RESULTS OF NON-DESTRUCTIVE SEM-EDX AND PGAA ANALYSES OF JADE AND ECLOGITE POLISHED STONE TOOLS IN HUNGARY

MAGYARORSZÁGI JADE ÉS EKLOGIT NYERSANYAGÚ CSISZOLT KŐESZKÖZÖK RONCSOLÁSMENTES VIZSGÁLATÁNAK EREDMÉNYEI

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Abstract

Good quality high pressure (HP) metaophiolites suitable for making stone implements, like jade and eclogite, are geologically absent in the Carpathian Basin and its surroundings. Therefore this raw material type was unknown among Hungarian findings for a long time, and henceforward this is one of the rarest types of raw material of polished stone implements in Hungary. Their investigation is very important because of their scarcity and distant origin. The nearest geological locality where these raw materials can be found is over 1000 kms away. The specific form and integrity of these stone implements indicate that they were transported as complete (finished) artefacts.

*Petrological investigations of large collections (Miháldy and Ebenhöch collections) revealed their presence among Hungarian findings (Friedel et al. 2008, Szakmány et al. 2013). In 2013 and 2014, several new pieces of HP stone implements were found in museums of Hungary, most of them from known archaeological context. Most samples were found in Transdanubia, only one piece turned up in Eastern Hungary (**Fig. 3.**). The 7 pieces of known archaeological context are from four localities (Zengővárkony (3), Szombathely Oladi plató (2), Alsónyék (1), Gorzsa (1)). Other pieces are stray finds. The localities and the main features of the tools are summarized in **Table 1**. In addition to the 3 pieces presented in Szakmány et al. 2013, 10 pieces of stone implements made of HP metaophiolites are presented in this work (**Fig. 1.**).*

Only non-destructive analytical methods were used in this study. Stone implements were divided into raw material type groups based on their mineral chemical and bulk rock analytical data. Our results are corresponding to results on HP metaophiolites of North-western Italy, obtained both on geological and archaeological samples (D'Amico et al. 2003, D'Amico 2012, Pétrequin et al. 2012). Based upon these facts, the HP metaophiolite stone implements in Hungary probably originated from the same raw material sources as Italian (and other Western European) HP metaophiolite stone tools. According to technical literature, these primary sources can be the Monviso, the Voltri Massif and secondary in the resedimented Oligocene conglomerates in Quaternary of River Po, Staffora and Curone equally (D'Amico et al. 2003, D'Amico & Starnini 2006, 2012, Pétrequin et al. 2012).

Kivonat

Jelenlegi ismereteink szerint a nagynyomású (HP) metaofiolit nyersanyagú eszközök igen csekély számban találhatók meg a magyarországi csiszolt kőeszközök leletegyütteseiben. Vizsgálatuk azért kiemelkedő fontosságú, mert nyersanyaguk a Kárpát-medencében, és annak közvetlen környezetében nem található meg. Legközelebbi, a neolitikumban bizonyítottan felhasznált nyersanyagforrásuk Magyarország jelenlegi területétől több, mint 1000 km távolságban található, ami, az eszközök formavilágát is figyelembe véve, arra utal, hogy ezek az eszközök kereskedelmi útvonalakon, használatra kész termékként érkeztek régészeti lelőhelyükre.

*Magyarországi jelenlétükre a Miháldy- és az Ebenhöch-gyűjtemények kőzettani szempontú feldolgozása során derült fény (Friedel et al. 2008, Szakmány et al. 2013). 2013-ban és 2014-ben további leletek kerültek elő különböző múzeumok gyűjteményeiből, a JADE2 program keretében végzett szisztematikus kutatások eredményeképpen. A leletek legnagyobb része a Dunántúlról került elő, mindössze egy darab származik az ország keleti részéből (**3. ábra**). Hét darab kőeszköz került elő részletesen dokumentált régészeti ásatásból (Zengővárkony (3), Szombathely Oladi plató (2), Alsónyék (1), Gorzsa (1), a többi szórványlelet. Lelőhelyeik és*

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legfontosabb adataik megtalálhatók az ***1. táblázat****ban. Ebben a munkánkban a Szakmány és munkatársai (2013) cikkben ismertetett 3 kőeszközön felül 10 további HP metaofiolit nyersanyagú kőeszközt mutatunk be részletesen (****1. ábra****).*

Munkánk során kizárólag roncsolásmentes analitikai módszereket használtunk. Eredményeink alapján kőzetszöveti, ásvány- és teljes kőzet kémiai adatok segítségével a nemzetközi archeometriai szakirodalomban található felosztás alapján csoportosítjuk a leleteket. Eredményeink jól egyeznek az Északnyugat Olaszország területéről leírt kőeszközök adataival (D'Amico et al. 2003, D'Amico 2012, Pétrequin et al. 2012), így az általunk vizsgált kőeszközök nyersanyagának forrásterülete feltehetőleg megegyezik azokéval. Ezek elsődleges előfordulása a Monviso, Voltri ill. az ÉNy-i Appenninek területe és másodlagosan az ezekről a területekről lepusztult, majd a negyedidőszakban áthalmozódott és a Pó síkságon, valamint a Curone és a Staffora folyók környezetében lerakódott konglomerátumok kavicsanyaga (D'Amico et al. 2003, D'Amico & Starnini 2006, 2012, Pétrequin et al. 2012).

KEYWORDS: POLISHED STONE TOOL, JADE, ECLOGITE, PROVENANCE, NON-DESTRUCTIVE ANALYSIS

KULCSSZAVAK: CSISZOLT KŐESZKÖZ, JADEITIT, EKLOGIT, NYERSANYAG FORRÁSTERÜLET, RONCSOLÁSMENTES ANYAGVIZSGÁLAT

Fig. 1.:

Stone implements from Hungary made of HP metaophiolite raw material. Most important form types can be seen on the picture.

1. ábra:

Néhány magyarországi HP metaofiolit anyagú kőeszköz. A képen az összes fontosabb alaki típus látható.

Introduction

Green or greenish colour is very popular among the prehistoric polished stone tools. This colour is characteristic for several different rock types, e.g. greenschist, contact metabasite, nephrite and serpentinite; however HP metaophiolites are outstanding among the 'greenstones' both by beauty and quality (**Fig. 1.**). Their elaboration is unique, in most of cases they have flat, elongated, lingular or triangular shape and they are of different shades of green and their surface is especially finely polished. These types of stone implements are very widespread all over Western Europe (D'Amico 2003, Pétrequin et al. 2011, Klassen 2012, Domínguez-Bella et al. 2015). Their penetration decreasing toward east, the easternmost known localities are in Bulgaria, however, they were practically unknown in the Carpathian Basin until the recent past (Pétrequin et al. 2011, Szakmány et al. 2013).

Their importance is derived from their scarcity and distant origin (**Fig. 2.**). Their first recognition is connected with the study of major polished stone tool assemblages: Friedel et al. (2008) mentioned six pieces of HP metaophiolite stone implements from Ebenhöch collection without detailed description. Szakmány et al. (2013) gave detailed information on three pieces of stone implements made of HP raw material. These pieces are from the collections accumulated by F. Ebenhöch and I. Miháldy in the 19th century. Because of the afore mentioned article is in Hungarian, these important artefacts will be presented in this work again (see below) for a wider audience.

Fig. 2.: Raw material sources of HP metaophiolites. 1 – Primary sources; 2 – secondary/tertiary sources (Oligocene conglomerates and their redeposited sediments). After D'Amico & Starnini 2012.

2. ábra: A HP metaofiolit nyersanyagok előfordulási területei. 1 – elsődleges előfordulások; 2 – másodlagos/harmadlagos előfordulások (Oligocén konglomerátumokban és azok áthalmozott üledékeiben). D'Amico & Starnini (2012) alapján.

General characterisation of jade and eclogite raw material

HP metaophiolites can be divided into two groups based upon the garnet content of these rocks. The most widespread group (in both area and mass/amount) is the eclogite group, which contains large amount of garnets beyond the alkaline pyroxenes (at least 5 % according to D'Amico 2003). Among the alkaline pyroxenes, omphacite is much more abundant than the other Na-pyroxene types, however jadeite dominated eclogites do exist as well. The group of 'alkaline-pyroxenites' or 'jades' is much smaller. Dominant mineral phases are the different alkaline-pyroxenes in this group, like jadeite and omphacite, sometimes aegirine and/or aegirine-augite. The quantity of alkaline-pyroxenes is more than 80% in these samples. In addition to the main phases, both groups contain some accessory minerals in these samples, e.g. rutile, zircon, ilmenite, apatite and titanite.

These two groups are formed under the same conditions, by high pressure (2-3 GPa) and low to medium temperature (500-600 °C) metamorphism from basaltic protoliths. Based upon their mineral and chemical composition, the HP metaophiolite raw material of these stone implements were formed by the younger, Alpean stage of orogenesis, during the Eocene epoch (Compagnoni 2007; Beltrando et al. 2010).

The Alpine type HP metaophiolites suitable implements for raw material can be found both in primary, secondary (Oligocene) and tertiary (recent) positions in NW-Italy. This latter were redeposited in the Quaternary period, and Prehistoric people used this as raw material source (D'Amico & Starnini 2006). Primary occurrences can be found in the eastern range of the Western Alps, from Monviso till the Aosta valley, and in the Voltri Massif at the north-western end of Apennines (Monte Beigua, **Fig. 2.**). The formation of these units occurred after the subduction of the floor of Tethys-ocean. Both areas were uplifted in the Alpean orogenic stage, after the high pressure metamorphism (Compagnoni et al. 2007). Secondary raw material sources can be found at the nearby piedmonts of the Western Alps and Voltri Massif and in the Po plain. They were formed during Oligocene by sedimentation of eroded materials from the Western Alps and Voltri Massif. These types of raw material sources were described from recent sediments of Staffora and Curone rivers

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(D'Amico & Starnini, 2012), and at the upper Po river, which originates from the Monviso region (Pétrequin et al. 2012) (**Fig. 2.**).

Material quality played an important role in the usage and spreading of these stone implements. Stone tools made of eclogite were used as wood-working tools by the local prehistoric people, however exceptionally shaped and finished stone implements made of jade can be found all over Europe. They are interpreted by the archaeologists as symbols of power and/or religion, and/or wealth rather than common work tools. The selection of raw material was really careful, it is indicated by the distribution of the quantity, the quality and the range of these stone implements. 'Jade' as raw material can be found at very limited area and in relatively small quantities, however, stone implements made of this raw material can be found more than 1500 km away from the source areas. On the other hand, the much more abundant eclogites were mostly locally (regionally) used as raw material for work tools (D'Amico 2005, D'Amico & Starnini 2012, Pétrequin et al. 2011).

Archaeological background

The analysed artefacts are from various localities within Hungary, some of them from archaeological excavations of prehistoric sites, others from recent field surveys or surface collections as stray finds in the old collection of various museums (**Table 1., Fig. 3.**). Their description here follows roughly the temporal order of their recognition among the polished stone artefacts in Hungary (Szakmány 2009, Szakmány et al. 2013).

Miháldy collection

The collection accumulated by István Miháldy (1833-1901) contains 378 polished stone tools, now curated in the Laczkó Dezső Museum, Veszprém. Unfortunately, the information on the exact provenance of the pieces was lost during re-inventorisation in the 1950-ies. We know, however, that all the samples were found at different sites in the Bakony Mts. and its surroundings (Horváth, T. 2001, Füri et al. 2004). One piece of HP metaophiolite artefact was found in this collection (sample 55.1276).

Ebenhöch collection

The collection was accumulated by Ferenc Ebenhöch (1821-1889), prebend of Győr, NE Hungary. His collection, partly donated, partly sold to the Hungarian National Museum contains more than 700 polished stone tools and it is now part of the Prehistoric Collection of the HNM, Budapest.

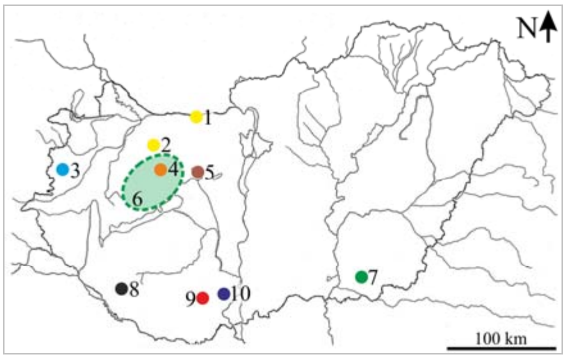

Fig. 3.: Archaeological localities of presented artefacts. 1 – Neszmély, 2 – Bakonypéterd, 3 – Szombathely, Olad-plateau, 4 – Zirc, 5 – Iszkaszentgyörgy, 6 – Bakony Mts., 7 – Hódmezővásárhely-Gorzsa, 8 – Lábod, 9 – Zengővárkony, 10 – Alsónyék.

3. ábra: A bemutatott kőeszközök régészeti lelőhelyei. 1 – Neszmély, 2 – Bakonypéterd, 3 – Szombathely, Oladi-plató, 4 – Zirc, 5 – Iszkaszentgyörgy, 6 – Bakony, 7 – Hódmezővásárhely-Gorzsa, 8 – Lábod, 9 – Zengővárkony, 10 – Alsónyék.

The artefacts were found in Western and Upper Hungary (latter is today Slovakia). The polished stone tools of this collection were studied in details by O. Friedel (Friedel et al. 2011). Two pieces of HP metaophiolite stone tools were investigated from this collection (samples 300/1876.247 and 300/1876.264). The localities involved are Bakonypéterd and Neszmély with no further archaeological context.

Gorzsa

Hódmezővásárhely-Gorzsa is a famous Late Neolithic (Tisza culture) tell settlement (Horváth, F. 2003, 2005). The site lies at the confluence of the Tisza and Maros rivers in the Great Hungarian Plain, north of Szeged. Approximately 1,000 square meters of the tell settlement were excavated and more than 1000 stone artefacts were collected from controlled stratigraphical context (Szakmány et al. 2008; Starnini et al 2015). One HP metaophiolite stone implement was identified in the rich the lithic assemblage, this is the easternmost example of HP stone tools in Hungary so far (sample 99.3.1863).

Szombathely (Olad plateau)

Olad plateau was a Late Neolithic settlement of the early Lengyel culture between the early and the classical periods (Lengyel Ib, Ilon 2011). Two very small fragments of HP metaophiolite stone implements were found in this site (samples Olad-321 and Olad-329).

Table 1.: Physical properties and basic petrographic description of the investigated HP metaophiolite samples. Abbreviations: LDM, Vp. – Laczkó Dezső Museum, Veszprém, HNM, Bp. – Hungarian National Museum, Budapest, RRM – Rippl-Rónai Museum, EW – private collection of Ernő Wolf, WMM – Wosinsky Mór Museum, JPM – Janus Pannonius Museum, SM – Savaria Museum, MFM – Móra Ferenc Museum, coll. – collection, ND – not determined.

1. táblázat: A vizsgált HP metaofiolit anyagú kőeszközök fizikai tulajdonságai és alapszintű kőzettani leírásai. Rövidítések: LDM, Vp. – Laczkó Dezső Múzeum, Veszprém, HNM, Bp. – Magyar Nemzeti Múzeum, Budapest, RRM – Rippl-Rónai Múzeum, EW – Wolf Ernő magángyűjteménye, WMM – Wosinsky Mór Múzeum, JPM – Janus Pannonius Múzeum, SM – Savaria Múzeum, MFM – Móra Ferenc Múzeum, coll. – gyűjtemény, ND – nem mért.

	Locality	Museum (collection)	Inv. number	L (mm)	W (mm)	H (mm)	MS ($x10^{-3}$ SI)	shape	colour	archaeological context	macroscopic description
1	Bakony	LDM, (Mihåldy collection)	55.1276	95	40	15	0,17	flat, elongated, triangular adze	medium to dark green	stray find	homogenous, lighter green veins and patches on the darker green background, tiny (smaller than 1 mm), white subhedral-anhedral crystals
2	Neszmély	HNM, (Ebenhöch collection)	300/1876.247	40	28	7	0,47	flat adze	dark green	stray find	homogenous, pale green, greyish green, grey and locally very dark green patches and veins
3	Bakonypéterd	HNM, (Ebenhöch collection)	300/1876.264	56	25	8	0,10	flat, elongated, lingular adze	pale green	stray find	homogenous, medium to dark green patches, locally rounded grains with darker and brighter shade than the matrix
4	Iszkaszent-györgy	HNM, (Prehistoric coll.)	39/1903	118	50	18	0,32	flat, strongly elongated, lingular adze	very dark green	stray find	homogenous, medium ét dark green patches, lot of relatively large rutile (larger than 1 mm), pale green-greenish white elongated crystals (jd?) locally
5	Lábod	RRM, Kaposvár	3127	95	50	25	ND	**thick,** elongated, lingular adze	dark green	stray find	slightly inhomogenous, dark and medium green patches
6	Zirc	EW, Zirc	81/W2 5	36	29	10	0,19	flat, slightly elongated, triangular adze	pale green	stray find	slightly inhomogenous with brigther green patches
7	Alsónyék	WMM, Szekszárd	M6.2010.10 B.3060.3	116	50	22	0,95	thick, strongly elongated triangular adze	greenish black	Lengyel culture	roughly homogenous, sheared, bands of different shades of green changing, large (max. 2 mms) brown and green grains locally
8	Zengővárkon y	JPM, Pécs	N.5/47-1939	40	30	7	0,19	flat, slightly elongated, triangular adze	bluish black	Lengyel culture	roughly homogenous, slightly foliated, tiny, white subhedral-anhedral crystals, crystalagglomerates and green veins
9	Zengővárkon y	JPM, Pécs	N.1/81-1938	62	39	13	0,20	flat, elongated, triangular adze	medium green	Lengyel culture	roughly homogenous, patches with differenet sheds of green, some large (max. 5 mms) white patches, 1-2 mm long dark green subhedral-euhedral crystals locally
10	Zengővárkon y	JPM, Pécs	N.11/169-1938	58	31	12	0,39	flat, strongly elongated, lingular adze	dark green	Lengyel culture	homogenous, medium and dark green patched, max. 2 mm long rutile crystals
11	Szombathely, Olad	SM, Szombathely	Olad-321	18	20	6	0,03	fragment of flat axehead	medium green and red	Lengyel culture	roughly homogenous, red grains in medium green matrix
12	Szombathely, Olad	SM, Szombathely	Olad-329	20	27	13	0,03	fragment of flat axehead	medium green	Lengyel culture	roughly homogenous, pale and medium green patches, tiny, locally white crystals
13	Hódmező-vásárhely, Gorzsa	MFM, Szeged	99.3.1863	40	25	9	0,24	flat, slightly elongated, triangular adze	dark green	Tisza culture	very dark green veins and patches in the dark green matrix, some (4-5) garnets with 1 mm of max diameter, few, elongated (max. 1 mm long) white crystals (zrn or ap)

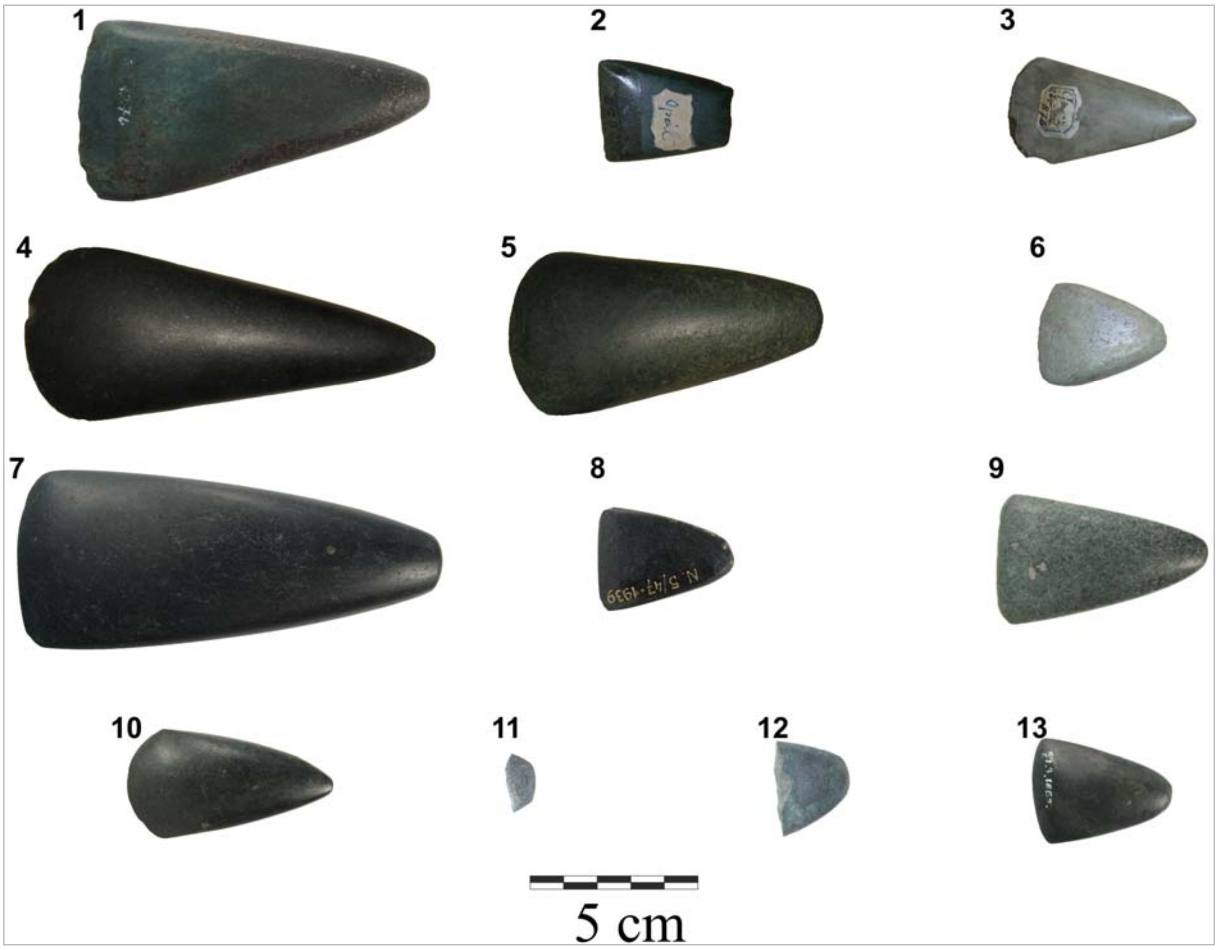

Fig. 4.: Artefacts presented in this work. Numbers are referring to first column on **Table 1**.

4. ábra: Az összes bemutatott kőeszköz. A sorszámok az **1. táblázat** 1. oszlopára vonatkoznak.

Zengővárkony

Classical settlement and cemetery of the Lengyel Culture, excavated and published by J. Dombay (Dombay 1939, 1960). The polished stone artefacts were investigated previously (Biró et al. 2003) and reconsidered for sourcing the HP metamorphites. Three HP metaophiolite artefacts were identified, all of them found in graves (samples N.1/81-1938, N.11/169-1938 and N.5/47-1939).

Alsónyék

The largest known cemetery of the Lengyel Culture, excavated in course of preventive rescue excavations. The elaboration of the site is in progress. One HP metaophiolite artefact was found in a princely grave (Grave 3060, sample M6.2010.10B.3060.3), published separately (Zalai-Gaál et al. 2011).

In addition, three stray finds were investigated, their localities: Iszkaszentgyörgy, Fejér county (HNM 39/1903); Lábod (3127), RMM Somogy county; Zirc (81/W2.5), private collection, Veszprém county.

All the objects with documented archaeological context are from the Late Neolithic. Most of them belong to Lengyel culture (Zengővárkony, Olad-plateau and Alsónyék). The object from Gorzsa is assigned to the Tisza culture, coeval with the Lengyel culture.

Methods

The stone implements made of jade and related rocks are generally intact, therefore only non-destructive methods can be used for their investigation. These non-destructive methods included macroscopic analyses, stereomicroscopy, magnetic susceptibility measurement, geochemical analysis by prompt gamma activation analysis, non-destructive X-ray diffractometry (performed at Miskolc University and presented in the paper by Kristály (2014) in this volume) and original surface analysis, explained in details below.

At the same time, spectroradiometrical analyses (Errera et al. 2007) were performed on the same items by M. Errera that will be reported on elsewhere.

Loupe and stereomicroscope were used for the basic petrographic description. Magnetic susceptibility (MS) measurements were made with portable Kappameter KT-5. Real MS values were calculated by different corrections of size and thickness (Bradák et al. 2005, 2009).

All of the BSE images and EDX analyses were made by non-destructive original surface analysis method (Bendő et al. 2013) that was developed for textural and mineral chemistry analysis of stone tools. Besides the standards listed in Bendő et al. 2013, jadeite (SPI #AS1195-AB), omphacite (Smithsonian Microbeam Standard, SMS, NMNH 110607), garnet (SMS, NMNH 87375) and pyrope (SMS, NMNH 143968) mineral standards were used. These measurements were made at the Department of Petrology and Geochemistry, Institute of Geography and Earth Sciences, Eötvös Loránd University, Budapest. The instrument was an AMRAY 1830 scanning electron microscope equipped with EDAX PV9800 energy dispersive spectrometer. Conditions of analysis: accelerating potential: 20 kV, beam current: 1 nA, beam diameter: focused electron beam (~50-100 nm), measurement time: 100 sec (livetime).

Non-destructive PGAA was performed at the Budapest Research Reactor operated by the Centre for Energy Research, Hungarian Academy of Sciences. The method is suitable for the quantitative determination of the average concentrations of the major elements (SiO_2, TiO_2, Al_2O_3, $Fe_2O_3^t$, MnO, MgO, CaO, Na_2O, K_2O and H_2O) and some trace elements (e.g. B, Cl, Sm, Gd) in a few cm^3 volume. Thermal equivalent beam intensity was 7.75×10^7 $cm^{-2}\times s^{-1}$. Calibrated Compton-suppressed HPGe detector was used to detect prompt gamma spectra, and Hypermet PC software was used for evaluation (Révay, 2009, Szentmiklósi et al. 2010, Szakmány et al. 2011).

Results

Physical properties and MS values of the stone implements are summarized in Table 1. with short macroscopic description of the samples. According to our results, the objects can be divided into six raw material groups. The first four groups are the 'Na-pyroxenites', those HP metaophiolite raw materials which do not contain significant amounts of garnet (less than 5 %). These groups are listed by increasing iron content. The fifth group contains the only glaucophane schist sample, and the last group comprises an iron eclogite specimen (**Fig. 4.**). Nomenclature diagram of alkaline pyroxenes (Morimoto et al. 1988) was used for the present mineral chemical grouping of samples. Garnets are presented also on a triangular diagram.

PGAA results are presented on multi-element diagrams normalized to Upper Continental Crust (UCC) data by McLennan (2001) for easier comparison. Data fields of Italian HP metaophiolite stone implements by D'Amico et al. (2003) are also presented on these diagrams for comparison.

Jadeitites

Two pieces of jadeitite stone implements were found until this time, both of them located in large old museum collections, one is from the Ebenhöch collection (300/1876.264), the other one is from the Miháldy collection (55.1276).

In respect of mineral composition, their raw material contains much more jadeite than omphacite (**Fig. 5.**). Both samples contain zircon and allanite as accessory minerals. Titanite and xenotime can also be found in them. The Na-pyroxenes are generally zoned, they have jadeite core and omphacite rim (**Fig. 6., Fig. 7.**). Deformation textures are missing in these samples.

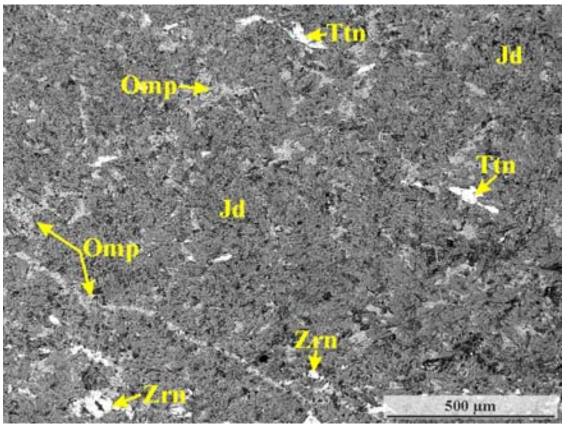

Fig. 5.: Typical texture of jadeitites: small amount of omphacite is scattered among jadeite. According to the textural characteristics, omphacite was formed in the latest stage of metamorphism in this sample (300/1876.264, Bakonypéterd).

5. ábra: Tipikus jadeitit szövet: kevés omfacit helyezkedik el szétszórva a jadeit kristályok között. Szöveti jellegzetességei alapján az omfacit a metamorfózis legutolsó fázisában keletkezett ebben a mintában (300/1876.264, Bakonypéterd).

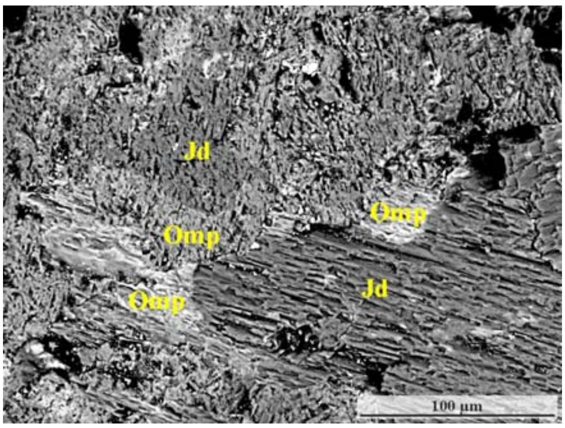

Fig. 6.: Zoned pyroxenes with jadeite core and omphacite rim. Sample 55.1276 (Bakony Mts.).

6. ábra: Zónás piroxénkristályok jadeit maggal és omfacit peremmel az 55.1276-os mintában (Bakony).

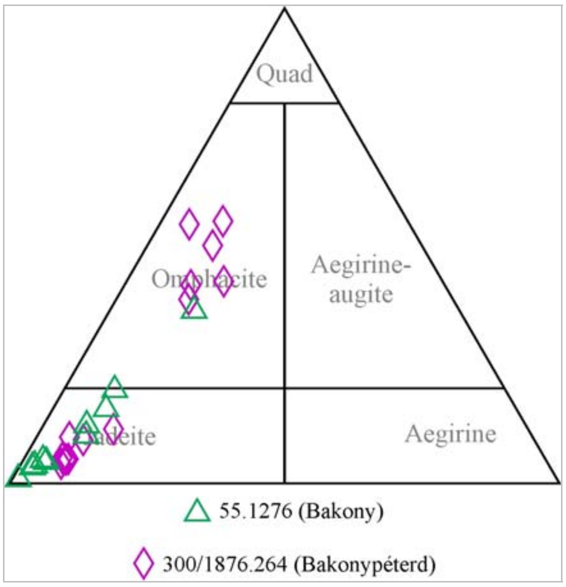

Fig. 7.: Pyroxene compositions in the jadeitite samples. Discrete compositions of jadeite and omphacite are very well visible.

7. ábra: A jadeitit minták piroxénjeinek összetétele. A jadeit és omfacit mezőkben elhelyezkedő összetételek határozott elkülönülést mutatnak.

Jadeitic pyroxenes are similar in composition in these two samples, but the omphacitic pyroxenes of sample 300/1876.264 contain much more omphacite than pyroxenes of sample 55.1276. Jadeitic pyroxenes appear along a steep trend line toward omphacitic pyroxenes but the continuity of this trend is doubtful based on these data (**Fig. 7.**).

Bulk-rock data are corresponding very well to data of D'Amico et al. (2003, grey field). According to these results these samples have fairly different calcium and magnesium content. This difference was probably caused by the higher omphacite content of sample 300/1876.264 (**Fig. 8.**).

Mixed jades

This jade group contains two pieces of stone tools. One of them was found at the excavations of Zengővárkony (N.1/81-1938), from documented archaeological context. The other one is a stray find from Zirc (81/W2.5).

Their raw material contains more jadeite than omphacite or aegirine-augite (**Fig. 9.**). Both samples contain rutile as accessory mineral. Zircon, allanite, titanite, xenotime and monazite can also be found in them. Retromorphic phases (albite, epidote) can be found in sample N.1/81-1938.

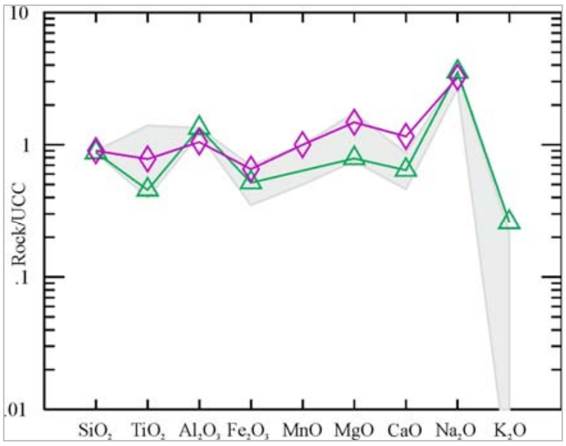

Fig. 8.: Bulk-rock compositions of jadeitite samples normalized to UCC by McLennan (2001). Results showing very well the correspondence to jadeitite compositions by D'Amico et al (2003). Differences in compositions are possibly caused by varieties in omphacite content of the samples.

8. ábra: A jadeitit minták teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). Az adatok nagyon jó egyezést mutatnak D'Amico et al. (2003) jadeitit adataival. A két minta közötti összetétel különbséget valószínűleg a minták különböző omfacit-tartalma okozza.

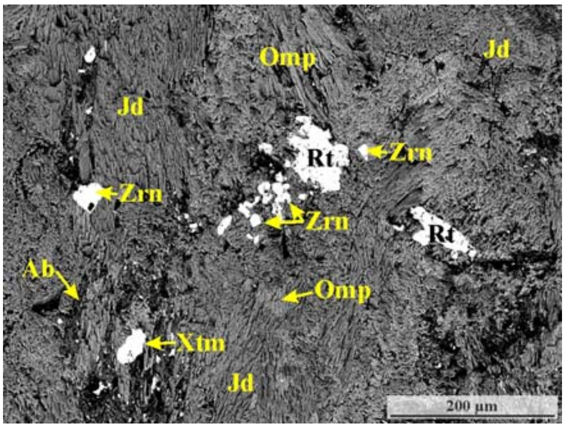

Fig. 9.: Texture of mixed jades with accessory and retromorphic minerals. Sample N.1/81-1938 from Zengővárkony.

9. ábra: Kevert jadeitit szövete, akcesszórikus és retromorf ásványai. N.1/81-1938 minta Zengővárkonyról.

Na-pyroxenes are frequently zoned, they have jadeite core and omphacite/aegirine augite rim (**Fig. 10**, **Fig. 11.**). Na-pyroxenes in sample 81/W2.5 have a 'radial' texture (**Fig. 10.**). Deformation structures are not present in these samples.

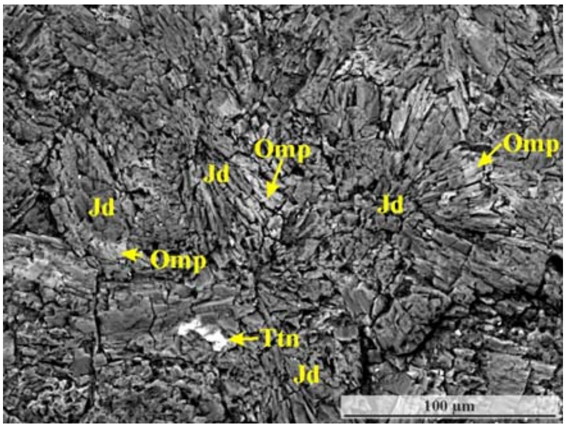

Fig. 10.: 'Radial' texture of alkaline pyroxenes in sample 81/W2.5 (Zirc). Pyroxenes shows 'normal' zonation, with jadeite core and omphacite rim.

10. ábra: Az alkáli piroxének „sugaras" szövete a 81/W2.5 mintában (Zirc). A piroxének normál zónásságot mutatnak, jadeit maggal és omfacit peremmel.

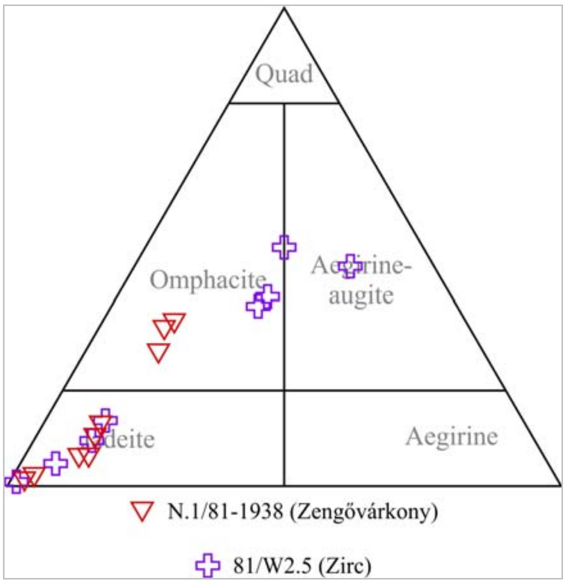

Fig. 11.: Pyroxene compositions in mixed jades. Jadeites have very similar composition in both samples, however omphacite compositions are rather different, omphacites of sample 81/W2.5 are richer in iron than omphacites of sample N.1/81-1938.

11. ábra: Piroxén összetételek a kevert jadeititekben. A jadeiteknek nagyon hasonló az összetétele a két mintában, míg az omfacitok összetétele meglehetősen eltérő, a 81/W2.5 mintában az omfacitok sokkal vasgazdagabbak, mint az N.1/81-1938 mintában.

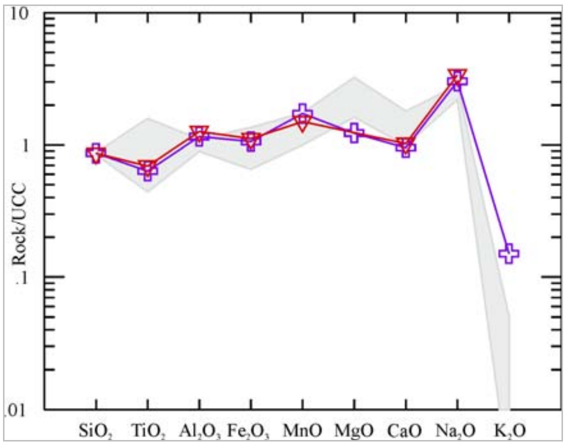

Fig. 12.: Bulk-rock compositions of mixed jade samples normalized to UCC by McLennan (2001). Two datasets are very similar to each other, and their correspondence to mixed jade data of D'Amico et al. (2003) is good too.

12. ábra: A kevert jadeititek teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). A két adatsor nagyon hasonló, és jó egyezést mutatnak D'Amico et al. (2003) kevert jadeitit adataival is.

Composition of jadeitic pyroxenes of the two samples are very similar, they appear along a relatively steep trend line. Omphacitic pyroxenes are very different, they show two distinct trends. Sample N.1/81-1938 trends towards the calcium rich omphacites, 81/W2.5 towards the iron rich aegirine-augites. The continuity of these trends is doubtful based on these data (**Fig. 11.**).

Bulk-rock data are very similar to each other. Their correspondence to data of D'Amico (2003, grey field) is good, differences probably caused by the larger natural variety of the raw material of the earlier investigated samples (**Fig. 12.**).

Iron-mixed jades

This jade group contains three pieces of stone tools, all of them found at different archaeological excavations. N.11/169-1938 was found in Zengővárkony, 99.3.1863 was found in Gorzsa and Olad-329 was found at Szombathely, Olad-plateau.

Their raw material contains a lot of jadeite with large amounts of omphacite and/or Fe-jadeite and/or aegirine-augite (**Fig. 13.**). Accessory minerals are zircon, allanite, rutile, apatite, ilmenite and titanite. Retromorphic phase (chlorite) can be found in one sample (Olad-329).

Na-pyroxenes are frequently zoned, in most of cases they have a jadeite core and Fe-jadeite or omphacite or aegirine-augite rim (**Fig. 14, Fig. 15.**).

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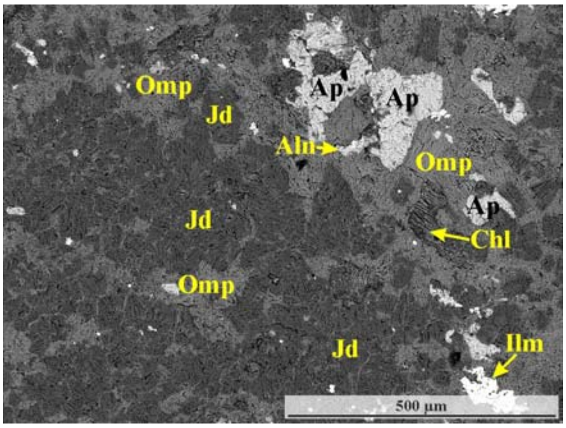

Fig. 13.: Textural view of an iron-mixed jade with accessory and retromorphic minerals. Sample Olad-329

13. ábra: Vas-kevert jadeitit szöveti képe, akcesszórikus és retromorf ásványai. Olad-329-es minta.

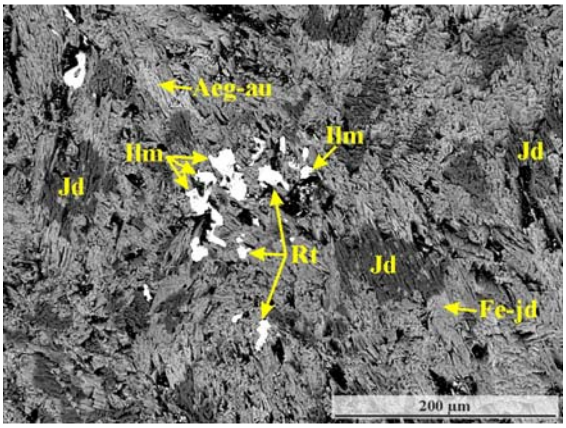

Fig. 14.: Zoned pyroxenes with jadeite core and iron-jadeite/aegirine-augite rim. Sample N.11/169-1938 (Zengővárkony).

14. ábra: Zónás piroxénkristályok jadeit maggal és vas-jadeit/egirinaugit peremmel. N.11/169-1938 minta (Zengővárkony).

The pyroxenes of these samples have rather different composition. Two of the stone tools contain three different compositional groups of pyroxene, one is near the clear jadeite composition, the second group is in the jadeite field with high iron content (iron-jadeite) and the third has omphacitic–aegirine-augitic composition. These pyroxenes have a flat trend-like line towards aegirine-augite across the omphacite field pyroxenes. Continuity of these trend lines are doubtful based on these data, again (**Fig. 15.**).

Bulk-rock data are corresponding very well to data published by D'Amico (2003, grey field). According to these results, these samples have fairly different manganese and magnesium content (**Fig. 16.**).

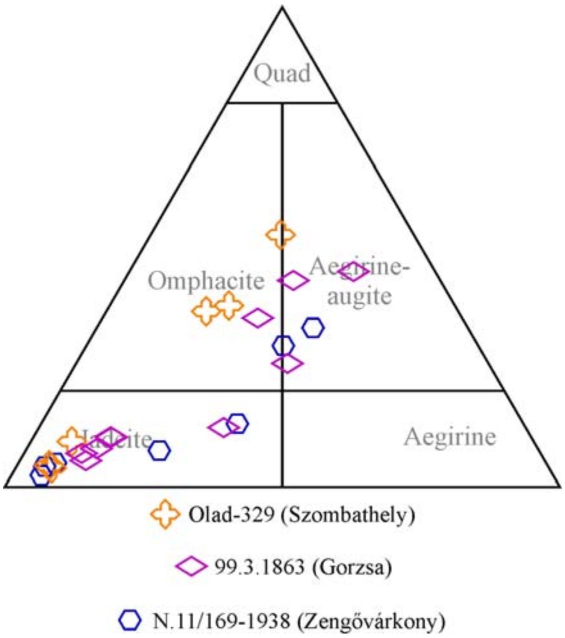

Fig. 15.: Very diverse pyroxene compositions in iron-mixed jades. Sample 99.3.1863 contains four different pyroxenes: jadeite, iron-jadeite, omphacite and aegirine-augite.

15. ábra: A vas-kevert jadeititek erősen eltérő piroxén összetételei. A 99.3.1863-as minta négyféle piroxént tartalmaz: jadeitet, vas-jadeitet, omfacitot és egirinaugitot.

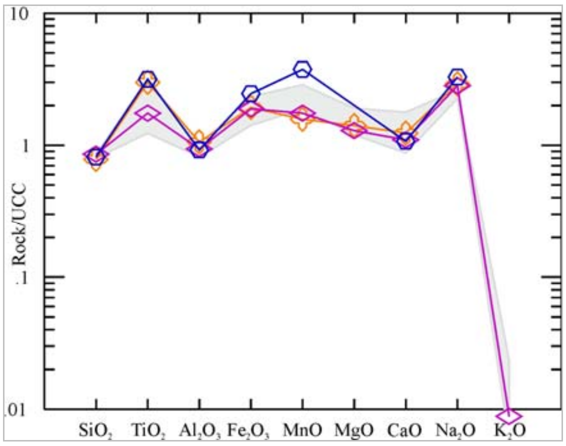

Fig. 16.: Bulk-rock compositions of iron-mixed jade samples normalized to UCC by McLennan (2001). Datasets are corresponding to iron-mixed jade data of D'Amico et al. (2003).

16. ábra: A vas-kevert jadeititek teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). Az eredmények jó egyezést mutatnak a D'Amico et al. (2003) által mért vas-kevert jadeititek értékeivel.

Iron jadeitite

Fe-jadeitite is a frequent raw material among the Hungarian HP metaophiolite stone tools, three pieces were found. Unfortunately all of them are stray finds without archaeological context. One piece is from the Ebenhöch Collection of the HNM (300/1876.247), the second is from Iszkaszentgyörgy (Prehistoric collection of Hungarian National Museum, Inv. nr. 39/1903), and the third is from Lábod from the collection of the Rippl-Rónai Museum, Kaposvár (Inv. nr. 3127).

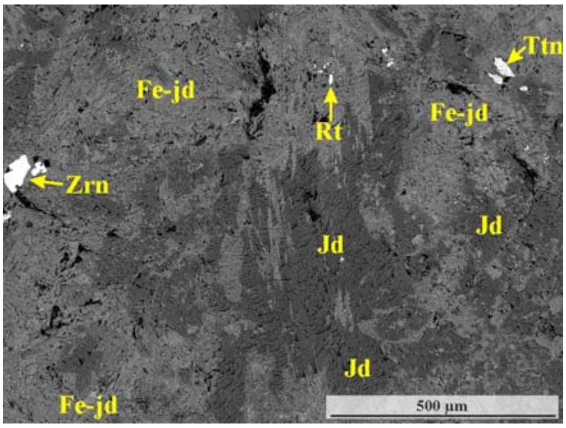

Fig. 17.: Texture and accessory minerals of iron-jadeite, sample 39/1903 (Iszkaszentgyörgy).

17. ábra: Vas jadeitit szöveti képe és akcesszórikus ásványai, 39/1903-as minta (Iszkaszentgyörgy).

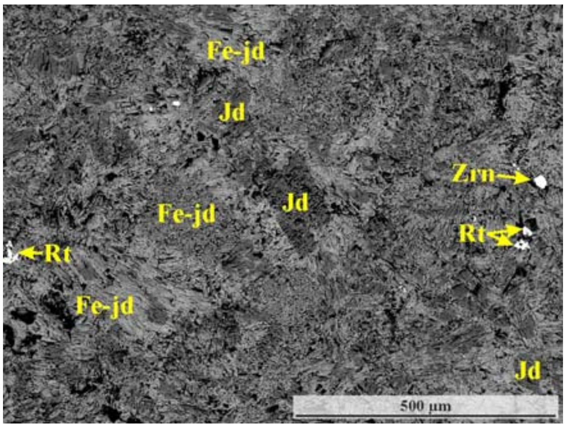

Fig. 18.: Zoned pyroxenes with jadeite core and iron-jadeite rim. Sample 3127 from Lábod.

18. ábra: Zónás piroxének jadeit maggal és vas-jadeit peremmel. 3127 minta, Lábod.

Their raw material contains jadeite and Fe-jadeite; while omphacite and aegirine-augite are absent (**Fig. 17., Fig. 19.**). Common accessory minerals are zircon and rutile. Titanite, allanite, ilmenite, xenotime and monazite can be found in them as well.

The Na-pyroxenes are generally zoned, they have jadeite core and Fe-jadeite rim (**Fig. 18, Fig 19.**). Deformation textures are absent from these samples.

Pyroxenes of sample 300/1876.247 contain more iron than pyroxenes of the other two samples. Pyroxenes appear along a very shallow/flat trend line towards aegirine (**Fig. 19.**).

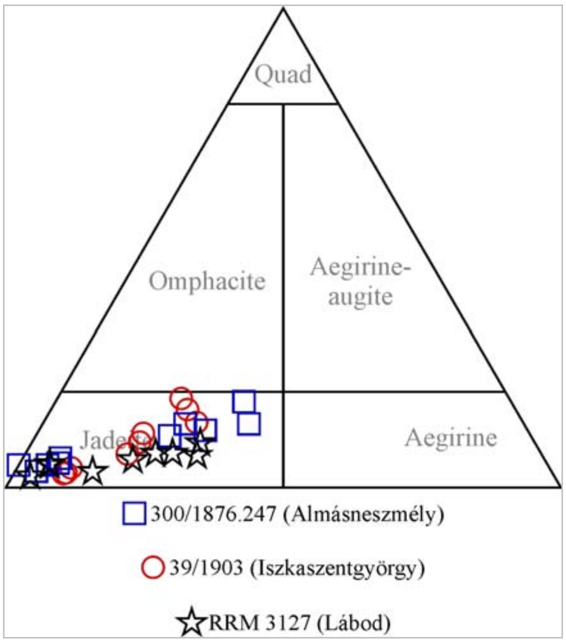

Fig. 19.: Pyroxene compositions in iron jadeitites. All of results fall to the jadeite field, however compositions are varying between iron poor and iron rich jadeites.

19. ábra: A vas-jadeititek piroxénjeinek összetétele. Minden mért érték a jadeit mezőbe esik, de az összetételek a vasszegény és vasgazdag jadeitek között váltakoznak.

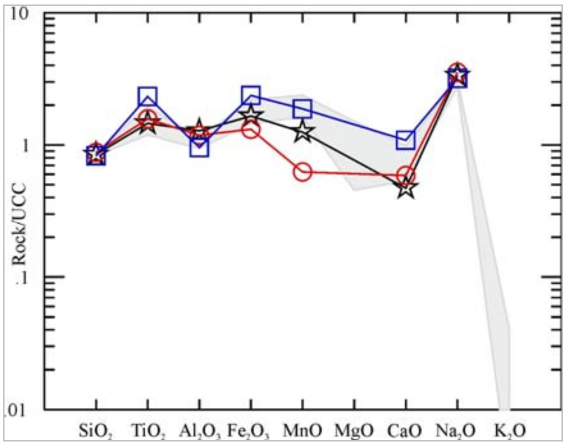

Fig. 20.: Bulk-rock compositions of iron jadeite samples normalized to UCC by McLennan (2001). Correspondence to iron jadeite data of D'Amico et al. (2003) is good.

20. ábra: A vas jadeititek teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). A mért adatok elég jól egyeznek D'Amico et al. (2003) vas-jadeitit adataival.

Correspondence of bulk-rock analyses to data of D'Amico (2003, grey field) is good, the variance is probably caused by the diversity of the raw material (**Fig. 20.**).

Glaucophane schist (retrograde omphacite schist)

This type of glaucophane schist is very rare among the Hungarian HP metaophiolite stone tools, only one piece was found until this time. This piece is from Zengővárkony archaeological site (N.5/47-1939).

Its raw material contains a lot of glaucophane and Na-pyroxenes as well. Most of the Na-pyroxenes are omphacite, jadeite occurs on the rim of omphacite crystals (**Fig. 21, Fig. 22.**). Accessory minerals are ilmenite, apatite, titanite (**Fig. 21.**).

Na-pyroxenes are zoned in this sample, but they show inverse zonation against the prior samples (**Fig. 21.**). According to the texture, glaucophane was formed in the latest, retrograde stage of metamorphism. N.5/47 has slightly foliated texture (**Fig. 21.**).

There are two compositional varieties of alkaline pyroxenes in this sample. The more abundant pyroxene is the omphacite, which has a steep trend line on the diagram. Jadeite compositions are much more close to each other, they appear in a pile and they do not form a trend line (**Fig. 22.**).

Bulk-rock data fit very well to glaucophane schist data of by D'Amico (2003, thick grey line). The only difference between the two compositions is the Na2O content, probably sample N.5/47-1939 has higher alkaline pyroxene content than the reference sample. Notably, the distribution of omphacite schist and glaucophane schist are very similar to each other (thick grey line and pale grey fields in **Fig. 23.**).

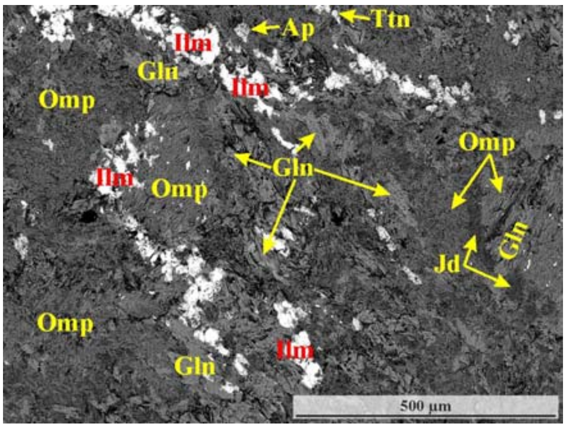

Fig. 21.: Texture of glaucophane schist with accessory minerals. Sample N.5/47-1939 (Zengővárkony).

21. ábra: Glaukofánpala szöveti képe és akcesszórikus ásványai. N.5/47-1939 minta (Zengővárkony).

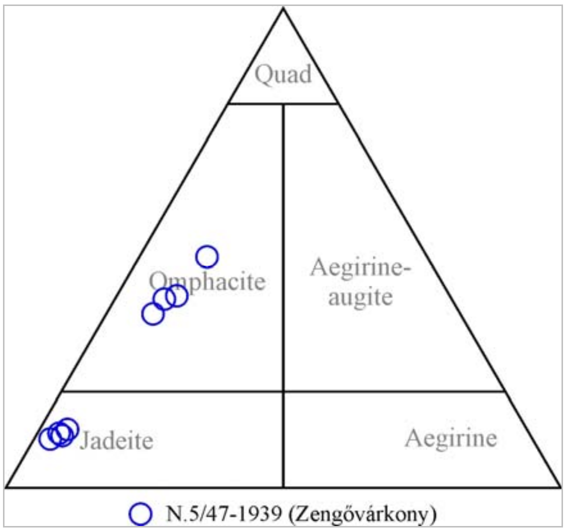

Fig. 22.: Alkaline pyroxene compositions in glaucophane schist. Compositions are showing two distinct group of jadeite and omphacite.

22. ábra: Alkáli piroxén összetételek a glaukofánpalában. A mért eredmények két különálló halmazt képeznek, egyet a jadeit és egyet az omfacit mezőben.

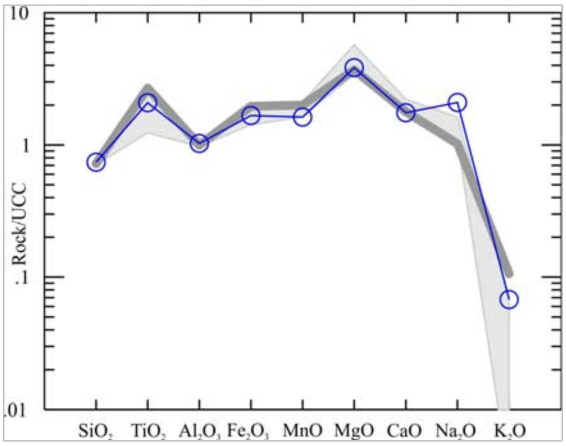

Fig. 23.: Bulk-rock compositions of glaucophane schist sample normalized to UCC by McLennan (2001). Dataset are very similar to glaucophane schist (thick gray line) and omphacite schist (pale gray field) data of D'Amico et al. (2003).

23. ábra: A glaukofánpala teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). Az adatsor nagyon hasonlít D'Amico et al. (2003) glaukofánpala (vastag szürke vonal) és omfacitpala (világosszürke mező) eredményeire.

Iron eclogites

Eclogite is rare raw material among the investigated HP metaophiolite stone tools, only two pieces were found. Both of them were found on excavated archaeological sites, M6.2010.10B.3060.3 was found in Alsónyék, Olad-321 was found at Szombathely, Olad plateau.

Their raw material contains a high amount of garnets in addition to alkaline pyroxene which is mainly omphacite (**Fig. 24.**). Accessory minerals are ilmenite, zircon, apatite and rutile. Retromorphic phase (epidote) occurs in the sample from Olad (**Fig. 25.**).

Alkaline pyroxenes seem to be homogenous in most of cases, but in some cases they have relatively jadeite-rich core (**Fig. 26.**).

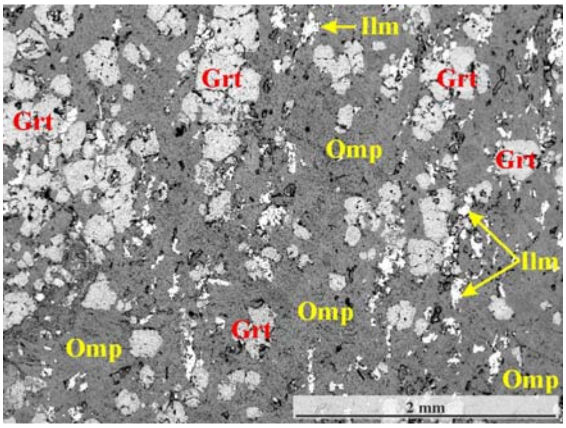

Fig. 24.: Typical texture of iron eclogites with accessory minerals and large amount of garnets. Moderate inhomogeneity of pyroxenes can be observed on this sample (M6.2010.10B.3060.3, Alsónyék). Foliation is visible on the placements of ilmenite crystals.

24. ábra: Tipikus vas eklogit szövet akcesszórikus ásványokkal és sok gránáttal. A piroxének összetételében enyhe inhomogenitás figyelhető meg. Az enyhe foliáció az ilmenit kristályok elhelyezkedésén látható. M6.2010.10B.3060.3 minta, Alsónyék.

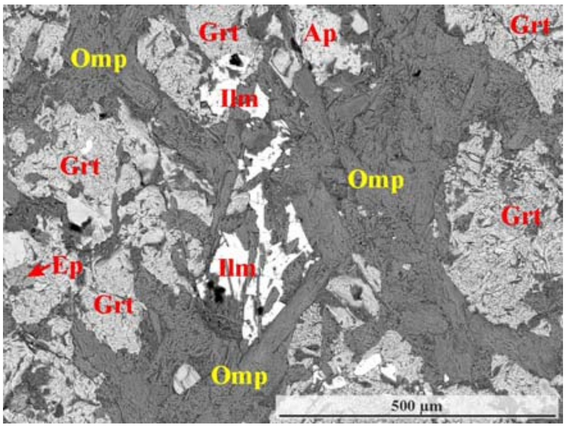

Fig. 25.: Elongated, homogenous omphacite crystals among garnets and ilmenites in the sample Olad-321.

25. ábra: Nyúlt, homogén omfacit kristályok a gránátok és az ilmenitek között az Olad-321 mintában.

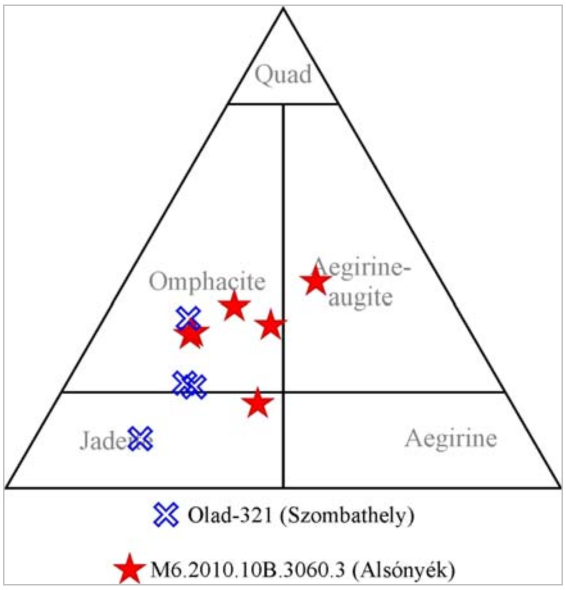

Fig. 26.: Pyroxene compositions in the iron eclogite samples. Most of the compositions fall into the omphacite field, however there are some jadeite and aegirine-augite, also.

26. ábra: Alkáli piroxének összetétele a vas eklogitokban. A legtöbb esetben omfacitos összetételt mértünk, de előfordulnak jadeit és egirinaugit összetételű mérések is.

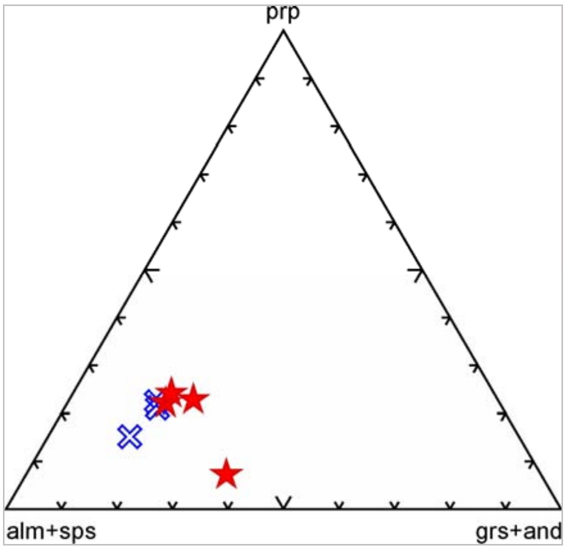

Fig. 27.: Garnet compositions in iron eclogite. Most of the compositions can be found in one narrow pile, however the distinct values are demonstrating the zonation of crystals.

27. ábra: Gránát összetételek a vas eklogitokban. A legtöbb érték egy szűk halmazban található, de néhány ettől eltérő összetétel is előfordul, amik a kristályok zónásságát bizonyítják.

Garnets are slightly zoned, probably, but this zonation is invisible on these rough surfaces (**Fig. 27.**). M6.2010.10B.3060.3 has slightly foliated texture (**Fig. 24.**).

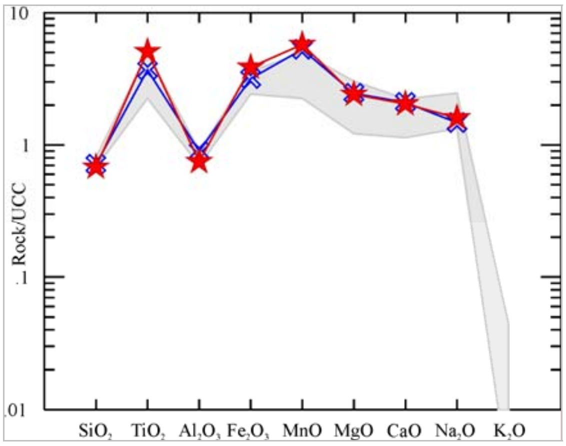

Fig. 28.: Bulk-rock compositions of iron eclogite samples normalized to UCC by McLennan (2001). Two datasets are very similar to each other, and they are corresponding very well to iron eclogite data of D'Amico et al. (2003), also.

28. ábra: A vas eklogitok teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). A két adatsor nagyon hasonló egymáshoz, és mindkettő nagyon jól egyezik D'Amico et al. 2003 vas eklogit adataival.

Pyroxene composition of these samples do not show any trends or alignment. Most of them are omphacite, however there are jadeitic and aegirine-augitic parts as well (**Fig. 26.**).

Most of the garnet compositions are in one pile, there are only two exceptions, and possibly they are indicating the zonation of the crystals (**Fig. 27.**).

Bulk-rock data are corresponding very well to data of D'Amico et al. (2003, grey field). According to these results these samples have very similar composition (**Fig. 28.**).

Discussion

Most of the investigated stone implements have characteristic, elongated, triangular or lingular shape which is similar to the stone implements described from Western Europe (D'Amico et al. 2003, D'Amico & Starnini 2006, Pétrequin et al. 2012). Instrumental analyses allow the grouping of these stone tools. According to our results, they can be divided into 6 raw material groups. These raw material groups are jadeitites, mixed jadeitites, iron-mixed jadeitites, iron jadeitites, glaucophane schist and iron eclogites (**Fig. 29.**).

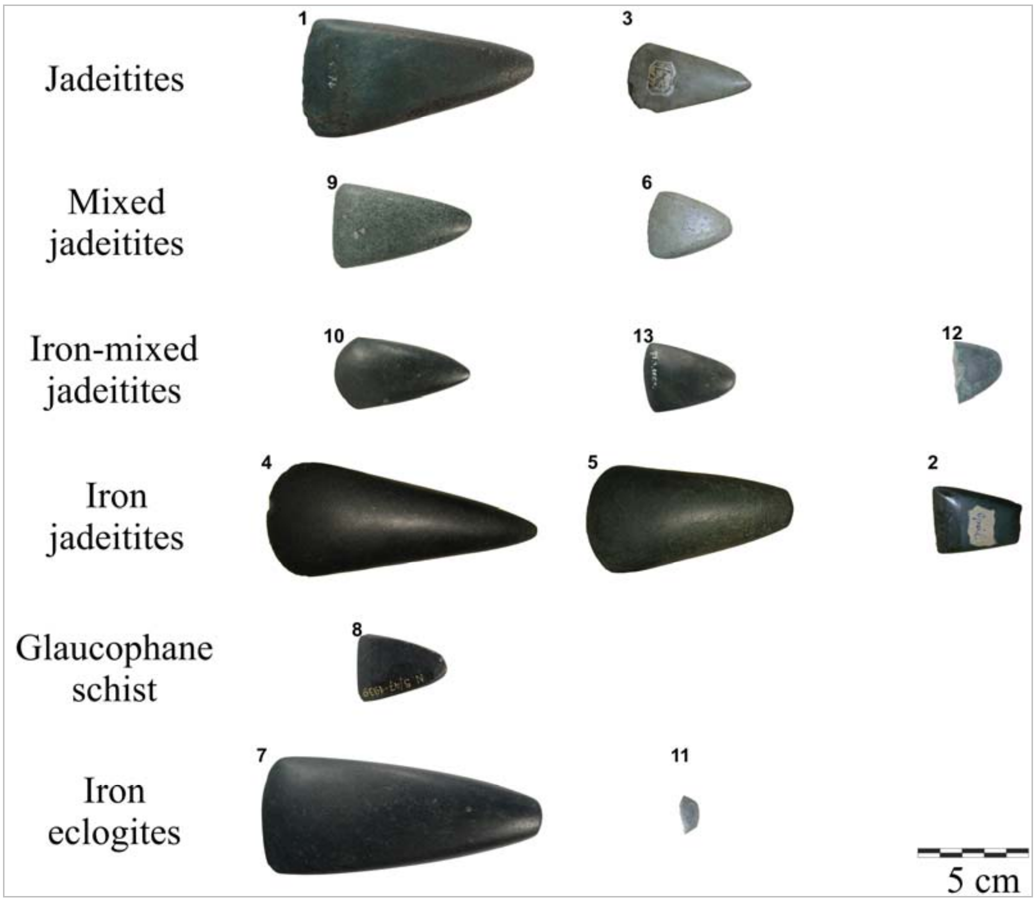

Fig. 29.: All of the presented artefacts arranged by their raw material type.

29. ábra: Az összes bemutatott kőeszköz a nyersanyag típusuk alapján elrendezve.

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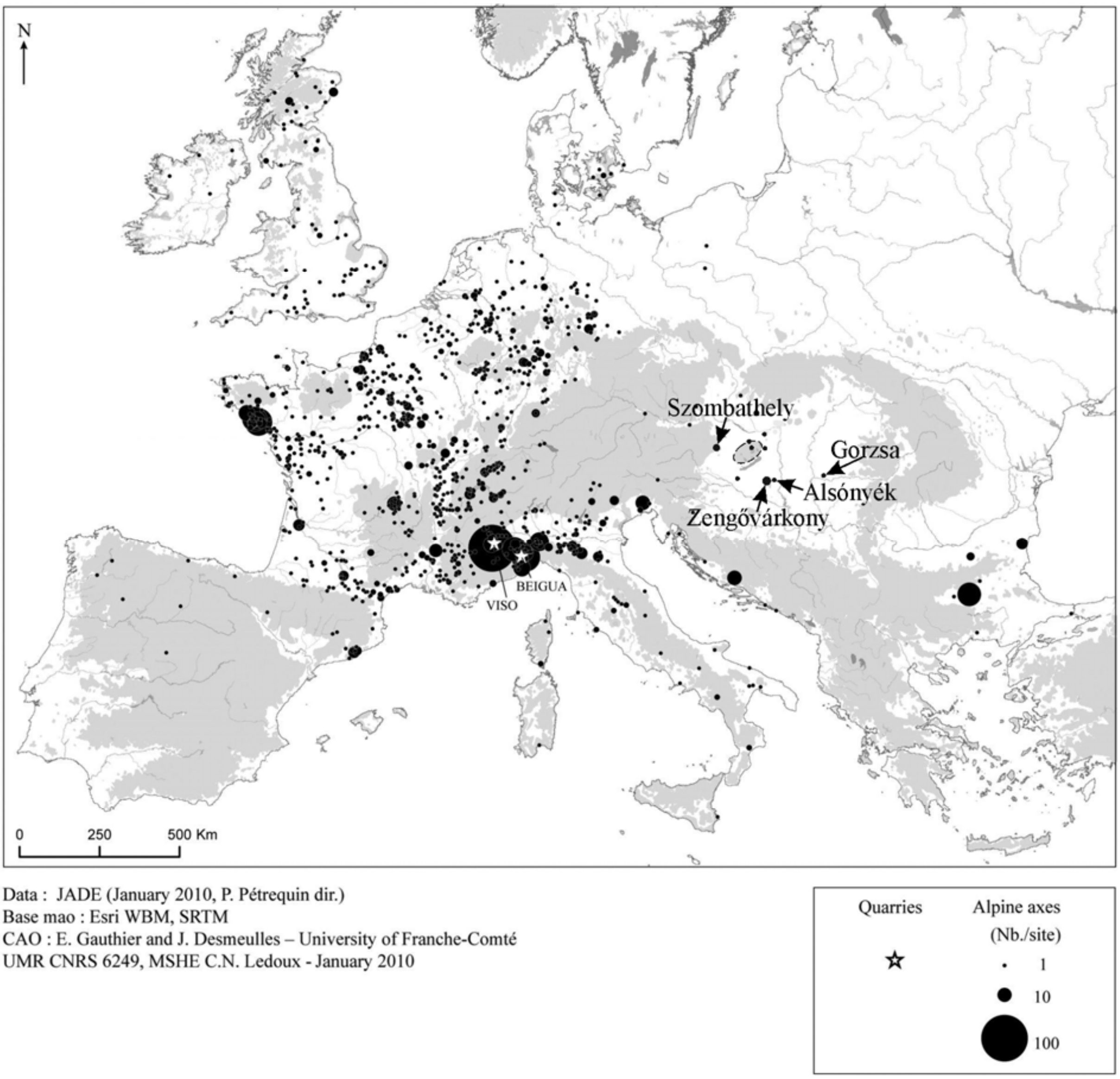

Fig. 30.: Distribution of HP metaophiolite stone implements in Europe completed with Hungarian artefacts. Archaeological excavations where the investigated finds were found are marked on map. Modified after Pétrequin et al. 2011.

30. ábra: HP metaofiolit nyersanyagú kőeszközök eloszlása Európában, kiegészítve a magyarországi adatokkal. A régészeti ásatások, melyek során a tanulmányozott leletek előkerültek, külön fel vannak tüntetve. Pétrequin et al. (2011) alapján módosítva.

Based on their inventory number, they can be grouped accordingly:

* Jadeitite: 55.1276 and 300/1876.264

* Mixed jadeitite: N.1/81-1938 and Zirc 81/W2.5

* Iron-mixed jadeitite: N.11/169-1938, 99.3.1863 and Olad-329

* Fe-jadeitite: 300/1876.247, 39/1903 and 3127

* Glaucophane schist: N.5/47-1939

* Iron-eclogite: M6.2010.10B.3060.3 and Olad-321.

Based upon data from the technical literature (D'Amico et al. 2003, D'Amico & Starnini 2006, Pétrequin et al. 2012), these raw materials, based on their mineral assemblage, chemical composition and textural properties, are identical to the North-western Italian HP metaophiolites which were formed by the Alpine orogenic stage. We do not make claims on the exact localisation of the raw material source proper because the relevant areas known from publications (see **Fig. 2.**) are rather large, and they are all at least about 1000 km from Carpathian Basin, practically in the same direction. Therefore, from the respect of prehistoric contacts they have the same significance.

Our results essentially increased the knowledge about the distribution range of these stone implements (**Fig. 30.**). Stone tools made of western Alpine HP metaophiolite raw material were formerly unknown in the Carpathian Basin, especially in Hungary. The artefacts presented here were mainly found in the Transdanubian region (West Hungary); there is one piece, however, from East Hungary as well (**Fig. 30.**).

In addition, very important result is the heterogeneity of lithotypes, not all of presented stone implements were made of the most widespread "pure jade" raw material, but eclogites and glaucophane schist are also occurring. This heterogeneity is explicable with several reasons. One possible scenario is the development of manufacturing and discovery of better and better quality sources for these stone implements in due time. Other probability is the changes in the transportation routes and/or procurement and contact modes in the function of time and/or regions. Last but not least these very distinct materials may be transported from multiple, coexistent sources/manufactures along multiple, coexistent trading routes.

Conclusion

In the last years there was a significant growth in the number of recognized and investigated stone implements made of HP metaophiolite raw material in Hungary (**Fig. 30.**). This growth is due to systematic quest for greenstones and the applied non-destructive methods (PGAA and original surface method by SEM-EDX). These methods proved their suitability to identify the raw material of stone implements and capability to provide good quality data for comparison with data in the international literature.

Artefacts from known archaeological context are from the Late Neolithic period and most of them belong to the Lengyel Culture. The only piece from East Hungary belongs to the Tisza Culture which is coeval to Lengyel Culture, and they were in regular contact with each other.

According to our results there is a large probability of finding other HP metaophiolite artefacts in Hungary, especially in the course of systematic petrologic investigations of findings of Lengyel and coeval cultures and from the old collections.

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Abbreviations on pictures

ab – albite

aeg-au – aegirine-augite

aln – allanite

ap – apatite

chl – chlorite

ep – epidote

Fe-jd – iron-jadeite

gln – glaucophane

grt – garnet

ilm – ilmenite

jd – jadeite

omp – omphacite

rt – rutile

ttn – titanite

xtm – xenotime

zrn – zircon

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