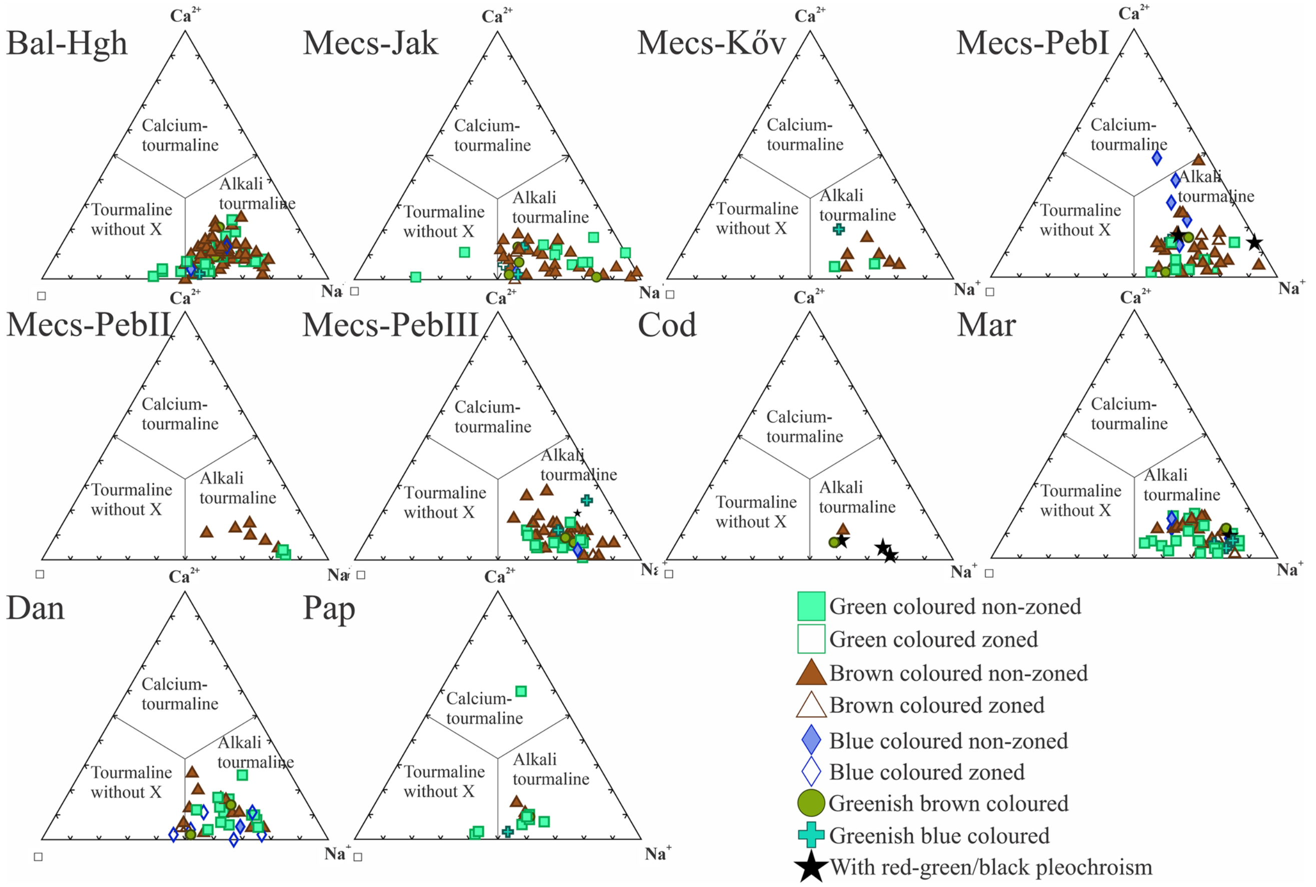

Fig. 12 X-cation content of the tourmalines of the geological samples is shown in the triangular diagram of the main tourmaline groups (diagram modified after Henry et al. 2011). Abbreviations: *Mecs – Kőv* Kővágószőlős Sandstone, *Mecs – Jak* Jakabhegy Sandstone, *Bal – Hgh* Balaton Highlands, *Cod* Codru, *Pap* Papuk, *Mar* Maros pebble, *Mecs – PebI* Mecsek pebble Type – I, *Mecs – PebII* Mecsek pebble Type – II, *Mecs – PebIII* Mecsek pebble Type – III, *Dan* Danube pebble

the heavy mineral content of the samples. All tourmalines of red-coloured sandstone tool types from Gorzsa were classified as alkali tourmalines. Blue-coloured ones often have higher Na^{+} content (Fig. 10). The greatest dispersion of the data can be noticed in type 'Red – 3'. In 'Red – 1', 'Red – 2' and 'Red – 4' types, brown and green-coloured tourmalines had similar composition, but in 'Red – 3', these versions were different, because the green ones were richer in calcium (Fig. 10). Most of the tourmalines were classified as dravite of metamorphic (low-, and medium-grade) origin. In the first two types, tourmalines only appeared with dravite compositions, but in type 'Red – 3' and 'Red – 4', schorl grains were also present (Fig. 11). Tourmalines of the Kővágószőlős Sandstone, the Maros- and the Danube Rivers together with most of the tourmalines of Mecsek pebble I ('Mecs – PebI', except for the blue ones and 'Mecs – PebIII') nicely overlap with the composition of tourmalines of both four Gorzsa tool types (Figs. 12 and 13). Sandstones of the Balaton Highlands, the Jakabhegy Sandstone and the Papuk areas can be excluded from the possible sources, as the X position of their tourmalines had more vacancies compared to torumalines of Gorzsa GSTs. In the Codru samples, a smaller proportion and other versions of tourmaline were observed, so this locality cannot be considered as a possible raw material source either. In the case of the tourmaline grains of the Mecsek pebble II ('Mecs – PebII'), the green-coloured varieties are richer in sodium compared to the brown-coloured ones (Fig. 12). This difference cannot be observed in the case of the tourmalines of the Gorzsa ground stone tools (Fig. 10), so these pebbles can also be excluded from the potential sources. Therefore, tourmaline grains were less usable indicators of the provenance of the red-coloured sandstone tools from Hódmezővásárhely-Gorzsa.

Beside the average distribution-data of the sandstone artefacts, Fig. 14 shows the representative distribution-data of the investigated geological samples of the possible raw material types. Subgroup 'Red – 1a' and 'Red – 1b' of the archaeological finds are sublitharenites and subarkoses. The composition of subgroup 'Red – 1a' does not overlap with any of the investigated possible sources (Fig. 14). In

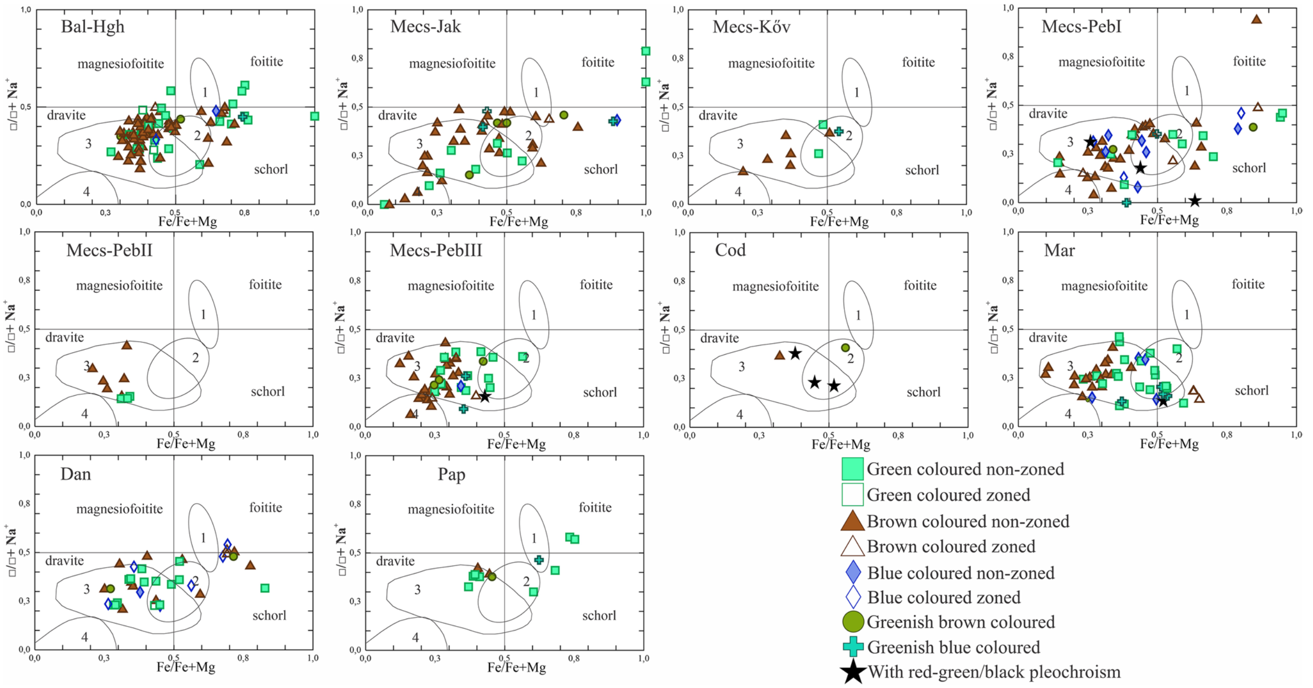

Fig. 13 The chemical composition of the tourmalines of the geological samples is represented in the $\mathrm{Fe}/(\mathrm{Fe}+\mathrm{Mg})^{Y}$ – $\square/(\square + \mathrm{Na})^{X}$ diagram. Fields marked with numbers show the typical compositional ranges of tourmalines of different metamorphic grades based on Henry and Dutrow (1996): 1 = diagenetic tourmalines; 2 = low grade tourmalines; 3 = medium grade tourmalines; 4 = high grade tourmalines. Abbreviations: *Mecs – Kőv* Kővágószőlős Sandstone, *Mecs – Jak* Jakabhegy Sandstone, *Bal – Hgh* Balaton Highlands, *Cod* Codru, *Pap* Papuk, *Mar* Maros pebble, *Mecs – PebI* Mecsek pebble Type – I, *Mecs – PebII* Mecsek pebble Type – II, *Mecs – PebIII* Mecsek pebble Type – III, *Dan* Danube pebble

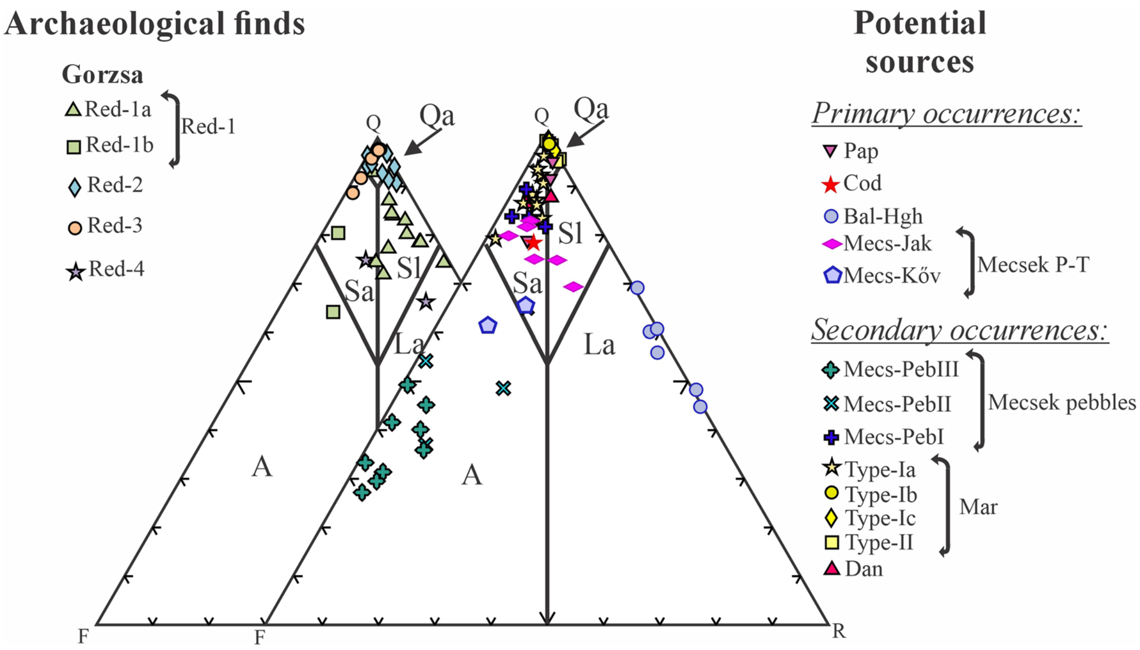

Fig. 14 Distribution of the framework grains of potential raw materials in quartz-feldspar-rock fragments – QFR triangular diagram. Abbreviations: *Q* Quartz content by using the determination procedure of Gazzi-Dickinson method, *F* Feldspar content by using the determination procedure of Gazzi-Dickinson method, *R* Rock fragment content by using the determination procedure of Gazzi-Dickinson method, *Qa* Quartz arenite, *Sa* subarkose, *Sl* Sublitharenite, *A* Arkose, *La* Litharenite, *Mecs – Kőv* Kővágószőlős Sandstone, *Mecs – Jak* Jakabhegy Sandstone, *Mecs P-T* Jakabhegy and Kővágószőlős Sandstone formations, *Bal – Hgh* Balaton Highlands, *Cod* Codru, *Pap* Papuk, *Mar* Maros pebble, *Mecs – PebI* Mecsek pebble Type – I, *Mecs – PebII* Mecsek pebble Type – II, *Mecs – PebIII* Mecsek pebble Type – III, *Dan* Danube pebble

contrast, subtype ‘Red – 1b’ of the Gorzsa tools shows a partial overlap with the Kővágószőlős and the Jakabhegy sandstones of the Mecsek Mountains, moreover with some of the Maros pebbles. Based on the presence of intermediate volcanic and granitoid rock fragments in the Permo-Triassic succession of the Mecsek Mountains, these geological formations can be excluded from the possible raw materials. Modal compositions indicate that Maros pebbles are the nearest to the ‘Red – 1’ tool type values (Fig. 14). A considerable diversity can be observed in the case of such pebbles. Its type ‘Ia’ (Fig. 14) can be considered as a probable source rock of the GSTs. In the case of the Gorzsa ‘Red – 1’ type connection with Maros pebbles could not be proven, because these stone tools do not have pebble origin (Fig. 3h). The petrographic compositions of the individual pebbles reflect the lithological diversity of the drainage area of the river. Since the control samples (pebbles) were collected from a river that contains various types of sandstones, it is not certain that all possible types or the rare ones will appear in the examined material. Due to this, it is possible that in present days (or in the past) there are pebbles in the Maros River with similar composition as the ‘Red – 1’ tool type. Thus, the pebbles of the Maros River cannot be excluded from the possible sources. In this case, it is assumed that the members of the community collected the raw material from an outcrop within the catchment area of the river or from its immediate surroundings. The general heavy mineral content of the ‘Red – 1’ tool type from Gorzsa is similar to the second character group (Group – CI), Kővágószőlős Sandstone and some of the Mecsek pebbles (‘Mecs-PebI’; Figs. 6 and 9), but significant differences can be observed based on their zircon, apatite and tourmaline content. Other potential sources can also be considered as possible raw materials, such as some of the pebbles of the Maros River, the Papuk, and the Balaton Highlands from the fourth character group (Group – CIV), but differences can be observed based on their tourmaline and apatite content (Figs. 6 and 9). Due to the numerous variations and differences of the heavy mineral variants, it is not possible to precisely determine the source of the ‘Red – 1’ tools of Gorzsa. Based on the heavy mineral composition of investigated geological samples, sandstones belonging to Group – CII and Group – CIV are the most similar sources of possible raw material. Based on the major elemental compositions of ‘Red – 1’ tool type shows similarities with the Jakabhegy and the Kővágószőlős Sandstone formations, samples of the Papuk Mountains, and with the pebbles of the Mecsek (‘Mecs – PebI’), and the Maros and the Danube Rivers (Suppl. Table 4). The minor elemental composition of the ‘Red – 1’ tool type is similar with the Jakabhegy and the Kővágószőlős Sandstone formations, the pebbles of the Mecsek (‘Mecs – PebI’ and ‘Mecs – PebIII’) and the Maros River. Based on the REE content of the ‘Red – 1’ tools, similarities were observed with the Kővágószőlős Sandstone Formation and the material of the Permo-Triassic succession of the Papuk Mountains (Fig. 15). The Th vs. La/Th ratio of the group ‘Red – 1’ show similar composition with the ‘Red – 3’ tools and with the Papuk samples, moreover with the pebbles of the Mecsek Mountains (‘Mecs – PebI’), the Maros and the Danube Rivers (Fig. 16a and c). The Th/Sc vs. Th/Cr content of the group ‘Red – 1’ show similar composition with the Kővágószőlős and Jakabhegy Sandstone formations, with the Papuk samples and the pebbles of the Danube River (Fig. 16b and d). Based on the REE

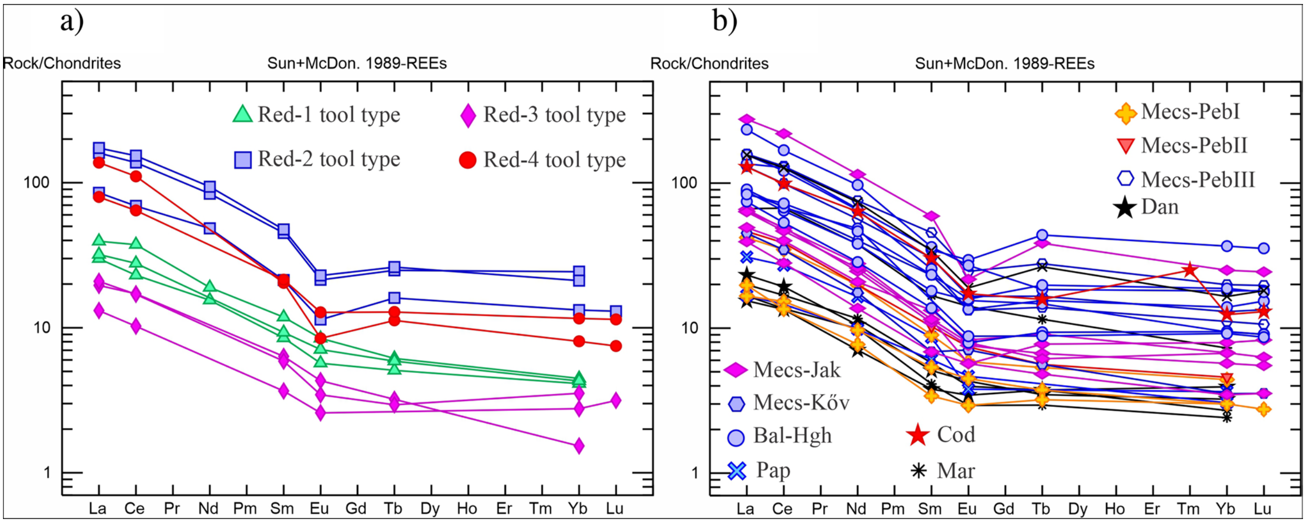

Fig. 15 Chondrite normalized rare earth element patterns. Chondrite-values by Sun and McDonough (1989). (**a**) Gorzsa artefacts, (**b**) Potential raw materials. Abbreviations: *Mecs – Kőv* Kővágószőlős Sandstone, M*ecs – Jak* Jakabhegy Sandstone, *Bal – Hgh* Balaton Highlands, *Cod* Codru, *Pap* Papuk, *Mar* Maros pebble, *Mecs – PebI* Mecsek pebble Type – I, *Mecs – PebII* Mecsek pebble Type – II, *Mecs – PebIII* Mecsek pebble Type – III, *Dan* Danube pebble

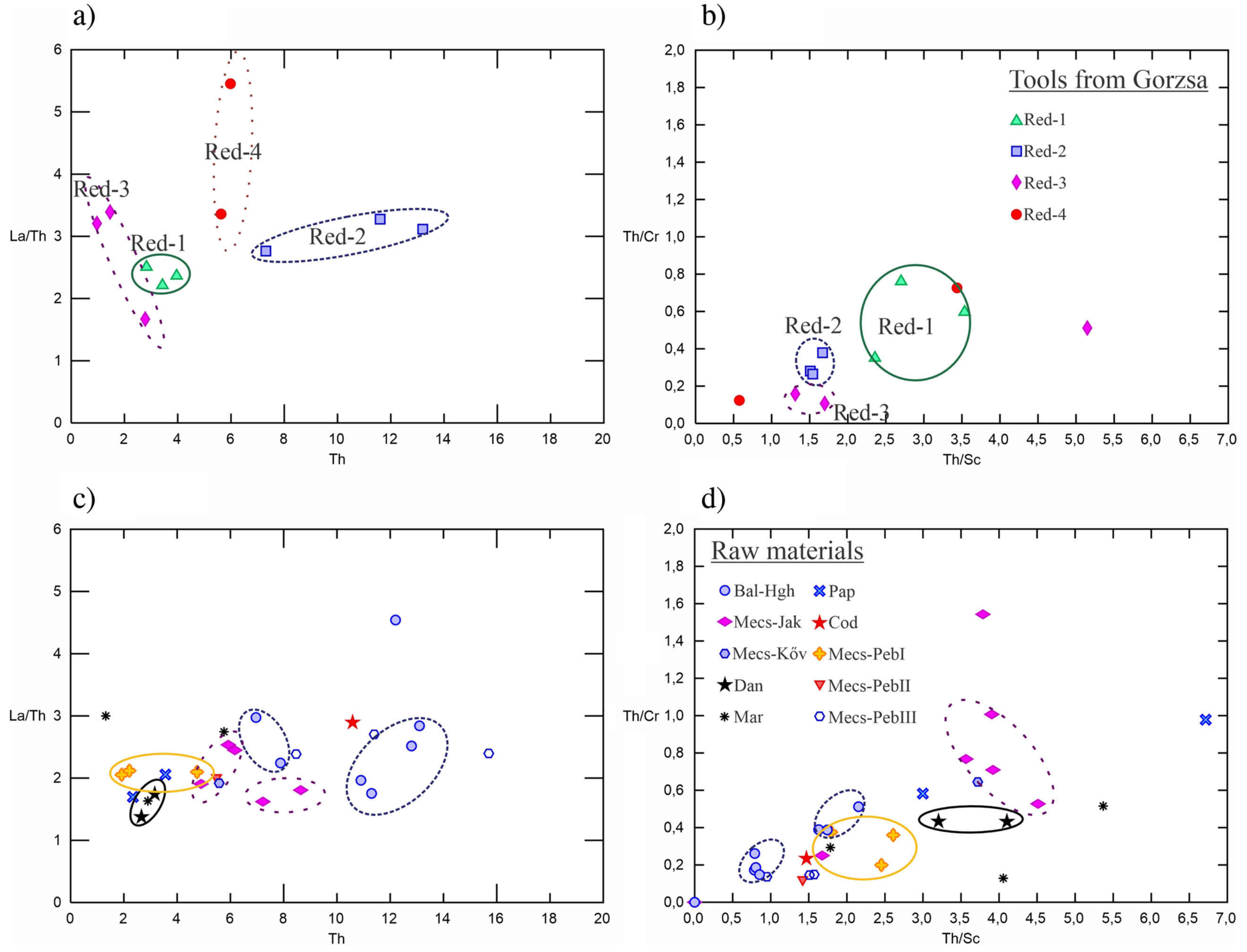

Fig. 16 (**a**) Th vs. La/Th (left) and (**b**) Th/Sc vs. Th/Cr (right) discrimination diagrams of the Gorzsa tools, (**c**) Th vs. La/Th (left one) and (**d**) Th/Sc vs. Th/Cr discrimination diagrams for possible raw materials. Abbreviations: *Mecs – Kőv* Kővágószőlős Sandstone, *Mecs – Jak* Jakabhegy Sandstone, *Bal – Hgh* Balaton Highlands, *Cod* Codru, *Pap* Papuk, *Mar* Maros pebble, *Mecs – PebI* Mecsek pebble Type – I, *Mecs – PebII* Mecsek pebble Type – II, *Mecs – PebIII* Mecsek pebble Type – III, *Dan* Danube pebble

content of the 'Red – 1' tools from Gorzsa an overlap was distinguished with the Maros and the Danube Rivers, and with the Mecsek pebbles ('Mecs – PebI'), moreover with the Papuk and probably with the Jakabhegy Sandstone samples (Fig. 17).

Group 'Red – 2' has transitional compositions between quartzarenites and sublitharenites very near to the Papuk and some pebbles of the Maros River (Fig. 14). Papuk samples have fine-medium grained, weakly, or medium sorted sandstones with high sericite (pseudomatrix) content. They have a few micas (mainly muscovite), high quartz content and some K-feldspar (Kfs > Pl, Table 3). Moreover, they have some tourmaline grains, which have brown-yellowish brown colour. These features cannot be seen in case of the group 'Red – 2' Gorzsa tools, because they are well sorted, fine grained sandstones or siltstones with high quartz quartz content and some plagioclase (Pl > Kfs) and green-coloured tourmalines. Therefore, samples of the Permo-Triassic succession of the Papuk Mountains were excluded from the possible sources. Regarding its origin, similar conclusions can be drawn as in the case of the subtype 'Red – 1b', namely it can be originated from an outcrop within the catchment area of the Maros River. The general heavy mineral composition of the 'Red – 2' tool type does not resemble any of the analysed potential source rocks. Based on the appearance of the tourmaline grains similarities can be observed with a few samples of the Maros River (Group – CIV, e.g. M-1/14 and/or M-1/18). Moreover, based on the appearance of tourmaline and apatite grains similarities can also be detected with the Codru sample (Group – CIII). Due to the numerous differences, it is not possible to determine the exact source of the 'Red – 2' tools from Gorzsa, but it might

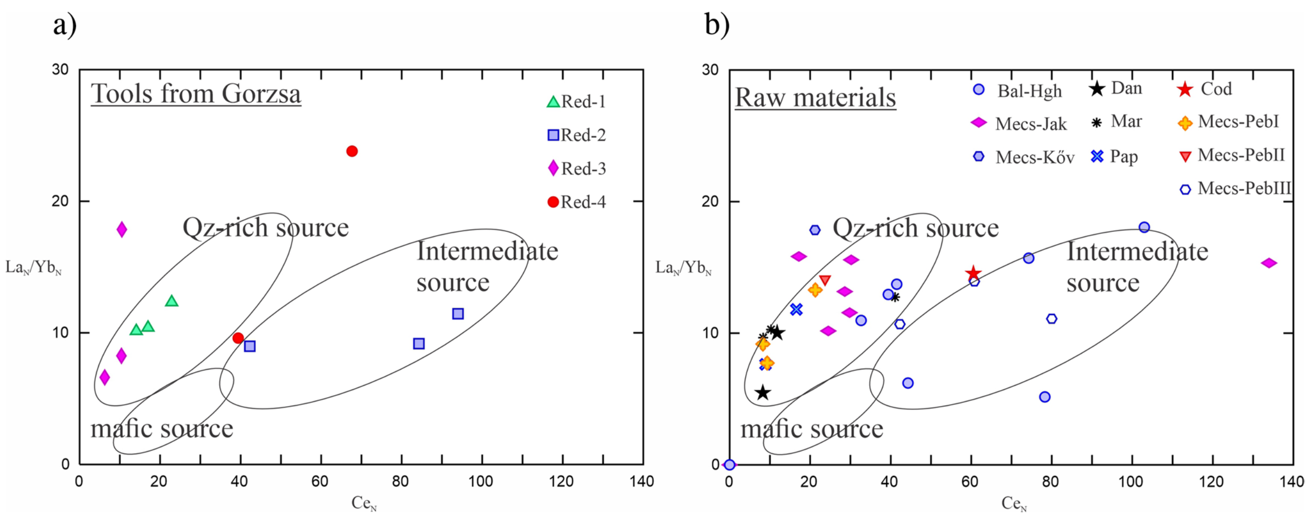

Fig. 17 Ce_N–La_N/Yb_N discrimination diagrams of the (**a**) archaeological finds and (**b**) the possible raw materials. The borders of the fields were after Alexander et al. (2000). Abbreviations: *Mecs – Kőv* Kővágószőlős Sandstone, *Mecs – Jak* Jakabhegy Sandstone, *Bal – Hgh* Balaton Highlands, *Cod* Codru, *Pap* Papuk, *Mar* Maros pebble, *Mecs – PebI* Mecsek pebble Type – I, *Mecs – PebII* Mecsek pebble Type – II, *Mecs – PebIII* Mecsek pebble Type – III, *Dan* Danube pebble

be connected to the Maros (‘Mar’) and the Transylvanian (‘Cod’) occurrences, based on the similarities of their tourmaline and general heavy mineral content (Figs. 6 and 9). The major elemental composition of ‘Red – 2’ group from Gorzsa shows similarities to the samples of the Balatonfelvidék Formation (Bal – Hgh) and pebbles of the Mecsek (Mecs – PebIII). The minor elemental content of the ‘Red – 2’ type is similar to the Balatonfelvidék Formation (‘Bal – Hgh’), however connection of ‘Red – 2’ tools with the Jakabhegy Sandstone and the pebbles of the Mecsek (‘Mecs – PebI’ and ‘Mecs – PebIII’) is also possible. Based on the REE content of the ‘Red – 2’ group from Gorzsa shows similarities with the ‘Red – 4’ tool type and with the Balatonfelvidék Formation (Bal – Hgh) and the Codru samples (Figs. 15 and 16). The Th vs. La/Th ratio of the group ‘Red – 2’ show similar composition with the Codru and the Balatonfelvidék Formation (Fig. 16a and c). The Th/Sc vs. Th/Cr content of the group ‘Red – 2’ show similar composition with the ‘Red – 3’ tools from Gorzsa and with the Balatonfelvidék Formation, Codru, Jakabhegy Sandstone Formation and with some pebbles of the Mecsek Mountains (‘Mecs – PebI’, Fig. 16b and d). Based on the REE content of the ‘Red – 2’ tool type from Gorzsa, similar compositions were identified with the Balaton Highlands and with the Mecsek pebbles (‘Mecs – PebIII’, Fig. 17).

Group ‘Red – 3’ has transitional compositions between quartzarenites and subarkoses that show an overlap with some of the Mecsek (‘Mecs – PebI’) and the Maros pebbles (Fig. 14). Pebbles group I of the Mecsek Mountains usually have similar composition (e.g. quartz and feldspar content), but they also have higher plutonic igneous rock fragment content (i.e. grains with granitoid composition), therefore they can be excluded from the potential sources. In this case, we also managed to prove the presence of tools of pebble origin (Fig. 3h). Based on the heavy mineral composition, similarities can be spotted between the ‘Red – 3’ tool type, the third- and fourth-character groups of the geological samples (Group – CIII: Codru sandstone and/or pebbles of the Danube River; Group – CIV: samples of the Papuk Mountains and the Balaton Highlands and/or pebbles of the Maros River) were determined as potential sources, although, differences could be detected in their zircon and tourmaline content (Figs. 6 and 9). Due to its high variation, it is not possible to determine the exact source of the ‘Red – 3’ tools from Gorzsa, but according to their heavy mineral content, Group – CIV seems to be the most potential source type. The major elemental composition of ‘Red – 3’ group shows similarities with ‘Red – 1’ group from Gorzsa and the pebbles from the Maros River. Based on the minor elemental compositions of the ‘Red – 3’ tool type, the same source components arise as for ‘Red – 1’ type, but the composition of the Papuk and the pebbles of the Maros River are closest to the material of this type. The REE elemental distributions of the ‘Red – 3’ group from Gorzsa shows similarities with the Papuk samples and with the Maros- and some of the Mecsek pebbles (Mecs – PebI, Figs. 15 and 16). The Th vs. La/Th ratio of the group ‘Red – 3’ show similar composition with ‘Red – 1’ tools from Gorzsa and with the pebbles of the Mecsek Mountains (‘Mecs – PebI’), the Maros and Danube Rivers and with the Permian-Triassic succession of the Papuk Mountains (Fig. 16a and c). The Th/Sc vs. Th/Cr content of the group ‘Red – 3’ show similar composition with the ‘Red – 2’ tools from Gorzsa and with the Codru and with some pebbles of the Mecsek Mountains (‘Mecs – PebII and III’, Fig. 16b and d). Based on the REE content of the ‘Red – 3’ tools from Gorzsa an overlap was distinguished with the Maros and the Danube Rivers, and with the Mecsek pebbles (‘Mecs – PebI’), moreover with the Papuk and probably with the Jakabhegy Sandstone samples, but in case of this archaeological group the most probable source is the Maros pebbles (Fig. 17).

Samples of the ‘Red – 4’ group from Gorzsa have transitional compositions between subarkoses and lithic arenites (i.e. litharenites) very near to the Codru, Papuk and the Jakabhegy Sandstone Formation (Fig. 14). This archaeological tool type contains a lot of metamorphic rock fragments, which is not presented in any of the investigated geological samples. Therefore, all of these geological, red-coloured sandstone occurrences can be excluded from the possible sources. Based on the petrographic observations, none of the investigated geological sandstone samples show similar compositions to ‘Red – 4’ tool type. Based on the heavy mineral content of the ‘Red – 4’ tool type does not match with any of the investigated geological sources, because this group has a very special, garnet dominated composition that is not an ordinary mineral phase in the case of the red-coloured sandstone occurrences of the Pannonian Basin. Based on the major elemental composition of ‘Red – 4’ tools the same source components were identified as for ‘Red – 2’ group, but in terms of composition the pebbles of the Mecsek (‘Mecs – PebII’) and the Codru samples can also be considered as possible raw materials. The minor elemental composition of ‘Red – 4’ tools, the same possible sources arise as for ‘Red – 2’ type, but in addition, the Jakabhegy, the Kővágószőlős and the Balatonfelvidék Sandstone formations together with the pebbles of the Mecsek (‘Mecs – PebII’ and ‘Mecs – PebIII’) and the Danube River can also be considered as possible sources. Based on the REE content of the ‘Red – 4’ group from Gorzsa shows similarities with the ‘Red – 2’ tool type and with the Balatonfelvidék Formation (Bal – Hgh) and the Codru samples (Figs. 15 and 16). The Th vs. La/Th ratio of the group ‘Red – 4’ show similar composition with some samples of the Balatonfelvidék Formation (Fig. 16a and c). The Th/Sc vs. Th/Cr content of the group ‘Red – 4’ show similar composition with some of the ‘Red – 1’ tools from Gorzsa and with the Jakabhegy and Balatonfelvidék formations (Fig. 16b and d). Based on the REE content of the ‘Red – 4’ tool type from Gorzsa, similar compositions were identified with the Balaton Highlands and with the Mecsek pebbles (‘Mecs – PebIII’, Fig. 17).

Conclusion

To locate the possible origin of the sandstone tools found at Gorzsa tell, a total of 109 fragments of red-coloured ground stone tools and 124 comparative geological samples from the Carpathian-Pannonian Region were investigated. Multiple analytical methods, principally petrographic and mineral chemical analysis of the heavy mineral species were applied as these are regarded as the most accurate provenance indicators.

1. Four raw material types of the Gorzsa red-coloured sandstone ground stone tools can be distinguished by the petrographic and geochemical investigations.
2. Similar heavy mineral composition can be identified in the case of the ‘Red – 1’, ‘Red – 2’ and ‘Red – 3’ types. Their main components are zircon, tourmaline, rutile, titanite and occasionally apatite (mostly in ‘Red – 1’). The main difference between them is the relative abundances of these mineral phases.
3. Type ‘Red – 4’ has a unique heavy mineral composition: high amount of garnet and lower amount of apatite, epidote, zircon, rutile and tourmaline.
4. The bulk-rock geochemical data shows a stronger relationship between ‘Red – 1’ and ‘Red – 3’, as well as the ‘Red – 2’ and ‘Red – 4’ tool types.
5. The comparison of sandstone tools and possible raw materials by petrography, whole-rock- and mineral chemistry reveals that the majority of the Gorzsa tools has similar compositions to the pebbles of the geographically close Maros River.
6. In the case of the Gorzsa ‘Red – 3’ type a strong connection with the pebbles of the Maros River could be proved.
7. In the case of the other red sandstone tool types (‘Red – 1’, ‘Red – 2’, and ‘Red – 4’), Maros source rocks (‘Type – Ia’) has highly similar petrographic and geochemical character, also considering that the Maros river is the closest geographical location of the tell site. However, none of the tools of these red sandstone types (‘Red – 1’, ‘Red – 2’, and ‘Red – 4’) has pebble origin. Therefore, it is assumed that the probable raw material source of these finds can be collected from an outcrop within the catchment area of the Maros River or from its immediate surroundings in the Apuseni Mountains (Romania).
8. Based on this recent investigation, other previously proposed raw materials (Piros 2010), such as the Jakabhegy Sandstone, the Papuk, the pebbles of the Danube River or the Mecsek Mountains could be excluded from the possible sources.

To clarify these results, additional possible raw material samples from the Maros River and the Apuseni Mts. (Transylvania, Romania) need to be investigated by petrographic, mineral- and geochemical methods.

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Declarations

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