






PROVENANCE OF POLISHED STONE TOOLS FROM THE BARADLA CAVE, AGGTELEK, NORTH-HUNGARY

AZ AGGTELEKI BARADLA-BARLANG (ÉSZAK-MAGYARORSZÁG) CSISZOLT KŐESZKÖZEINEK NYERSANYAG EREDETE •

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This article is dedicated to the memory of Gábor Rezi Kató, who made an enduring contribution to the archaeological exploration of the Baradla Cave.

Abstract

The Baradla Cave has been an open and used natural cavity since ancient times. 20 pieces of polished stone tools of various raw materials were revealed from the cave site, which were analysed using non-destructive methods. Source areas were identified for those raw material types which have detailed mineralogical and rock chemistry data available. The main raw material types are blueschist and contact metabasite, defined as a close and a distant raw material, respectively. The latter was widely known for its excellent mechanical properties. In addition, subordinate greenschist-amphibolite, serpentinite, sandstone, limestone, basalt litho-typed polished implements were also identified from the Cave, but their source area can be determined only tentatively from the available analyses and previous publication data.

Kivonat

A Baradla-barlang ősidők óta nyitott és használt természetes üreg volt. 20 darab változatos nyersanyagú csiszolt kőeszköz került elő innen, ezek archeometriai vizsgálatát végeztük el roncsolásmentes módszerekkel és azonosítottunk forrásterületet azoknál a nyersanyag típusoknál, amelyeknél részletes ásvány- és kőzetkémiai adatok rendelkezésünkre álltak. A fő nyersanyag típusok a közeli nyersanyag típusként azonosítható kékpala és az elterjedt távoli nyersanyag típus, a kontakt metabázit, ami kiváló kőzetmechanikai tulajdonságai miatt széles körben ismert volt. A fentiekben túl alárendelten zöldpala-amfibolit, serpentinit, homokkő, mészkő, bazalt anyagú kőeszközök is meghatározásra kerültek a barlangból, ám ezek forrásterülete a rendelkezésre álló mérési és eddigi publikált adatokból csak feltételesen állapítható meg.

KEYWORDS: POLISHED STONE IMPLEMENTS, PROVENANCE, BARADLA CAVE, METABASITE

KULCSSZAVAK: CSISZOLT KŐESZKÖZÖK, FORRÁSTERÜLETEK, BARADLA-BARLANG, METABÁZIT

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Introduction

The Baradla-Domica Cave system is situated at the border of North Hungary and South Slovakia. The Baradla Cave is the second largest one in Hungary with its 30 km length. The nearly 6 km long Domica is located in the Slovak Republic.

In the prehistoric times the Cave system was open on both sides of the recent borders. In Aggtelek (Hungary) there are two anciently opened entrances, both of them are situated at the distinctive cliff face, recently called as the Main Entrance/Főbejárat and Bat branch/Denevér-ág) (Székely 2003) (Fig. 1.). Proved by many archaeological findings, the Domica entrance must have been opened up in the prehistoric times but collapsed and/or clogged somewhen after the Neolithic period and was rediscovered only in 1926.

In the Aggtelek site the first documented archaeological excavations were led by Jenő Nyáry in 1876-1877 (Nyáry 1881). In the ancient times the Cave was used as a temporary dwelling, a shelter, a cemetery and a possible ritual site for a long time. From both Domica and Baradla, Bükk and Late Bronze ceramics besides traces of fireplaces, pile-huts, skeletons and polished stone tools were found even farther from the natural entrances (Tompa 1937, Holl 2007, Laczi 2011, Rezi Kató 2014). As the cave system was inhabited and used for over long and different periods of time, determining the exact age of the polished stone tools is challenging. According to Holl (2007) and Rezi Kató (2014) the archaeological findings are mixed, furthermore detailed information about the circumstances of the excavations are even not described in the inventory book of the Hungarian National Museum. The latest archaeological survey in the cave was led by Gábor V. Szabó (ELTE) in 2019, which survey excavated the BD 25-28 polished stone tools linked to Bükk ceramic fragments (Nyíró et al. 2022).

Generally speaking, most of the polished implements are flat chisels therefore we presume that they belong to the Bükk culture, Middle Neolithic period. Regarding the 1911.21.28. curved hoe, 1929.64.75. handstone and 1929.64.63. stone implements, their shapes do not fit exclusively to the Middle Neolithic era; they might also date from the Late Bronze Age. In 2014, an archaeometric study began to process the Neolithic polished stone tools of the Herman Ottó Museum being the largest archaeological collection of Northeast Hungary. During this investigation period many lithotypes were learned and matched with provenance. In the framework of this archaeometric research, five stone tools from the Baradla Cave were discovered (Kereskényi 2021).

The aim of this study is the archaeometric analysis of all the polished stone tools excavated from the

Baradla Cave. Surprisingly, only 20 polished stone tools are known from the excavations so far, officially belonging to the Institute of Archaeology of Eötvös Loránd University, the Hungarian National Museum and the Herman Ottó Museum (App. Table 1.). The exact archaeological locality of the implements was not documented (Holl 2007), except for the BD25-28 polished stone implements which were excavated from the Fox branch (Fig. 1.). In this paper, from the aspect of provenance we compare the formerly published data with the compositions of the Baradla polished stone implements.

Analytical and calculation methods

As most of the implements are intact and/or borrowed, exclusively non-destructive mineralogical and petrological analytical methods were allowed to carry out.

All samples were macroscopically described and measured for their magnetic susceptibility with KT-5 Kappameter using thickness correction (Bradák et al. 2009).

The mineral chemistry analyses were performed by electron microprobe analysis of the stone tools using the original surface method (Bendó et al. 2013). The mineral association and textural relations were studied by a JEOL JXA 8600 Superprobe electron-microprobe in energy-dispersive mode. The accelerating voltage was 15 kV and the beam current was 20 nA.

BD25 implement was measured by a Zeiss EVO/MA 10 electron microscope using 20 kV accelerating voltage in EDS mode.

Non-destructive X-ray diffraction (XRD) analyses were accomplished on six stone implements with a Bruker D8 Advance X-ray diffractometer. Parameters of XRD analyses: CuK α source, 40 kV and 20 mA generator settings, parallel beam geometry (Göbel-mirror), Vantec1 position detector (1° window opening degree), 0.1 mm collimator. After the measurements, the crystallographic phase identification was performed with Bruker DiffracPlus EVA based on ICDD PDF2 and COD (Crystallography Open Database) database, using Search/Match algorithm. Rietveld fitting was performed on high-priority samples using TOPAS4 software (Kristály & Kereskényi 2016).

Both SEM-EDS and XRD measurements were carried out at the Institute of Mineralogy and Geology, while the BD25 implement were studied at the Department of Metallurgy at University of Miskolc.

Bulk elemental composition of 17 polished stone tools has been determined by prompt-gamma activation analysis (PGAA) at the 10⁸ n/cm²·s intensity external cold neutron beam of the

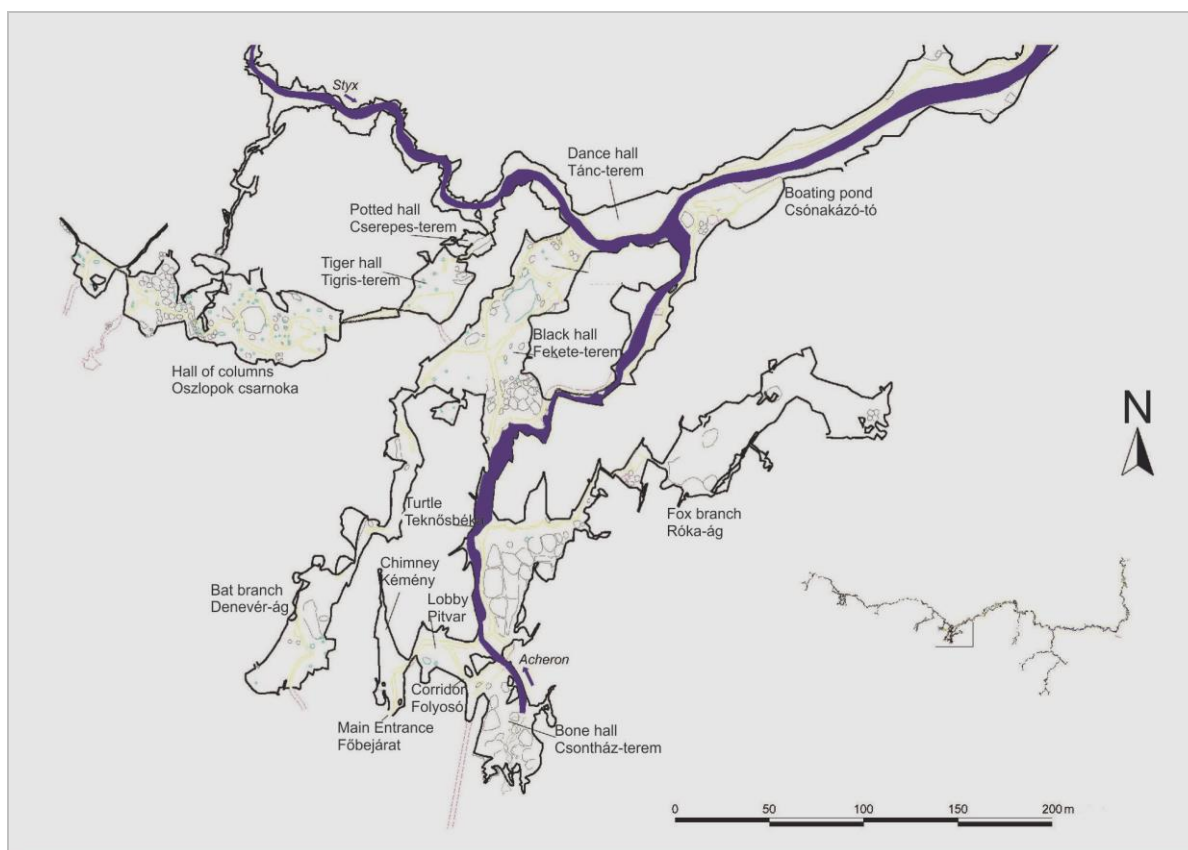


Fig. 1.: Surroundings of the Main entrance in Aggtelek, where most of the archaeological findings were excavated (after Ország et al. 1989).

1. ábra: Az aggteleki főbejárat környéke, ahonnan a legtöbb régészeti lelet előkerült (Ország et al. 1989 után).

Budapest Research Reactor. The upgraded Budapest PGAA facility has been described by Szentmiklósi et al. (2010).

The smaller pieces have been analysed at the PGAA station, while the bigger ones have been analysed at the NIPS-NORMA station. The beam size was adjusted between 24 mm² and 44 mm². Since the neutrons can penetrate a few cm deep into the sample, the results provided will be characteristic for the bulk material. The acquisition time has been set between 1300 s and 4100 s, in order to obtain statistically satisfactory counts in the spectrum peaks. With the PGAA method, it was possible to determine quantitatively the amount of the major components of SiO₂, TiO₂, Al₂O₃, Fe₂O₃, MnO, MgO, CaO, Na₂O, K₂O, H₂O, occasionally CO₂, which indicate the presence of carbonates and SO₃, as well as trace elements of B, Cl, Sc, V, Cr, Ni, Sm and Gd non-destructively as it was demonstrated by Kasztovszky et al. (2008), Szakmány and Kasztovszky (2004), Szakmány et al. (2011).

To calculate cation numbers of amphiboles the ACES Excel spreadsheet was used. The initial M³⁺/ΣM ratio was not retained, the algorithm

sought electroneutrality by adjusting the valences of Fe and Mn automatically selecting one or more of four cation normalization schemes: sum Si to Ca (+Li) = 15; sum Si to Mg (+Li) = 13; sum Si to Na = 15; sum Si to K = 16 apfu (atomic proportion per formula unit). Schemes were selected automatically based on the smallest maximum deviations from criteria (Locock 2014). Amphiboles were named following the classification of Hawthorne et al. (2012).

Results and discussion

Blueschist

Macroscopic description

Eight flat chisels proved to be blueschist. Macroscopically they are fine-grained (grains cannot be distinguished by naked eye), having bluish, bluish green, greenish, greyish blue colour with fibrous texture. Dominantly they are massive and non or slightly foliated. A few of them have characteristic green patches on their surface, while on some chisels these spots are elongated in the direction of foliation (Figs. 2/A, /C, /E). The magnetic susceptibility is low, and ranges in a narrow interval, 0.35–0.64*10⁻³ SI.

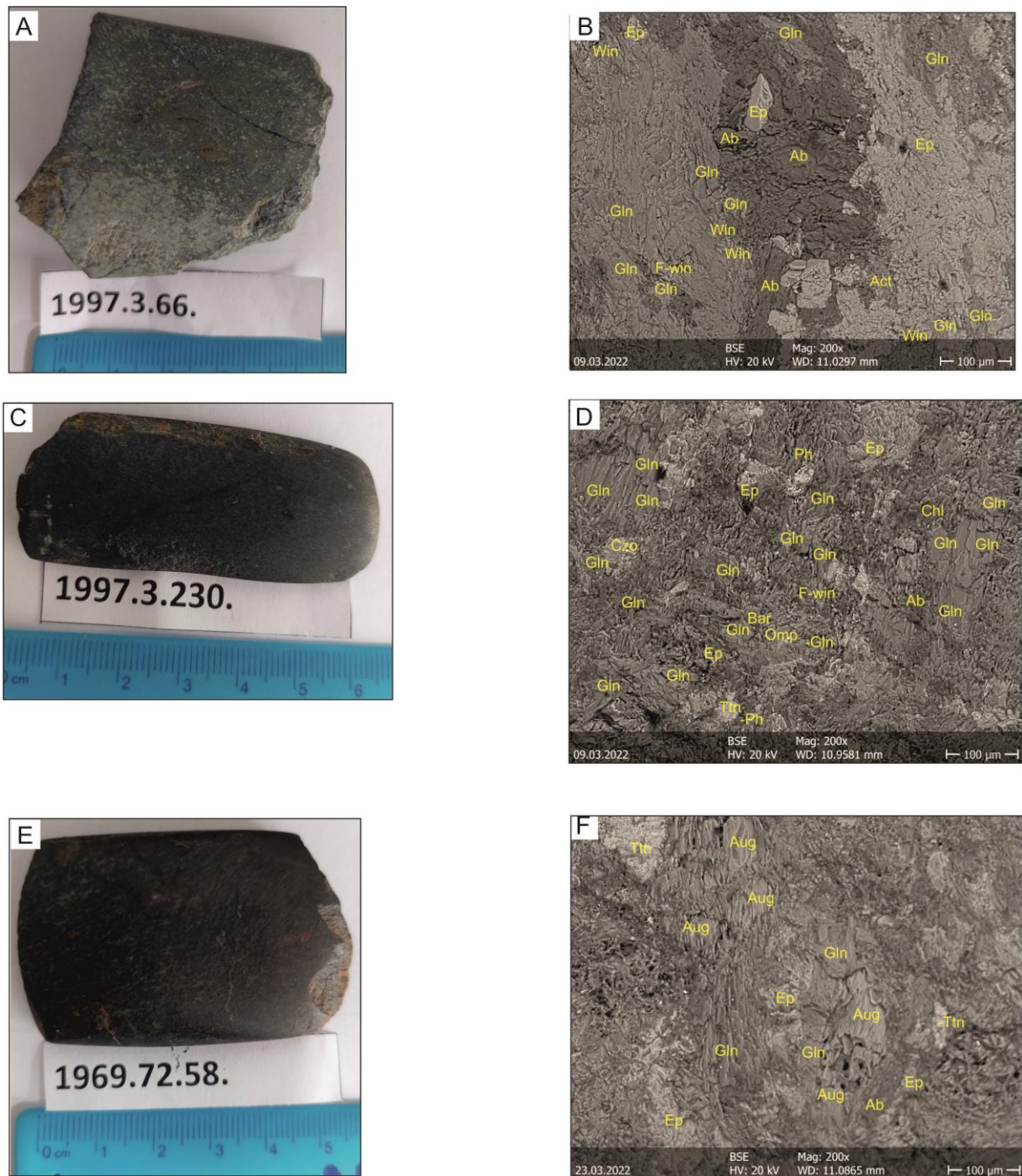


Fig. 2.: Macroscopic (A) and SEM-BSE image (B) of 1997.3.66. blueschist polished stone implement with green patches on its surface. Macroscopic (C) and SEM-BSE image (D) of 1997.3.230. blueschist polished stone implement. The presence of omphacite indicates prograde metamorphism towards eclogite facies. Macroscopic (E) and SEM-BSE image (F) of 1969.72.58. blueschist polished implement, with green patches elongated parallel with the foliation. Relict augite grains are preserved from the magmatic phase.

2. ábra: Makroszkópos (A) és SEM-BSE kép (B) az 1997.3.66. jelű kékpala csiszolt kőeszközről. Makroszkópos (C) és SEM-BSE kép (D) az 1997.3.230. jelű csiszolt kőeszközről. Az omfacit megjelenése a progresszív metamorfózist jelöli az eklogit fácies felé. Makroszkópos (E) és SEM-BSE kép (F) az 1969.72.58. jelű csiszolt kőeszközről. A zöld foltok a foliáció irányával párhuzamosan megnyúltak. Relikt augit szemcsék a magmás fázisból őrződtek meg.

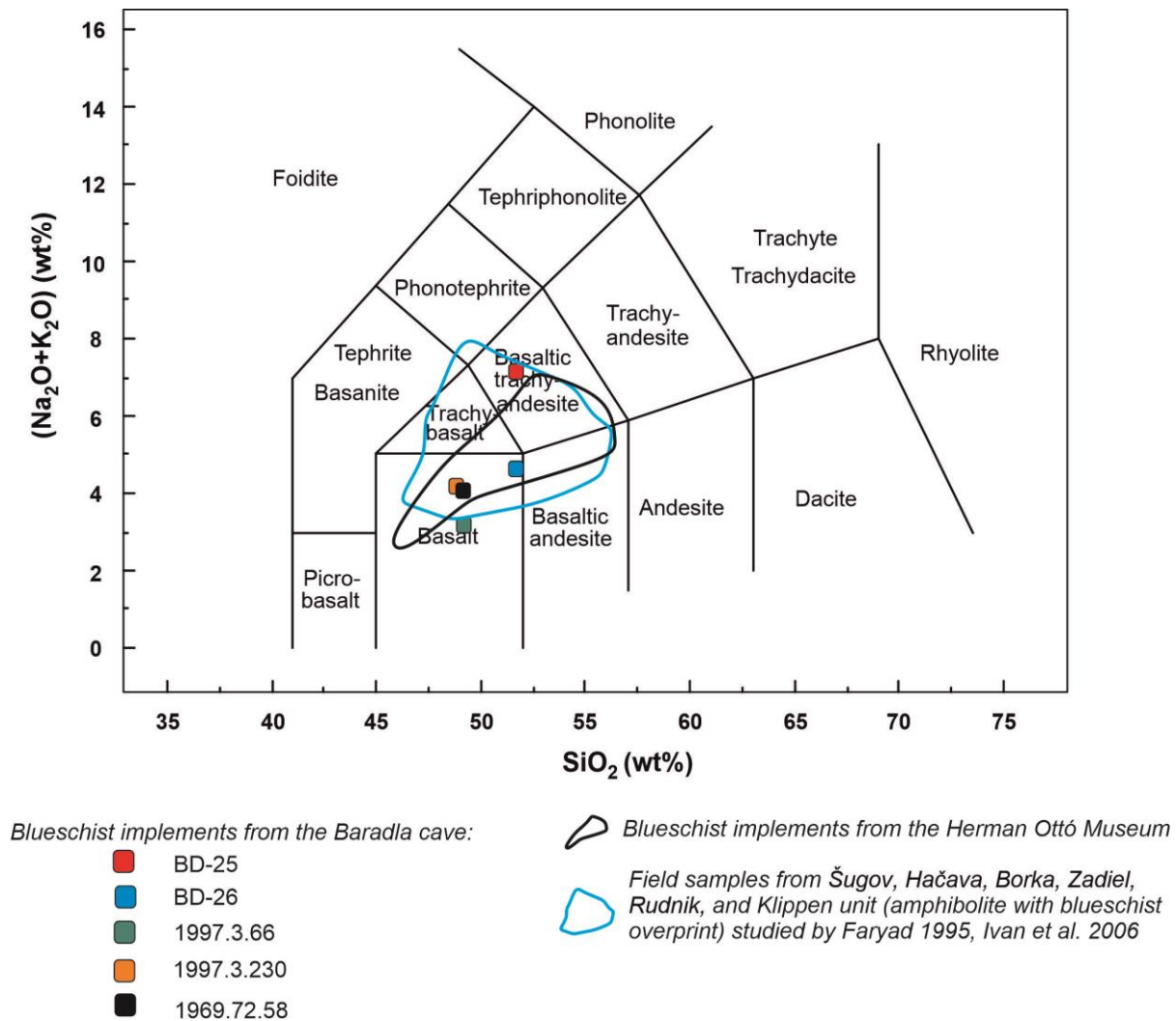


Fig. 3.: Chemistry of blueschist implements from the Baradla Cave compared with formerly studied implements from the collection of Herman Ottó Museum (Kereskényi et al. 2018) and field samples (Faryad 1995, Ivan et al. 2006) plotted in TAS diagram (Le Bas et al. 1986).

3. ábra: A Baradla-barlangból előkerült kékpala kőszközők kőzetkémiai eredményei TAS diagramon (Le Bas et al. 1986) ábrázolva, összevetve a Herman Ottó Múzeum gyűjteményébe tartozó, korábban vizsgált kőszközőkkel (Kereskényi et al. 2018) és a terepi mintákkal (Faryad 1995, Ivan et al. 2006).

Bulk chemistry

Compositional data of five implements were studied and listed in **App. Table 2**. The SiO_2 content is low and ranges in a narrow interval from 45.85 to 49.86 wt%. The CaO content varies in a quite wide range from 2.34 to 11.23 wt%, while the Na_2O covers an interval with 2.96–6.73 wt%. Four samples plot into the basalt field, while BD25 matched to the basaltic trachyandesite field in the TAS diagram (**Fig. 3**.) due to the highest Na_2O content in the group.

In the AFM diagram four implements show tholeiitic affinity, while the BD25 has calc-alkaline feature (**Fig. 4**).

Petrography and mineral chemistry

Six blueschist polished stone tools contain glaucophane as a rock-forming mineral. Glaucophane yielded as nematoblasts which are parallel oriented to foliation. Some analysed points of blue amphiboles were classified as ferro-glaucophane in the BD26 and 1969.72.58 implements. The compositional ranges of sodic amphiboles, as a function of $\text{Fe}^{2+}/(\text{Fe}^{2+}+\text{Mg})$ vs $\text{Fe}^{3+}+\text{Al}$ in formula, is shown in **Fig. 5**. Chemical compositions of Na-amphiboles are presented in **App. Table 3**.

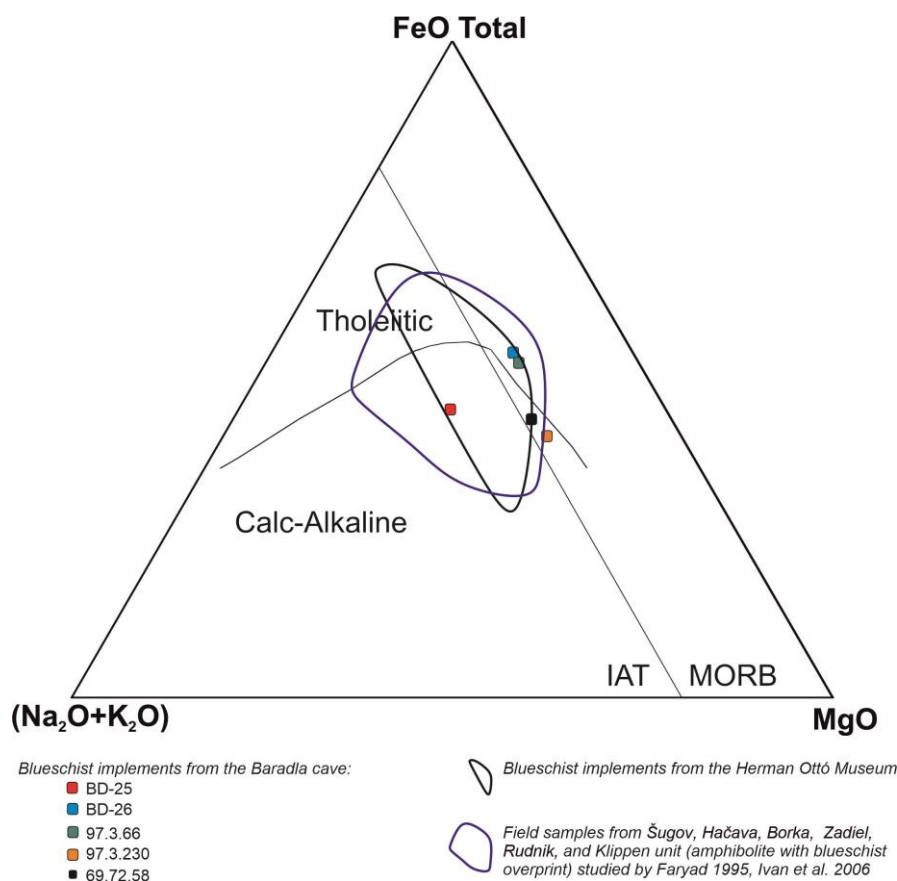


Fig. 4.: Chemistry of blueschist implements from the Baradla Cave and formerly studied implements from the collection of the Herman Ottó Museum (Kereskényi et al. 2018) and field samples (Faryad 1995, Ivan et al. 2006) plotted in the AFM diagram (Irvine & Baragar 1971). A = alkali (Na₂O + K₂O), F = FeO (total), M = MgO. IAT = island arc tholeiite, MORB = mid-ocean ridge basalt.

4. ábra: A Baradla-barlangból előkerült kékpala kőszközök kőzetkémiai eredményei AFM diagramon (Irvine & Baragar 1971) ábrázolva, összevetve a Herman Ottó Múzeum gyűjteményébe tartozó, korábban vizsgált kőszközökkel (Kereskényi et al. 2018) és a terepi mintákkal (Faryad 1995, Ivan et al. 2006).

Winchite, ferro-winchite and ferri-winchite were perceived from the rims of a glaucophane of the 1997.3.66. and 1997.3.230. implements (**Figs. 2/B, /D**). Chemical compositions are represented in **App. Table 4**.

In the 1997.3.230. sample barroisite (**App. Table 4**.) was observed, its textural relation is unclear (**Fig. 2/D**).

Magnesio-ferri-hornblende was perceived in the BD26 sample (**App. Table 4**.), its textural relation cannot be recognised.

Actinolite was observed at the edges of glaucophane in the 1997.3.66. implement (**Fig. 2/B**) (**App. Table 4**).

Augite (En_{0.52-0.57} Fs_{0.06-0.09} Wo_{0.35-0.40}) with cc. 150 µm crystal size was detected as a relict phase next to glaucophane and albite in the 1969.72.58. sample (**Fig. 2/F**).

Omphacite grain with cc. 60 µm was described next to glaucophane from the 1997.3.230. sample (**Fig. 4/D**).

Albite is observed in all tools as helictic poikiloblasts in large amount (**Figs. 2/B, /D, /F**).

Epidote/clinozoisite were detected in all Baradla blueschist samples in various amounts (**Figs. 2/B, D, F**).

Chlorite was detected as clinochlore in BD25, BD26 and 1997.3.230. stone implements, however, it is supposed that it may be also present in other blueschist tools as it was one of the most common mineral phases in the previously studied implements. Due to the limitation of the original surface method clinochlore phase cannot be noticed in each case.

Titanite is a very common accessory mineral in all tools (**Figs. 2/D, F**). Its Al₂O₃ content is typically high which can be explained by the increase of metamorphic pressure during titanite crystallization (Smith 1981, Faryad 1995).

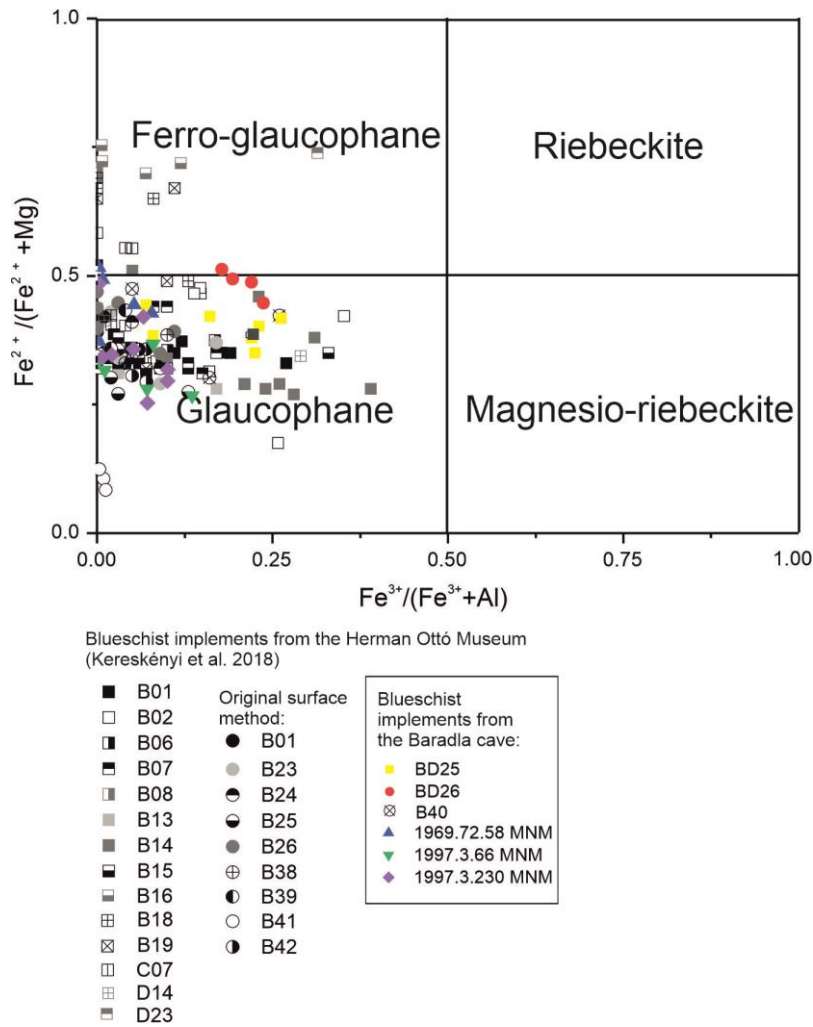


Fig. 5.: Composition of sodic amphiboles in blueschist implements from the Baradla Cave and formerly studied implements from the collection of the Herman Ottó Museum (Kereskényi et al. 2018) plotted in the $Fe^{3+}/(Fe^{3+}+Al)$ vs. $Fe^{2+}/(Fe^{2+}+Mg)$ diagram.

5. ábra: A Baradla-barlangból előkerült kékpala kőeszközök Na-amfiboljainak összetétele a $Fe^{3+}/(Fe^{3+}+Al)$ vs. $Fe^{2+}/(Fe^{2+}+Mg)$ diszkriminációs diagramon ábrázolva, összevetve a Herman Ottó Múzeum gyűjteményébe tartozó, korábban vizsgált kőeszközökkel (Kereskényi et al. 2018).

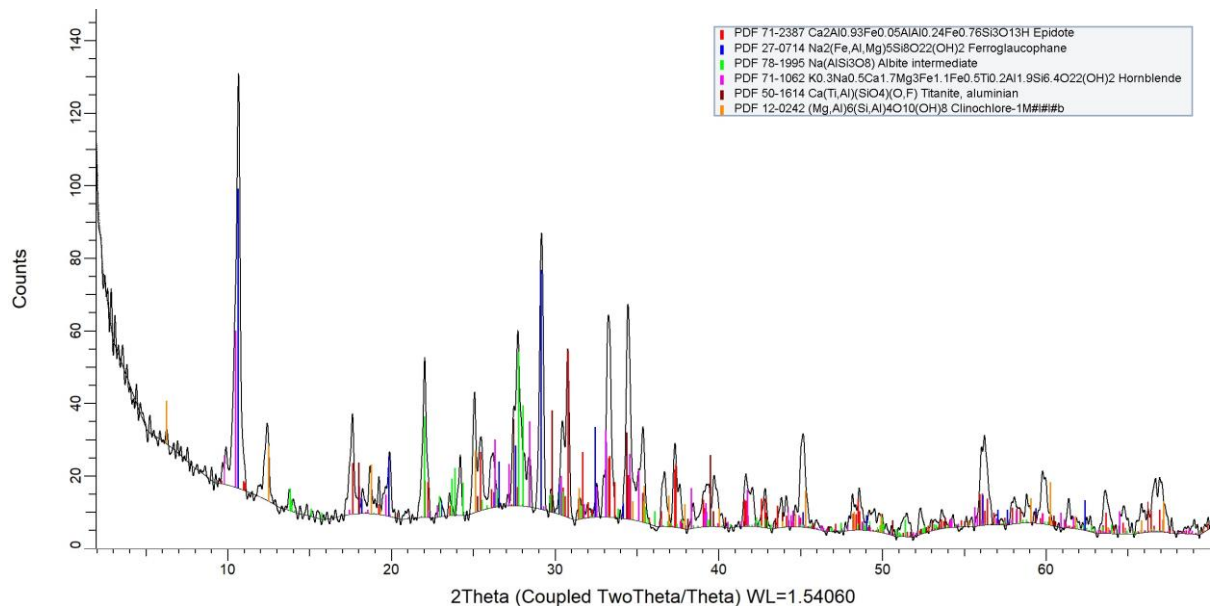


Fig. 6.: XRD pattern of 53.188.1. blueschist implement.

6. ábra: 53.188.1. jelű kékpala kőbaltá XRD felvétele.

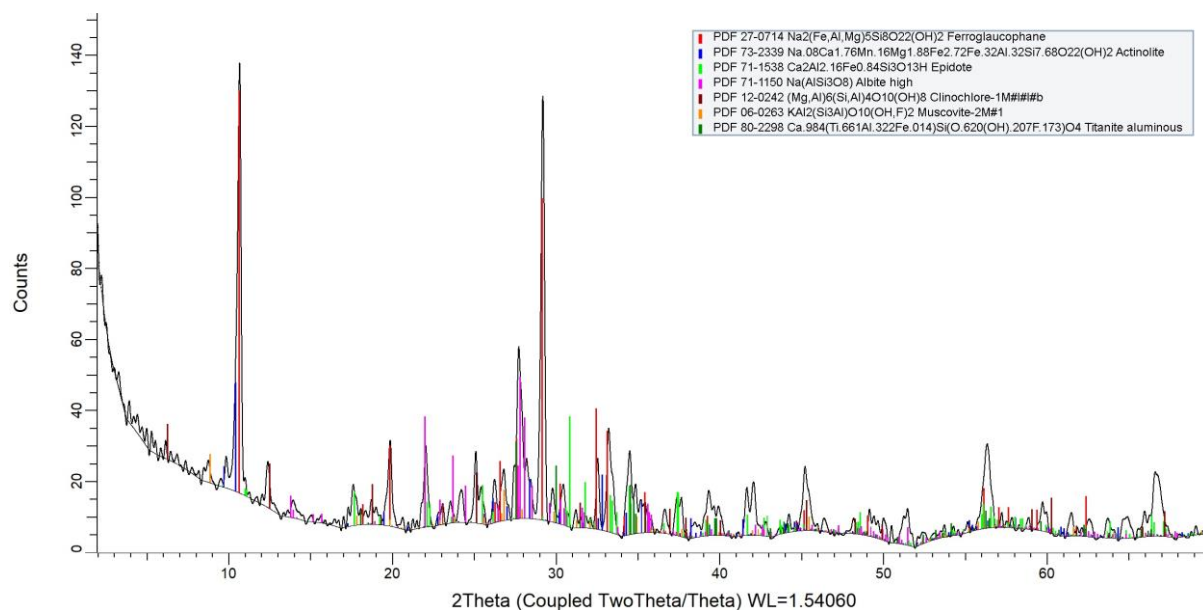


Fig. 7.: XRD pattern of 53.188.2 blueschist implement.

7. ábra: 53.188.2. jelű kékpala kőbalta XRD felvétele.

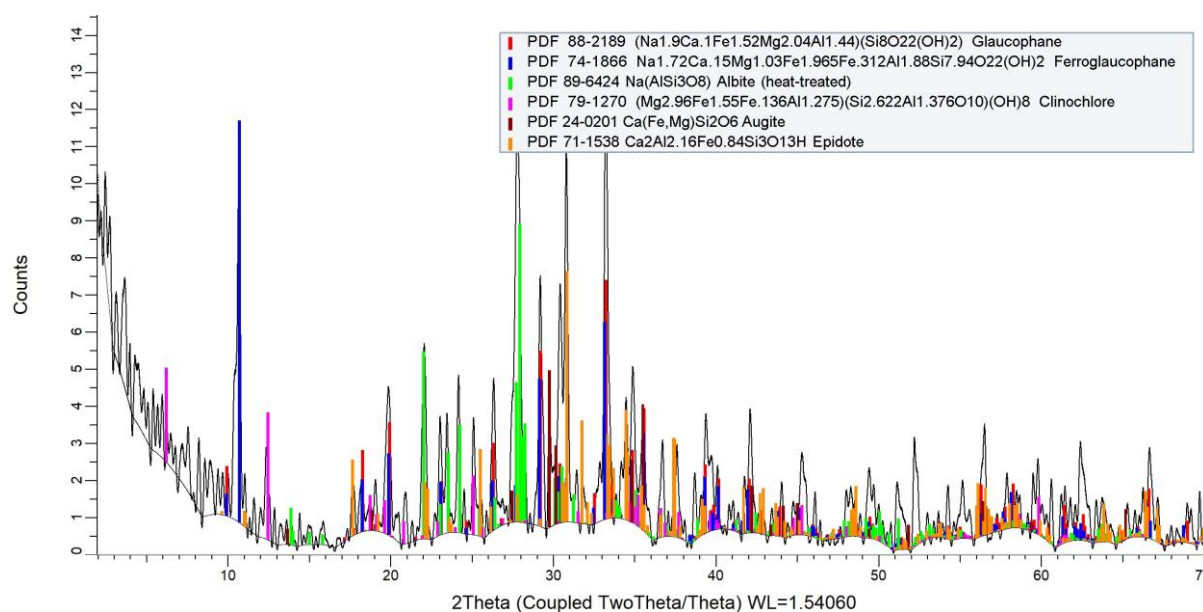


Fig. 8.: XRD pattern of 1969.72.58. blueschist implement.

8. ábra: 1969.72.58. jelű kékpala kőbalta XRD felvétele.

XRD of blueschist implements

In a previous study we managed to identify glaucophane/ferro-glaucophane and distinguish them from other amphiboles by XRD (Kristály & Kereskényi 2016). This method helped to recognize blueschist implements in a relatively rapid, non-destructive way. In this work three blueschist implements were studied by XRD from the Baradla Cave.

The detected minerals by XRD from sample 53.188.1. are ferro-glaucophane, epidote, albite, hornblende, titanite and clinochlore (**Fig. 6.**). Sample 53.188.2. analysed by XRD yielded ferro-glaucophane, actinolite, epidote, albite, clinochlore, muscovite and titanite (**Fig. 7.**). Implement 1969.72.58. was also studied by XRD. The detected minerals are glaucophane, ferro-glaucophane, albite, clinochlore, augite, epidote (**Fig. 8.**), which association was confirmed by SEM-EDS either, except for ferro-glaucophane.

Possible provenance of the blueschist implements

Formerly large numbers of blueschist polished stone implements were examined in details and compared to the most possible provenance field. The blueschist outcrops linked to a 20 km long subduction zone, Bôrka Nappe in Meliata unit in Southeast Slovakia (Kereskényi et al. 2018, Kereskényi 2021). The bulk chemistry data of Bôrka Nappe and Neolithic polished stone implements of Northeast Hungary were compared to the Baradla blueschist implements in the TAS and AFM diagrams and showing high correlation affinity (Figs. 3. and 4.).

The observed mineral assemblage of the Baradla implements are varied: glaucophane ± ferro-glaucophane ± winchite ± ferri-winchite ± ferro-winchite ± barrosite ± magnesio-ferri-hornblende ± actinolite ± omphacite ± augite + albite + epidote/clinozoisite + chlorite + titanite. The mineral chemistry and texture were also compared to the previously published data. In BD25, BD26 and B40 samples only glaucophane and ferro-glaucophane were detected as rock-forming

amphiboles. In 1997.3.66. and 1997.3.230. samples, Na-Ca amphiboles were perceived at the rims of glaucophane (Figs. 2/B, /D) showing a temperature increase and a pressure decrease after the blueschist metamorphism (Otsuki & Banno 1990). In 1997.3.66. sample actinolite appears at the rims of winchite/glaucophane demonstrating a greenschist facies overprinting (Fig. 2/B).

In the 1997.3.230. sample, the presence and textural position of omphacite indicates a progressive metamorphism towards the eclogite facies (Fig. 2/D).

In 1969.72.58. sample, the large relict augite is preserved (Fig. 2/F) from the magmatic phase due to the lack of volatiles.

Observing the mineral phases and their textural relations in the case of Baradla blueschist implements a polyphase metamorphism can be outlined such as in the case of the implements of Northeast Hungary and the Meliata samples (Faryad 1995, Kereskényi et al. 2018, Kereskényi 2021).

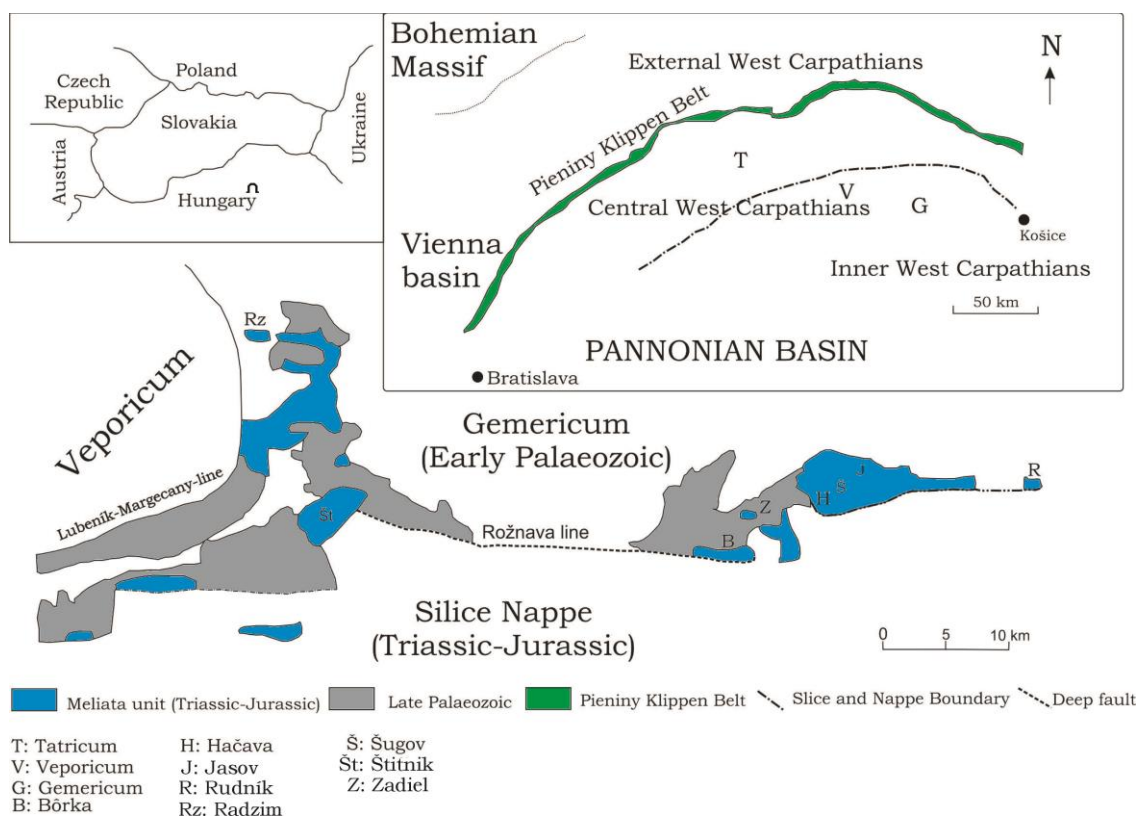


Fig. 9.: The most possible provenance field of blueschist implements in a geological setting of the Meliata blueschists in a framework of the Western Carpathian geotectonic division. The schematic geological map of the southern part of the Gemicum after Bajanič et al. (1983).

9. ábra: A Mellétei-egység kékpala előfordulásai a Nyugati-Kárpátokkal, vázlatos geológiai térképen ábrázolva Bajanič et al. (1983) után.

Based on the proximity and similarity of the blueschist outcrops, we suggest Bôrka Nappe as the most possible provenance (**Fig. 9.**) of the Baradla Cave blueschist implements. In addition, this is supported by the fact that the blueschist implements are very frequent in archaeological collections of North Hungary while farther away the number of the blueschist artefacts shows decreasing numbers. Blueschist was the most abundant polished stone tool raw material in the Baradla Cave either.

Contact metabasite

Macroscopic description

Five polished flat chisels were identified as contact metabasite. Their colour is black, greyish black, very fine-grained and foliation cannot always be

perceived. The MS values are variable ($0.21\text{--}0.46 \cdot 10^{-3}$ SI), while the 1951.101.10. implement provides $40.38 \cdot 10^{-3}$ SI, an extreme large magnetic susceptibility among the contact metabasites.

Bulk chemistry

Bulk chemistry data of five contact metabasite implements are listed in **App. Table 2.** SiO_2 content ranges in a very narrow interval: 47.78–50.50 wt%. TiO_2 concentration show elevated value with 1.85–3.70 wt% in all contact metabasites.

Representing data in a TAS diagram, all Baradla contact metabasite implements plot to the basalt field and have subalkaline characteristics (**Fig. 10.**), while the AFM diagram describes tholeiitic affinity (**Fig. 11.**).

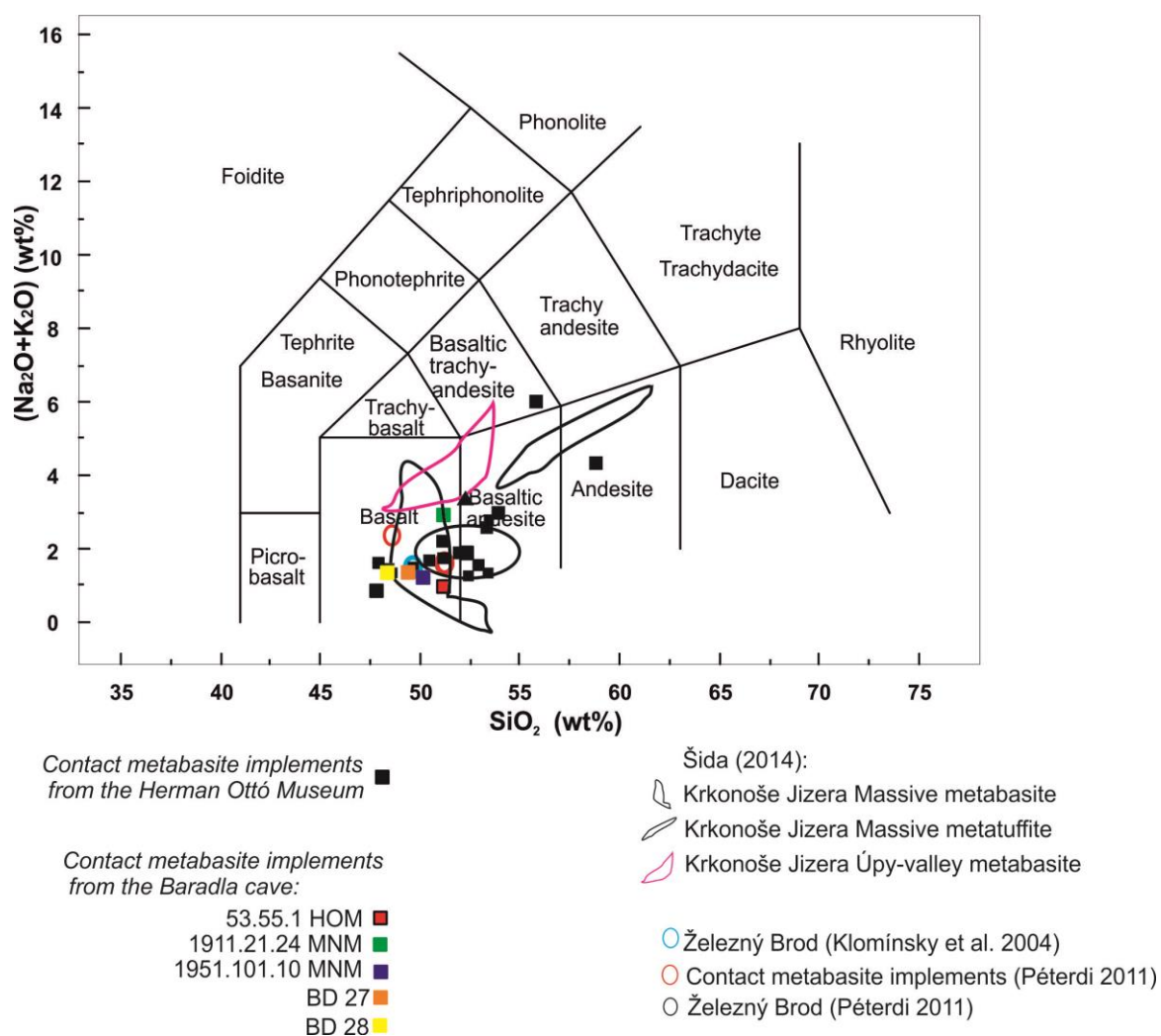


Fig. 10.: Contact metabasite implements from the Baradla Cave compared with previously studied implements from the Herman Ottó Museum (Kereskényi 2021) and field samples (Šida 2014, Klominsky et al. 2004, Péterdi 2011) plotted in TAS diagram (Le Bas et al. 1986).

10. ábra: A Baradla-barlangból előkerült kontakt metabázit kőeszközök kőzetkémiai eredményei TAS diagramon (Le Bas et al. 1986) ábrázolva, összevetve a Herman Ottó Múzeum gyűjteményébe tartozó, korábban vizsgált kőeszközökkel (Kereskényi et al. 2018) és a terepi mintákkal (Šida 2014, Klominsky et al. 2004, Péterdi 2011).

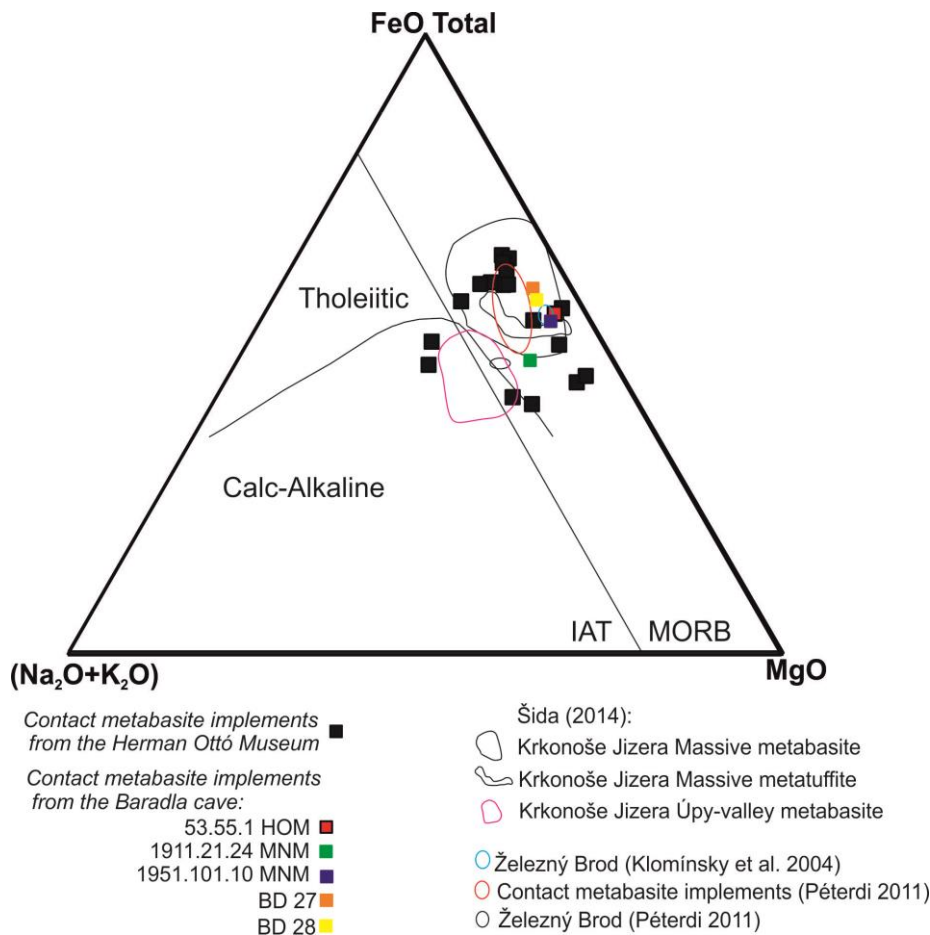


Fig. 11.: Contact metabasite implements from the Baradla Cave compared with formerly studied implements from the Herman Ottó Museum (Kereskényi 2021) and field samples (Šida 2014, Klominsky et al. 2004, Péterdi 2011) plotted in AFM diagram (Irvine & Baragar 1971) diagram. A = alkali ($\text{Na}_2\text{O} + \text{K}_2\text{O}$), F = FeO (total), M = MgO. IAT = island arc tholeiite, MORB = mid-ocean ridge basalt.

11. ábra: A Baradla-barlangból előkerült kontakt metabázit kőeszközök kőzet-kémiai eredményei AFM diagramon (Irvine & Baragar 1971) ábrázolva, összevetve a Herman Ottó Múzeum gyűjteményébe tartozó, korábban vizsgált kőeszközökkel (Kereskényi et al. 2018) és a terepi mintákkal (Šida 2014, Klominsky et al. 2004, Péterdi 2011).

Petrography and mineral chemistry of contact metabasite

As a common feature, the radially oriented acicular amphiboles appear in all five samples. Magnesio-hornblende or magnesio-ferri-hornblende are preserved in the core of amphiboles (Figs. 12/B, /D, /F) with $\text{Al}_{(\text{tot})} = 1.11\text{--}2.09$ apfu (App. Table 5.), while at their rim cummingtonite or grunerite crystallized due to the process of contact metamorphism (Figs. 12/B, D) (App. Table 6.). In the case of 1951.101.10. tool magnesio-hornblende was observed without any Fe-Mg amphibole (Fig. 12/F). At particular rims of magnesio-hornblende actinolite and ferro-actinolite crystallized.

Bulk chemistry data of the raw materials and formerly studied polished stone implements were

compared to the recently studied Baradla tools. Subalkaline and mainly basaltic characteristic describes the geological and other archaeological samples in the TAS diagram (Fig. 10.).

Interpreting the data on the AFM diagram, tholeiitic affinity is the dominant feature both of the polished stone tools and the raw materials (Fig. 11.).

According to the mineralogical, petrological observations and the previously published archaeometric data (Péterdi 2011, Kereskényi 2021), submitted that four contact metabasite polished stone tools occurring in Baradla Cave originate from the Krkonoše Jizera Massif, Železný Brod area, while the raw material of the 1951.101.10. sample derived from the closer Želešice area (Fig. 13.).

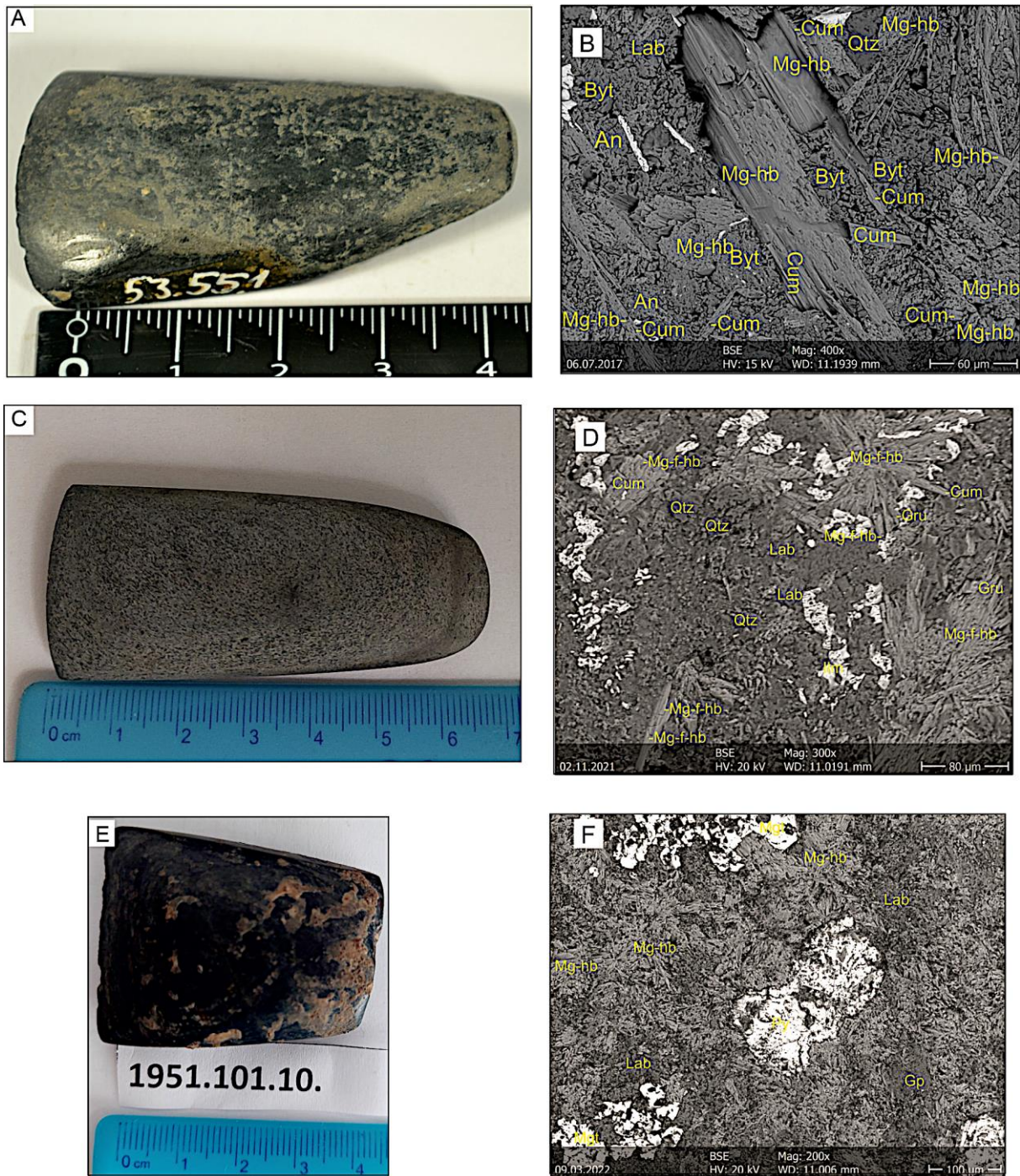
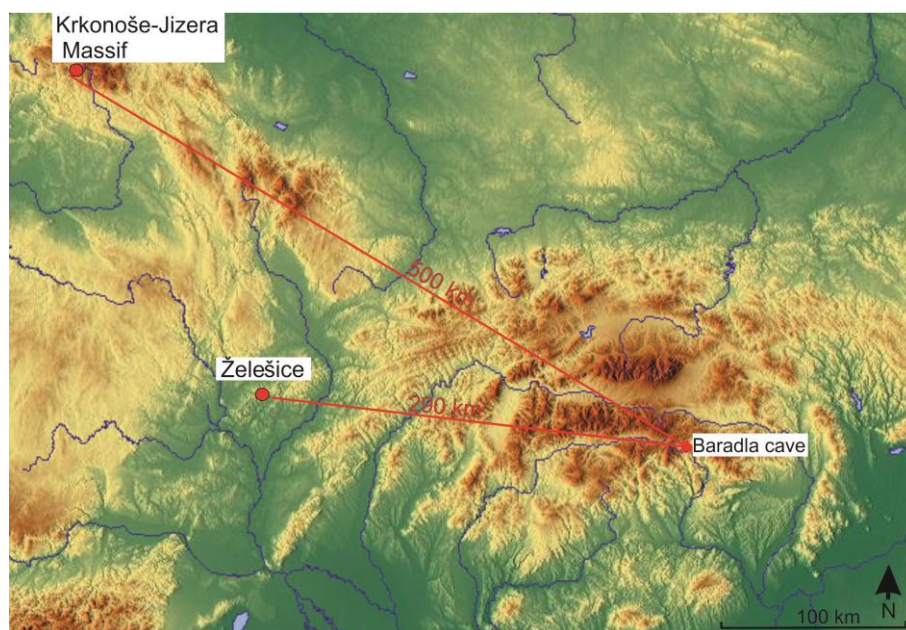


Fig. 12.: Macroscopic (A) and SEM-BSE image of 53.55.1. polished implement, radially oriented acicular amphiboles in contact metabasite, cummingtonite crystallized at the rim of magnesio-hornblende. Macroscopic (C) and SEM-BSE image (D) of BD27 implement; at the rim of magnesio-ferri-hornblende, cummingtonite and its Fe-rich counterpart, grunerite can be observed. Macroscopic (E) and SEM-BSE image (F) of 1951.101.10. sample; radially oriented magnesio-hornblende and large magnetite grains are observed in the 1951.101.10., indicating a Želešice type of contact metabasite.

12. ábra: Makroszkópos (A) és SEM-BSE kép az 53.55.1. jelű csiszolt kőeszkörről. Radiálisan orientált amfibolok kontakt metabázitban, cummingtonit a magneziohornblende szegélyén. Makroszkópos (C) és SEM-BSE kép a BD27 jelű csiszolt kőeszkörről. A magnezioferrihornblende szegélyén cummingtonit és annak vasgazdag párja, a grünerit figyelhető meg. Makroszkópos (E) és SEM-BSE kép (F) az 1951.101.10. jelű csiszolt kőeszkörről. Radiálisan szétseprűződő magneziohornblende és nagy magnetit szemcsék, melyek a Želešice-típusú kontakt metabázitot valószínűsítik.

**Fig. 13.:**

Contact metabasite derived as a long-distance raw material from the Krkonoše Jizera Massif and Želešice to the Baradla Cave.

13. ábra:

A kontakt metabázit távoli nyersanyagként érkezett a Baradla-barlanghoz, ugyanis a forrásterület Krkonoše Jizera Masszívum, illetve Želešice környéke.

Greenschist-amphibolite

Three polished stone tools were sorted to the greenschist-amphibolite rock type. The name is indicated because only XRD analyses were carried out of two implements therefore their textural relations cannot be observed to determine the precise lithotype.

Macroscopic description

1939.36.5. and 1984.9.1. are flat chisels (**Fig. 14/A**), while 1998.1.26 is a semi-finished artefact.

1939.36.5. sample is covered with a thick coating originating the burial process, although in some spots the greenish colour can be observed. Magnetic susceptibility of $0.48 \cdot 10^{-3}$ SI was measured for this implement. 1984.9.1. is very fine-grained, black coloured, and having a higher magnetic susceptibility of $7.20 \cdot 10^{-3}$ SI.

1998.1.26. implement has white and green foliation bands in the texture; its magnetic susceptibility is $0.42 \cdot 10^{-3}$ SI.

Bulk chemistry of greenschist-amphibolite implements

Bulk compositional data of the greenschist-amphibolite implements are listed in **App. Table 7**. The tools are characterized by a narrow SiO₂ content interval ranging from 46.20 to 48.07 wt%.

Representing the data in a TAS diagram, the analysed samples plot in the basalt field having

subalkaline characteristics, since the Na₂O + K₂O content varies between 3.03 and 3.24 wt% (**Fig. 15.**). In the AFM diagram most of the samples show tholeiitic affinity (**Fig. 16.**).

Petrography and mineral chemistry of 1984.9.1. greenschist

40–120 μm actinolite and magnesio-hornblende with low Al content (**App. Table 5.**) were described from the sample as rock-forming amphiboles. The plagioclases correspond to albite (An_{4.01-4.07}). Epidote, clinozoisite, clinocllore are observed in large amount representing the greenschist metamorphic facies. The stone implement contains titanite and cc. 300 μm magnetite grains as accessory phases (**Fig. 14/B**). These large magnetite grains cause the high magnetic susceptibility value of the tool.

XRD analyses

Only XRD analyses were accomplished both on 1998.1.26. and 1939.36.5. inventory numbered implements, because of their large size, do not fit into the sample chamber of the electron microscope.

Actinolite, albite, epidote and quartz were recorded from the 1998.1.26. sample (**Fig. 17.**), while magnesio-hornblende, chlorite, albite, andesine, quartz, illite and gypsum phases were detected from 1939.36.5. implement (**Fig. 18.**). Illite and gypsum may have deposited to the surface during the burial process, as a thick coating on it; although illite may have come from the weathering of feldspars either.

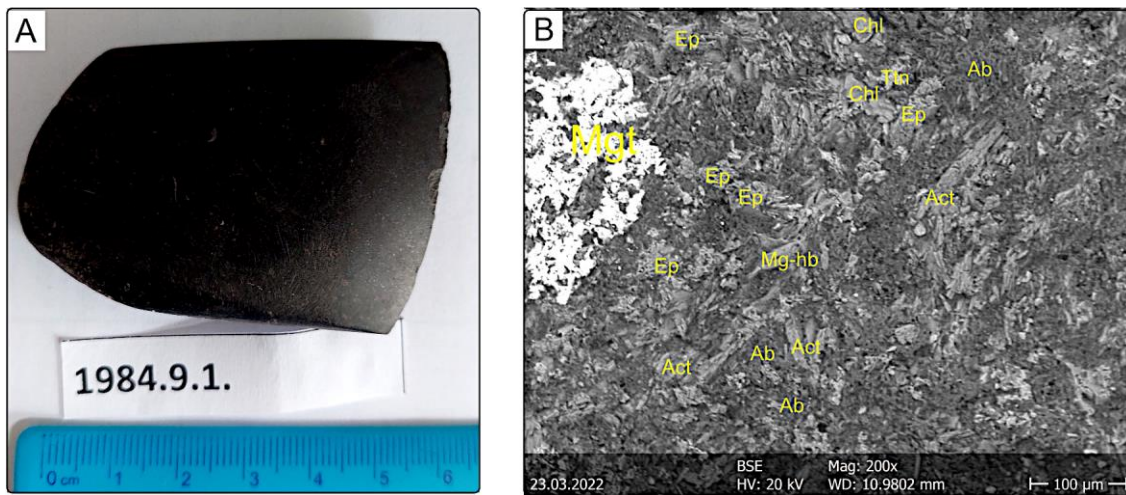


Fig. 14.: Macroscopic picture (A) and SEM-BSE image (B) of 1984.9.1. greenschist-amphibolite implement.
14. ábra: Makroszkópos (A) és SEM-BSE kép (B) az 1984.9.1. jelű zöldpala-amfibolit csiszolt kőszekéről.

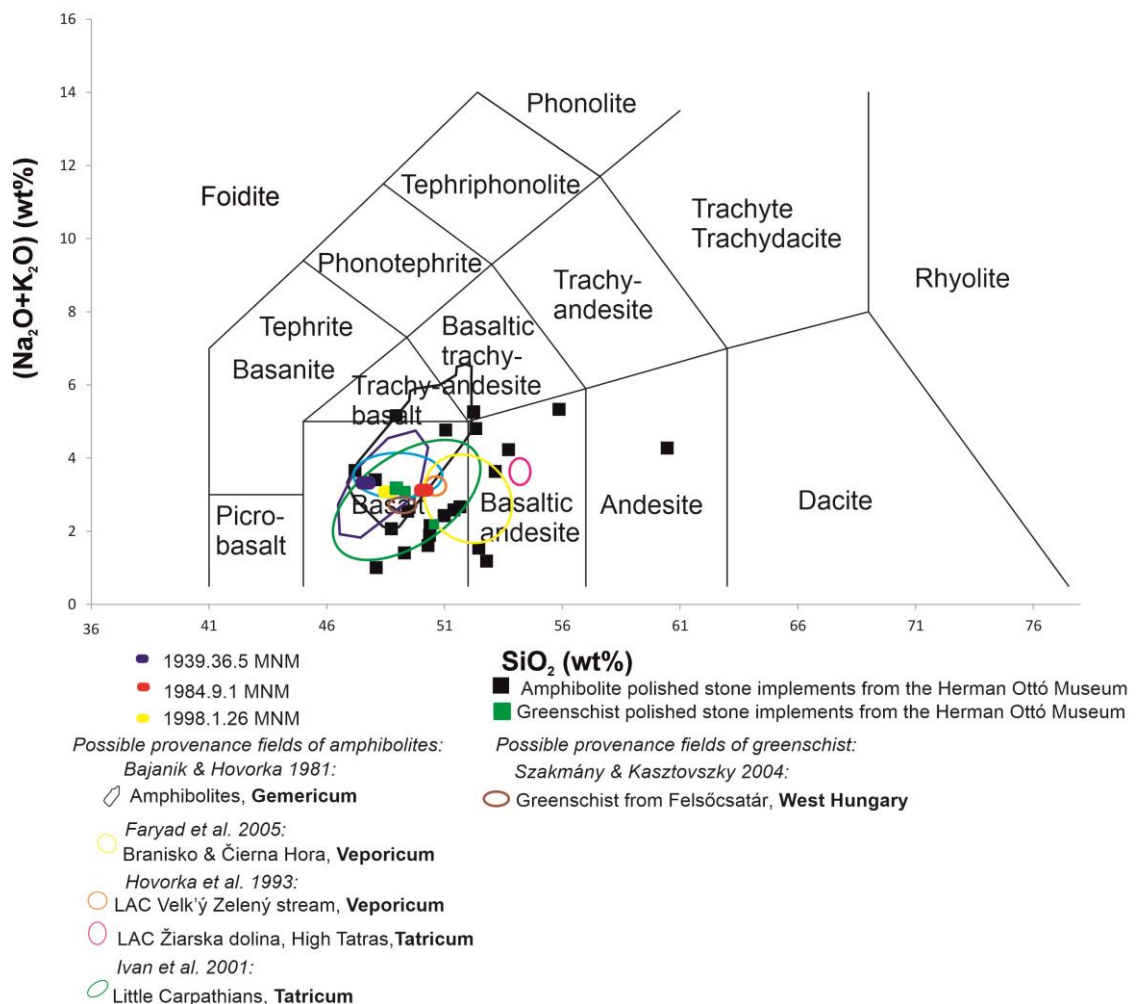


Fig. 15.: Greenschist-amphibolite implements from the Baradla Cave compared to the implements from the collection of the Herman Ottó Museum and the possible provenances plotted in the Total Alkali-Silica (TAS) diagram (Le Bas et al. 1986).

15. ábra: A Baradla-barlangból előkerült zöldpala-amfibolit kőszeközök kőzetkémiai eredményei TAS diagramon (Le Bas et al. 1986) ábrázolva, összevetve a Herman Ottó Múzeum zöldpala-amfibolit csiszolt kőszekőzeivel és a lehetséges forrásterület adataival.

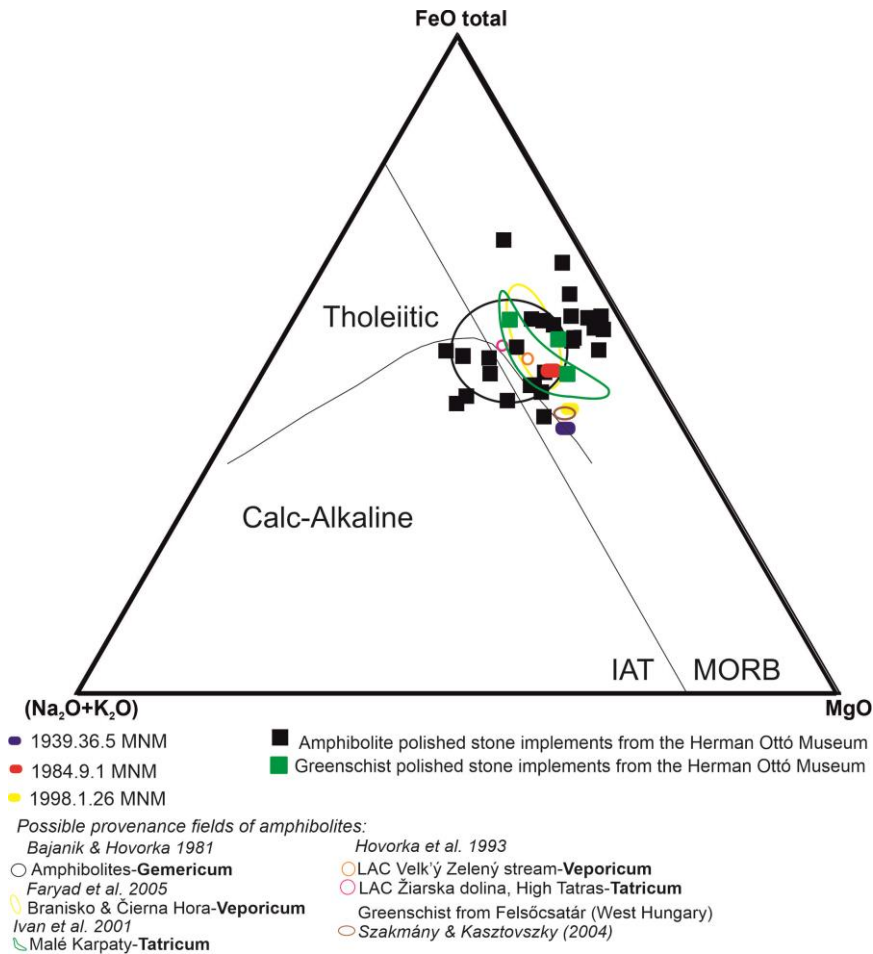


Fig. 16.:

AFM diagram of the investigated greenschist-amphibolite implements of the Baradla Cave compared to the implements from the collection of Herman Ottó Museum and their possible field localities plotted in AFM (Irvine & Baragar 1971) diagram. A = alkali ($\text{Na}_2\text{O} + \text{K}_2\text{O}$), F = FeO (total), M = MgO. IAT = island arc tholeiite, MORB = mid-ocean ridge basalt.

16. ábra:

A Baradla-barlangból előkerült zöldpala-amfibolit kőszeközök kémiai eredményei AFM diagramon (Irvine & Baragar 1971) ábrázolva, összevetve a Herman Ottó Múzeum zöldpala-amfibolit csiszolt kőszeközével és a lehetséges forrásterület adataival.

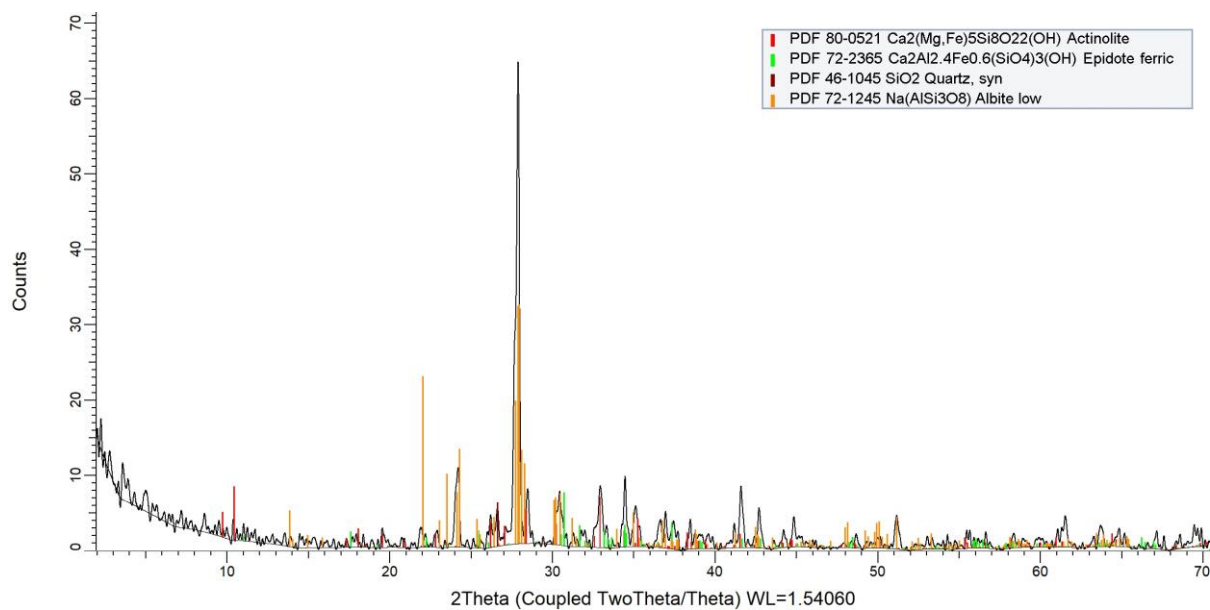


Fig. 17.: XRD pattern of 1998.1.26. greenschist-amphibolite tool.

17. ábra: 1998.1.26. jelű zöldpala-amfibolit kőszeköz XRD felvétele.

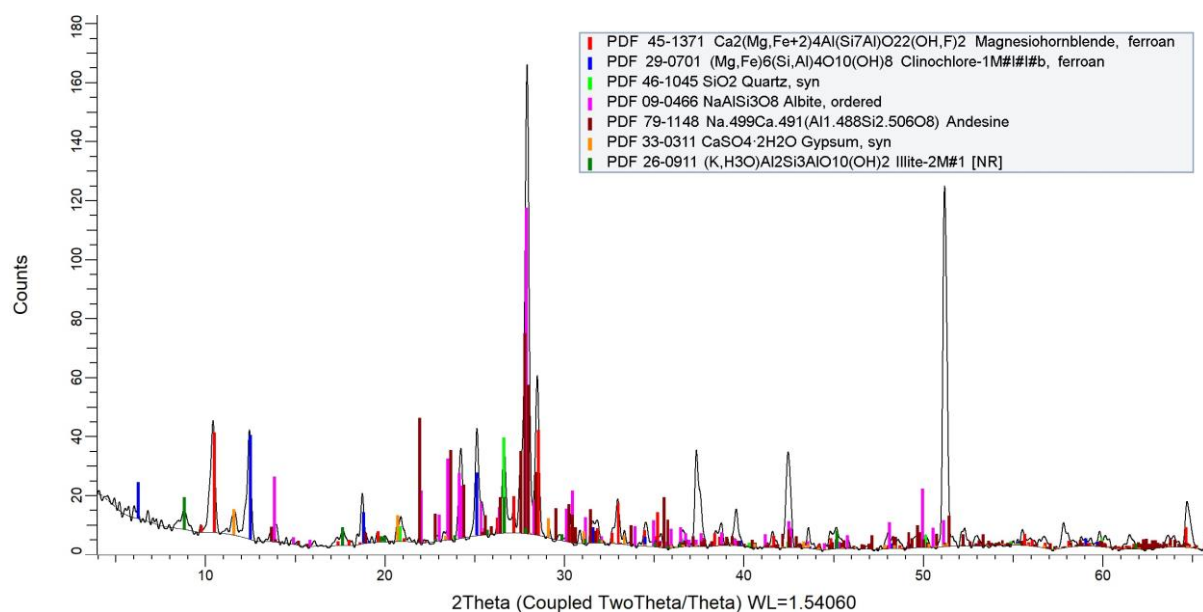


Fig. 18.: XRD pattern of 1939.36.5. greenschist-amphibolite implement.

18. ábra: 1939.36.5. jelű zöldpala-amfibolit köeszköz XRD felvétele.

Possible provenance of amphibolite-greenschist implements

Amphibolite and greenschist is a very common raw material of polished stone tools in the northern part of Carpathian basin in the Neolithic era (Hovorka et al. 2001, Méres et al. 2001, Přichystal 2013, Kereskényi et al. 2020). In addition, many geological amphibolite and greenschist outcrops expose in large amounts in the Western Carpathians (e.g. Plašienka et al. 1997). Distinguishing them on the base of main elemental bulk chemistry is challenging because those overlap both at the TAS and AFM diagrams (**Figs. 15., 16.**).

Since only X-ray diffraction analyses were performed on two stone tools, only the greenschist-amphibolite rock group can be laid down from the mineral chemistry and the macroscopic appearance.

Greenstones and amphibolites with high magnetite content were described from the Little Carpathians (Ivan et al. 2001) and the Slovak Ore Mountains (Faryad & Peterec 1987). Therefore, the raw material of the 1984.9.1. polished stone tool might originate from these territories.

In formerly studied amphibolite polished stone tools, thermobarometric estimations helped to identify possible provenance (Kereskényi et al. 2020) but due to the limitations of non-destructive methods, the metamorphic evolution cannot be reconstructed from the data obtained, so we do not attempt a more definite provenance identification.

Other rock types

This chapter involves those rock types which are difficult to be determined for more reasons. The

non-destructive methods are not sufficient enough to collect mineralogical and petrological data or the number of such artefacts is low in Hungary or those are widespread lithotypes with plentiful possible source areas.

Serpentinite

A very fine-grained, black coloured, 1948.37.37. inventory numbered flat chisel (**Fig. 19/A**) with typical high magnetic susceptibility value (MS: $52.71 \cdot 10^{-3}$ SI) was identified as serpentinite.

The bulk rock chemistry provided by PGAA reveal low SiO₂, high MgO and significant H₂O content, which refers to an ultrabasic rock (**App. Table 7.**).

Presence of massive fibrous serpentine minerals and cc. 20-300 μm magnetite grains were observed in the BSE images (**Fig. 19/B**).

Serpentinite is a rare rock type; as only four of the cc. 500 polished stone tools of the Herman Ottó Museum were found to be serpentinite. This observation is also supported by previous archaeometric studies in Hungary, in which serpentinite stone implements are mainly concentrated in the Transdanubian region and only a small number of them appear in archaeological collections (Szakmány 2009, Péterdi 2011).

At the surroundings of the Carpathian Basin there are several significant occurrences of serpentinites: Eastern Alps, Sowie Góry, Czech Massif, Vardar Belt, and around Dobšiná (Szakmány 2009). Although Dobšiná (Slovakia) is the closest serpentinite outcrop to the Baradla Cave, because of its rock quality, previously it has been excluded as a possible source (Hovorka & Illášová 1995).

The small number of serpentinite stone tools suggests that it is not a widespread rock type, probably the low hardness of serpentinite limited its suitability for polishing stone implements. Due to this fact and the scarcity of archaeometric data, the archaeometric processing of serpentinite as a raw material type is still in its initial phase. Comparison of mineralogical and petrological data to possible source areas at this stage of the research is not yet possible.

Sandstone hoe

A large, slightly curved implement with 1911.21.28 inventory number is named hoe in the inventory book of the Hungarian National Museum. One end is flattened, while the other is oval. The oval end must have been used for hoeing, with a shard of use that broken off (Fig. 20.).

Macroscopically the hoe has brown colour, only quartz crystals can be recognized by naked eye. The magnetic susceptibility is low: $0.06 \cdot 10^{-3}$ SI.

PGAA bulk rock chemistry shows high SiO₂ values with 84.41 wt%, moreover large concentration of Al₂O₃ also occurs (App. Table 7.).

The mineral phases detected by XRD are quartz, albite, calcite and gypsum (Fig. 20.). The latter two

mineral phases may have been deposited on the stone tool during the burial process.

The macroscopic appearance and mineral composition suggest a sandstone lithotype. Several sandstone excavations and quarries are known in Northeast Hungary, Slovakia and Ukraine close to the archaeological site. At present, based on the mineralogical and petrological data, identification of the source area is impossible in the absence of a diagnostic mineral.

Limestone white handstone

1929.64.75. inventory numbered white handstone (Fig. 21.) which was denominated from the inventory book of the Hungarian National Museum coming from a 1929 excavation, the precise archaeological locality is not mentioned, but three localities from the Baradla Cave (Bat branch, Fox branch or Bone hall) are listed in the inventory book. Magnetic susceptibility is 0.00.

The major chemical composition of the stone tool is 52.55 wt% CaO and 47.26 wt% CO₂ (App. Table 7.). In the implement the only phase is calcite recorded by XRD (Fig. 21.).

The host rock of the Cave covers three limestone formations and a precise identification might be performed by palaeontological studies.

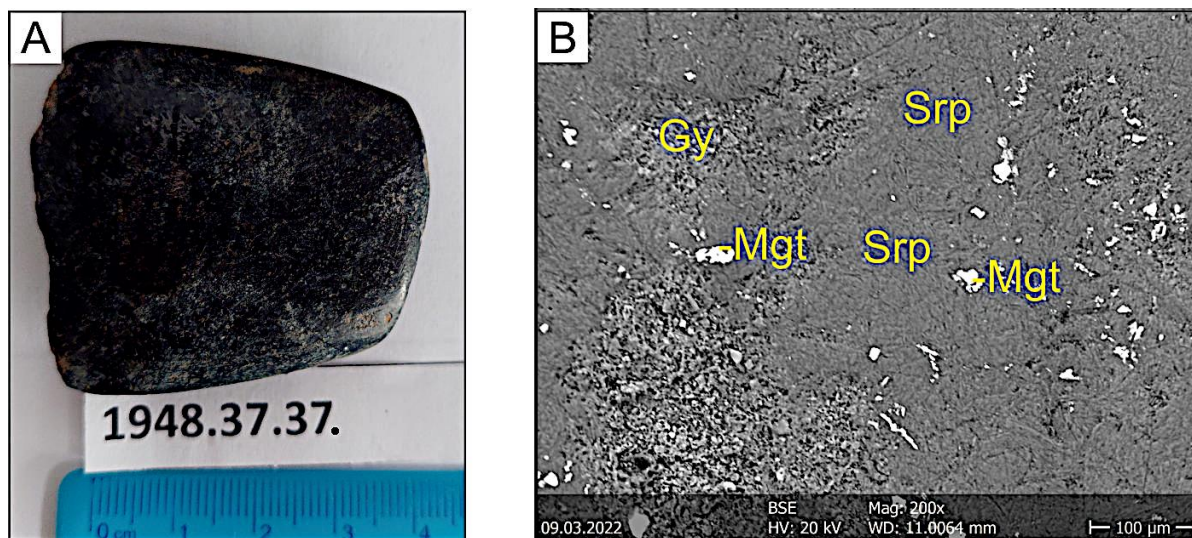


Fig. 19.: Macroscopic (A) and SEM-BSE image (B) of 1948.37.37. serpentinite implement. Masses of fibrous serpentinite bundles with magnetite.

19. ábra: Makroszkópos (A) és SEM-BSE (B) kép az 1948.37.37. jelű szerpentinít csiszolt kőszkőzről. A szerpentinkegerek tömegesen jelennek meg magnetitvel.

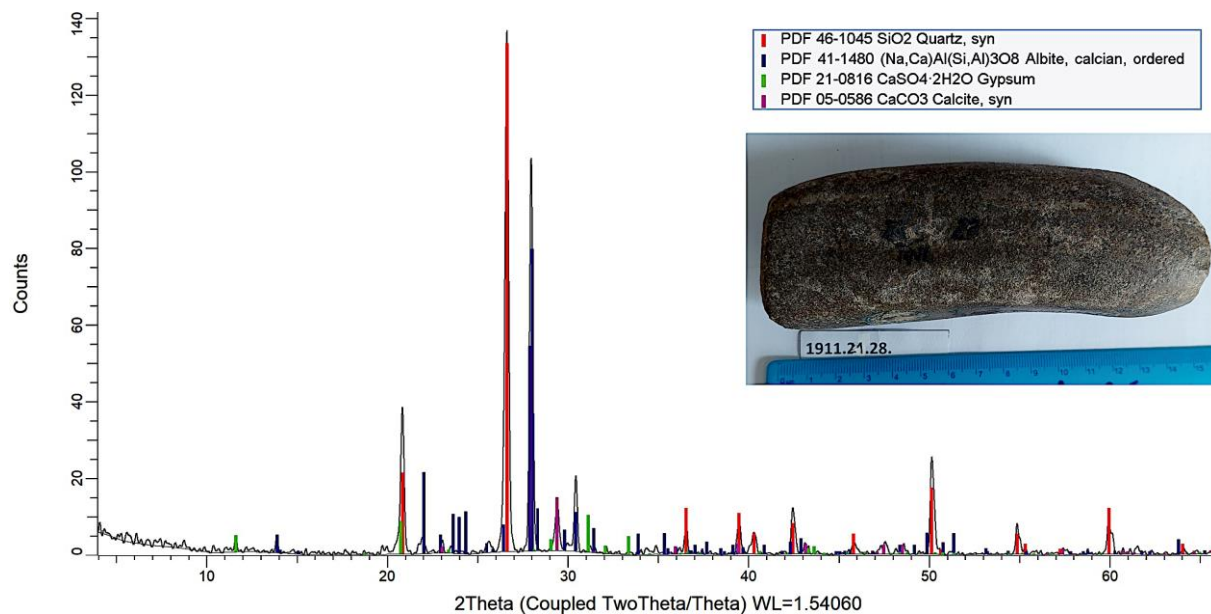


Fig. 20.: Macroscopic image and XRD pattern of 1911.21.28. sandstone implement.

20. ábra: Az 1911.21.28. jelű homokkő nyersanyagú kőeszköz makroszkópos képe és XRD felvétele.

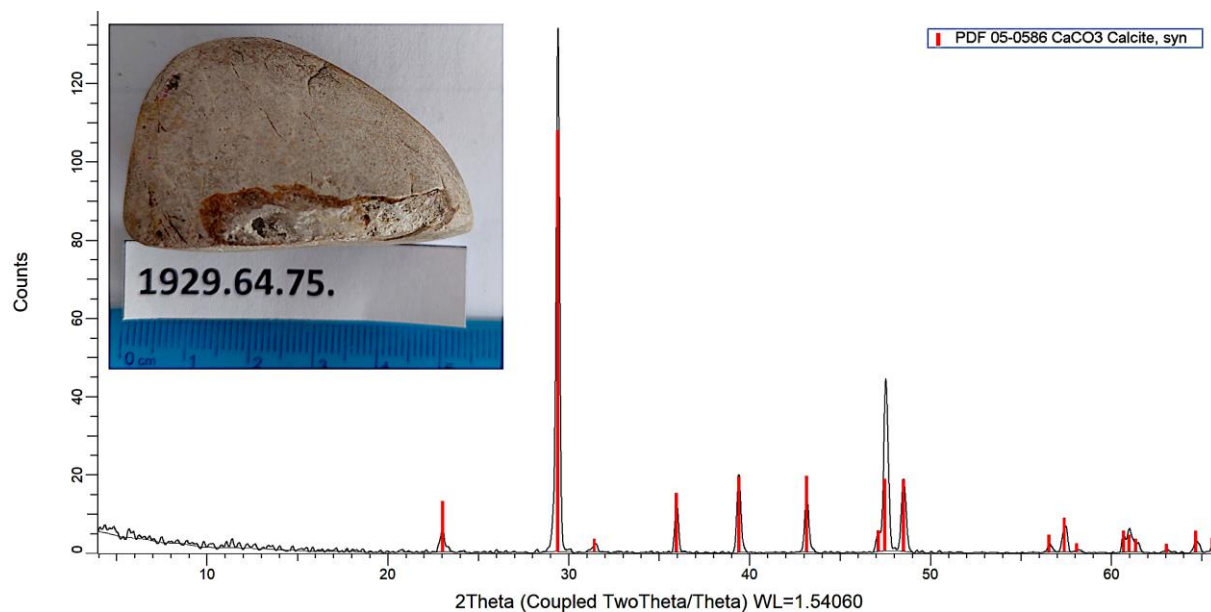


Fig. 21.: Macroscopic image and XRD pattern of 1929.64.75. limestone implement.

21. ábra: Az 1929.64.75. jelű mészkő kőeszköz makroszkópos képe és XRD felvétele.

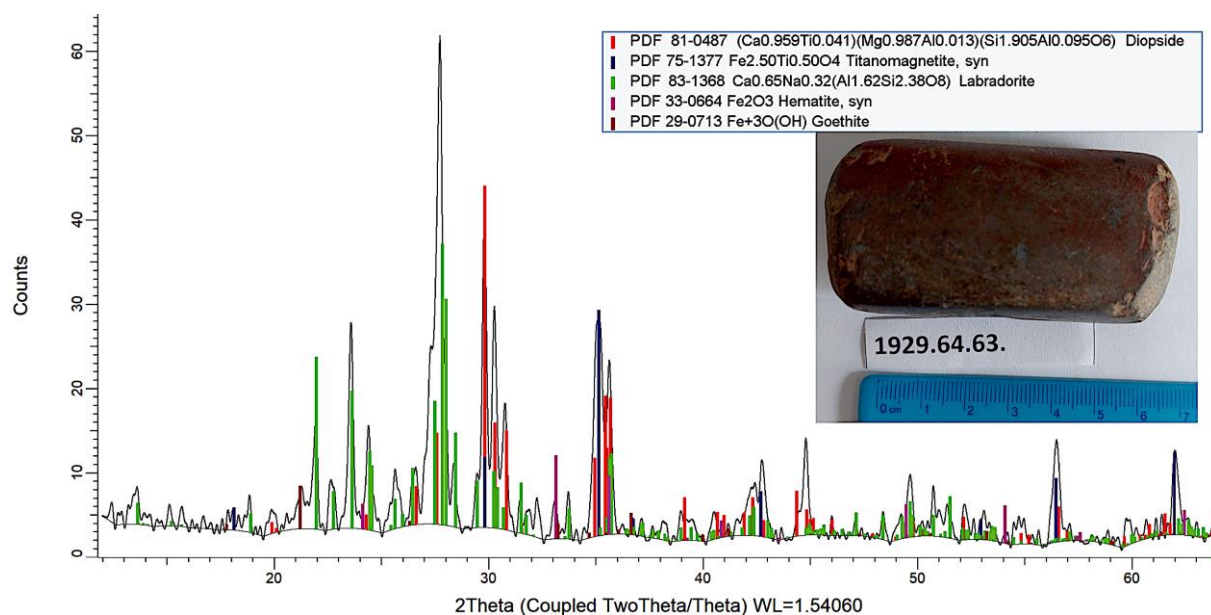


Fig. 22.: Macroscopic image and XRD pattern of 1929.64.63. basalt stone implement.

22. ábra: Az 1929.64.63. jelű bazalt nyersanyagú kőeszköz makroszkópos képe és XRD felvétele.

Basalt polished stone tool

1929.64.63. inventory numbered, a larger flat polished stone implement (**Fig. 22.**) originating also from the 1929 excavation with the same three possible localities as in the case of the previously mentioned artefact.

The surface of the stone axe shows traces of red staining, otherwise it is fine-grained, its magnetic susceptibility is $12.71 \cdot 10^{-3}$ SI. According to the bulk rock chemistry data it plots to the trachybasalt field in the TAS diagram (**App. Table 7.**).

XRD analysis confirmed the presence of goethite and hematite which cause the red colour of the surface. In addition, diopside, labradorite and titanomagnetite have been detected in the implement (**Fig. 22.**).

The mineral association and the geochemical data suggest basalt rock-type.

Füri et al. (2004) made suggestions for differentiation of basalts. The Baradla basalt implement does not correspond to any Hungarian basalts investigated by Füri et al. (2004) by comparing the Gd vs Sm and $\text{Na}_2\text{O}/\text{K}_2\text{O}$ vs $\text{TiO}_2/\text{Al}_2\text{O}_3$ data either. We assume that in the strongly altered basalt alkali might have mobilised, Na depleted, while the K preserved which can cause the low $\text{Na}_2\text{O}/\text{K}_2\text{O}$ ratio.

The fact of red staining on the surface of the tool, and the presence of hematite and goethite yielded by the XRD may influence the Fe_2O_3 content, therefore comparing the bulk chemistry to other basalt data should be treated with caution.

Hence SEM-EDS investigation is needed to get more information about the rock forming minerals and textural relations to compare to possible raw materials.

Conclusion

20 polished stone implements were analysed with non-destructive methods from the Baradla Cave. Summarizing the results, blueschist is the most abundant lithotype, which is already well-known raw material in the archaeological collections of North Hungary. Its provenance can be found nearby, in Bôrka Nappe (Southeast Slovakia), in 30–50 km distance from the archaeological locality.

The second common rock-type is the contact metabasite, a mechanically well-behaving lithotype, which was widespread in the Neolithic Central Europe. The long-distance raw materials are originated from Želešice and Krknoše Jizera Crystalline Unit, near Železný Brod, Czech Massif, meaning about 500 km trade route to the archaeological locality.

Three implements were sorted to the greenschist-amphibolite rock-type. The applied original surface method of SEM-EDS, XRD and PGAA are not sufficient enough to provide relevant results to determine precise provenance of greenstones. In a previous paper, thermobarometry was additionally applied to constrict the provenance of the greenstone implements which resulted to determination of different parts of the Western Carpathians (Kereskényi et al. 2020, Kereskényi 2021). In the case of 1984.9.1. polished stone tool, significant magnetite content was observed,

therefore the Little Carpathians (Ivan et al. 2001) or the Slovak Ore Mountains (Faryad & Peterec 1987) might be concerned as a source area, meaning a nearby provenance field.

A few pieces of archaeometrically not well-processed lithotypes (serpentinite, sandstone, limestone, basalt) were identified from the Baradla Cave. Furthermore, the approximate cultural period of the latter three stone implements is also questionable, because their shapes deviate from the other flat implements. On the basis of the limited information available, no precise source area can be determined, and further exploration is recommended both on the archaeological finds and possible raw materials.

Mineral abbreviations applied in the paper: Ab: albite, Act: actinolite, An: anorthite, Aug: augite, Bar: barroisite, Bs: blueschist, Byt: bytownite, Chl: chlorite, Cm: contact metabasite, Cum: cummingtonite, Czo: clinozoisite, Ep: epidote, F-win: ferro-winchite, F3-win: ferri-winchite, Fgln: ferro-glaucophane, Gln: glaucophane, Gru: grunerite, Gp: gypsum, Ilm: ilmenite, Lab: labradorite, Mg-hb: magnesio-hornblende, Mg-f-hb: magnesio-ferri-hornblende, Mgt: magnetite, Omp: omphacite, Ph: phengite, Py: pyrite, Qtz: quartz, Srp: serpentinite, Ttn: titanite, Win: winchite.

Contribution of authors

Kereskényi Erika Conceptualization, Methodology, Validation, Investigation, Resources, Data Curation, Writing – Original draft, Visualization, Writing – Review and Editing, Visualization. **Fehér Béla** Methodology, Validation, Investigation, Resources, Writing – Original draft, Supervision. **Kristály Ferenc** Methodology, Validation, Investigation, Resources, Writing – Original draft, Visualization. **Szilágyi Veronika** Investigation, Writing – Original draft. **Kasztovszky Zsolt** Methodology, Validation, Resources, Investigation, Writing – Original draft, Visualization, Funding acquisition. **Szakmány György** Conceptualization, Writing – Original draft, Supervision.

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