ARCHAEOMETRIC INVESTIGATION OF AN ALKALI BASALT NEOLITHIC POLISHED STONE TOOL FROM NAGY-FERTŐ LOCALITY, NEAR MEZŐKÖVESD (BORSOD-ABAÚJ-ZEMPLÉN COUNTY, NE HUNGARY)•

ALKÁLI BAZALT NEOLIT CSISZOLT KŐESZKÖZ ARCHEOMETRIAI VIZSGÁLATI EREDMÉNYEI MEZŐKÖVESD, NAGY-FERTŐ LELŐHELYRŐL (BORSOD-ABAÚJ-ZEMPLÉN MEGYE, MAGYARORSZÁG)

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Abstract

A Neolithic alkali basalt polished stone tool originated from Nagy-Fertő locality, near Mezőkövesd, was studied from archaeometric aspect. Its mineral association was detected by EDS/SEM and XRD. The decisive mineral components of the stone axe are olivine, clinopyroxene, labradorite, sodalite, natrolite and spinel. Data of bulk chemistry WDXRF analyses were compared with previously published data on similar raw materials and it shows a good match with the stone axe and the alkali basalts from Bulhary, Ceres Mountains. As other sodalite-bearing basalts are not known from the Carpathian basin and its surroundings and according to the similar mineralogical assemblage of the stone axe and the alkali basalt raw material, in addition the bulk chemistry data confirmed the presumption of the possible provenance field is Bulhary in the Ceres Mountains (Slovakia).

Kivonat

Egy alkáli bazalt nyersanyagú neolit csiszolt kőeszköz archeometriai vizsgálati eredményeit mutatjuk be Mezőkövesd, Nagy-Fertő lelőhelyről. A kőeszköz fő kőzetalkotó ásványai: olivin, klinopiroxén, labradorit, szodalit, nátrolit és spinell. Az EDS/SEM és XRD vizsgálatok eredményei nagy hasonlóságot mutattak a Cseres-hegységben (Szlovákia) előforduló szodalit-tartalmú alkáli bazaltokkal. Elvégeztük a bolgáromi kőbányából származó kőzetminta EDS/SEM, XRD elemzését is, továbbá kőzetkémiai (WDXRF) vizsgálatok is történtek mindkét mintán, amelyek megerősítették a hasonlóságot. A Kárpát-medence környékéről más szodalit-tartalmú alkáli bazalt nem ismert. Ez a tény, illetve az ásvány- és kőzetkémiai azonosságok is alátámasztják a kőbalta nyersanyagának cseres-hegységi eredetét.

KEYWORDS: ALKALI BASALT, SODALITE, PROVENANCE, HERMAN OTTÓ MUSEUM

KULCSSZAVAK: ALKÁLI BAZALT, SZODALIT, FORRÁSTERÜLET, HERMAN OTTÓ MÚZEUM

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Fig. 1.: Excavation area is signed with green circle in the Borsod-Abaúj-Zemplén county map

1. ábra: A kőeszköz régészeti lelőhelye zöld körrel jelölve a térképen

Fig. 2.: Alkali basalt stone axe (D19 sample) from Mezőkövesd, Nagy-Fertő excavation area

2. ábra: Alkáli bazalt kőbalta (D19 minta) Mezőkövesd, Nagy-Fertő lelőhelyről

Introduction

The archaeological collection of the Herman Ottó Museum contains approximately 500 Neolithic polished stone tools. Most of the axes are metabasites, mainly different types of amphibolites and contact metabasites (Kereskényi et al. 2017). 36 polished stone tools proved to be blueschists (Kereskényi et al. 2018). Lithology of the quarter of the collection has eruptive origin and roughly 50 pieces of stone artefacts are of sedimentary origin. A few axes are determined to be hornfels and some metaultrabasite polished stone tools also occur in the archaeological collection.

There is one piece made from alkali basalt among the eruptive stone tools. In this paper mineralogical and petrological investigations of this alkali basalt polished stone tool are presented and the results are compared to the possible provenance field of the raw material.

Mineral abbrevations applied in the paper: Ol: olivine, Di: diopside, Aug: augite, Lab: labradorite, Ano: anorthoclase, San: sanidine, Nep: nepheline, Ntr: natrolite, Sdl: sodalite, Sme: smectite, Ca: calcite, Usp: ulvospinel, Mgt: magnetite, Ilm: ilmenite, Ap: apatite, Ze: zeolite.

Archaeological background

The archaeological locality of the polished stone tool is Nagy-Fertő excavation area near Mezőkövesd, on the track of M3 motorway, locality No. 76 (**Fig. 1.**). It came to surface with Spondylus findings and Middle Neolithic ceramic fragments belonging to the Szakálhát culture (Csengeri 2013, Gál 2018). The broken implement must have had shoe-last form (**Fig. 2.**). There has not been published archaeometrical work about Szakálhát cultured polished stone implements yet.

Samples and methods

The stone axe is a non-catalogized artefact and belongs to the collection of the Department of Archaeology, Herman Ottó Museum. The alkali basalt sample of Bulhary (Bolgárom) belongs to the collection of the Department of Mineralogy, Herman Ottó Museum (Inventory number: 2011.167, Locality: Bulhary quarry, Collectors: Ferenc Kristály and Sándor Szakáll, Year: 2011).

Analyses were carried out at the Institute of Mineralogy and Petrology, Miskolc University. Macroscopic and microscopic description were made on the stone axe using Zeiss Stemi DV4 stereo microscope. Polished sections were made from the stone tool and the raw material. EDS/SEM analyses were prepared using a JEOL JXA-8600 Superprobe electron-microprobe in energy-dispersive mode. The accelerating voltage was 20 kV and beam current was 20 nA.

Non-destructive X-ray diffraction (XRD) analyses were carried out both on the stone implement and the alkali basalt with Bruker D8 Advance X-ray diffractometer. Parameters of XRD analyses: CuKα source, 40 kV and 20 mA generator settings, parallel beam geometry (Göbel-mirror) Vantec1 position detector (1° window opening degree), 0.1 mm collimator. The exact method of the analyse is resumed in the earlier published papers (Kristály 2014, Kristály & Kereskényi 2016).

Bulk chemistry determined by wavelength dispersive X-ray fluorescence (WDXRF) Rigaku Supermini spectrometer. Parameters of the WDXRF: 200 W air-cooled Pd cathode-tube, 50 kV accelerating voltage, 4.0 mA current, 1.2-1.6 Pa vacuum pressure. In basic circumstances the required sample quantity is 3 g for WDXRF, but due to the unique aspect of stone implement only 0.6 g was available.

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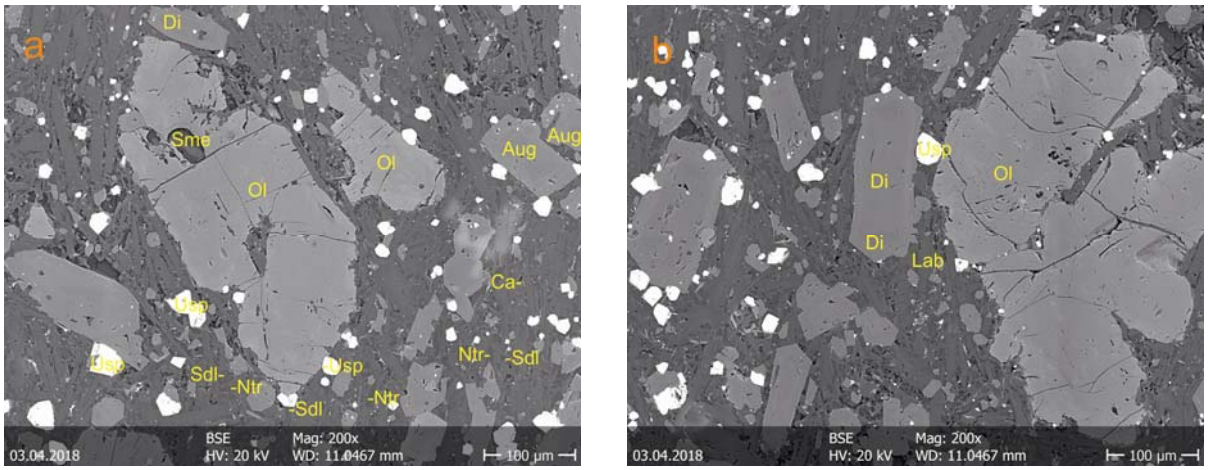

Fig. 3: BSE images of the stone axe

3. ábra: A kőbalta BSE képe

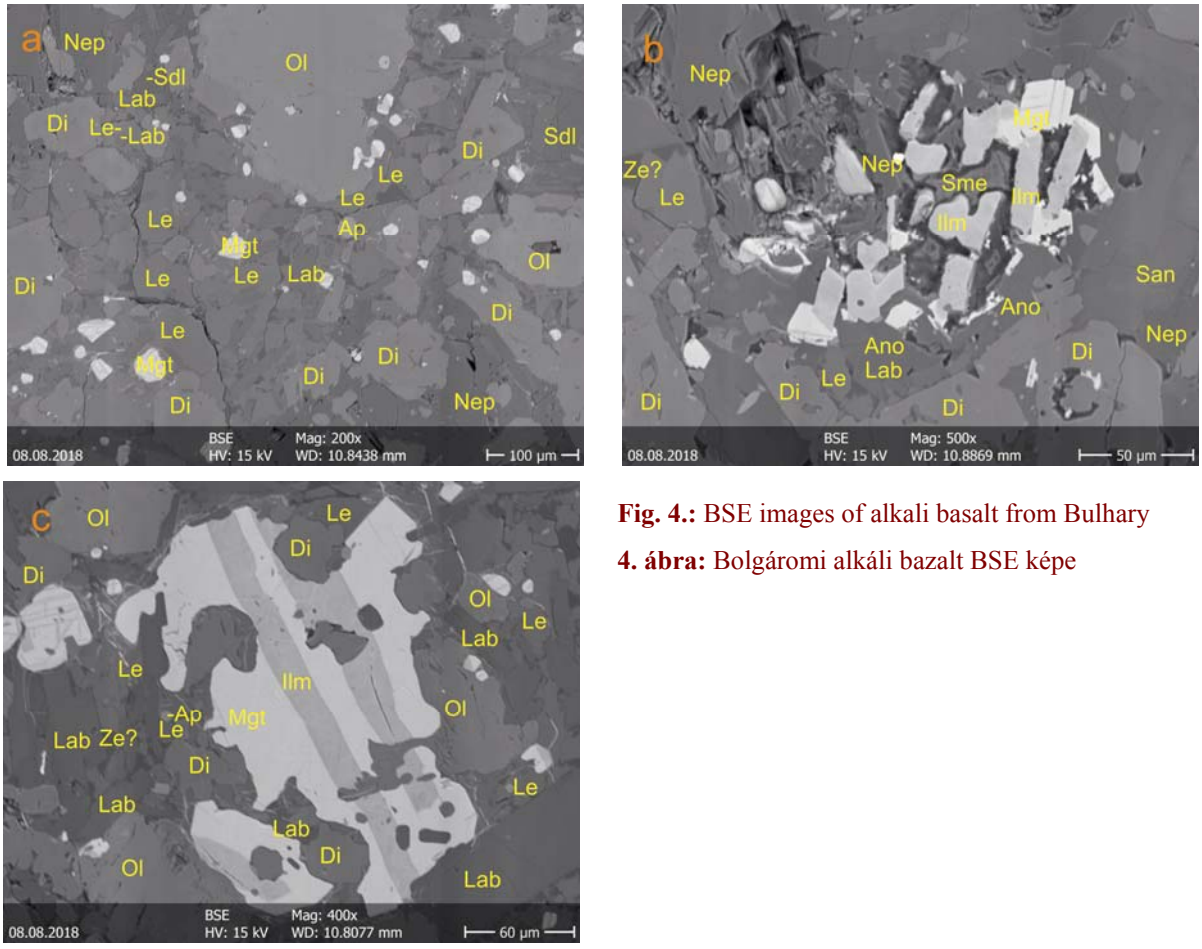

Fig. 4.: BSE images of alkali basalt from Bulhary

4. ábra: Bolgáromi alkáli bazalt BSE képe

Hence 0.6 g sample amount was mixed with 2.4 g analytical purity glass sand in both cases of analysing of the stone implement and the possible source material. In the case of controlled analyses 3 g of alkali basalt was measured.

Geological setting of the provenance field

Alkali basaltic fields located in Cerová Highlands in southeastern of Slovakia. The southern border of Ceres Mountains is the Slovakian-Hungarian state border. In Hungary the Ceres mountain ranges in Karancs, Medves and Heves-Borsod hills (Konečný et al. 2004). The alkaline basaltic volcanism have

developed Ceres Basalt Formation in 5 episodes since 5.5-4 million years forming maars, lava streams, cinder cones, dykes and tuffs (Forgáč 1970, Farsang et al. 2014). The comparison sample was collected from one of the largest quarry, the Bulhary volcanic complex is situated next to Bulhary village.

Results

Macroscopic and microscopic description

The dimensions of the polished stone tool are 3.7 cm x 1.8 cm x 1.5 cm. Macroscopically the stone tool is dark grey and max. 5 mm off-white rounded spots can be recognized in even distribution on its surface and inside the tool (**Fig. 2.**). Along the spots the axe falls apart to blocks, showing the phenomenon of sunburning (Zagożdżon 2003). Due to burial, sporadical light yellow coating can be studied on the surface of the axe.

Microscopically porphyric texture with red and green olivine and black pyroxene phenocrysts can be recognized in the dark, homogenous matrix.

EDS/SEM analyses of the stone axe

EDS/SEM investigation has shown that the size of subhedral olivines can reach the 700 µm. The composition of the olivines varies continuously representing Fe-content increase from core to rim, while the Mg-content decreases and enriches in the core of olivines. From core to rim the fayalite content is 25-34%. Formation of smectites can be observed inside of some olivine crystals (**Fig. 3a**). Zoned euhedral and subhedral pyroxenes are identified and have diopside and augite composition (**Fig. 5.**). Among the feldspars only plagioclase was recognized. Composition of plagioclase corresponds to labradorite (An_{52-58}) (**Fig. 6.**), its size can reach 500 µm. The equant crystals of ulvospinel crystals are up to 40 µm and distributes in the sample equally. Ulvospinel xenoliths can be observed in pyroxenes and olivines. The groundmass contains interstitial sodalite and natrolite crystals sporadically, and sporadical calcite needles. Glass component is not observed in the sample (**Fig. 3a-b**).

EDS/SEM analyses of the Bulhary basalt

Since there is no knowledge of sodalite-bearing basalt in the surroundings of the Carpathian Basin, except is at Bulhary in South Slovakia (Farsang et al. 2014, Fehér et al. 2016), mineral chemistry of alkali basalt from Bulhary quarry was studied for comparison.

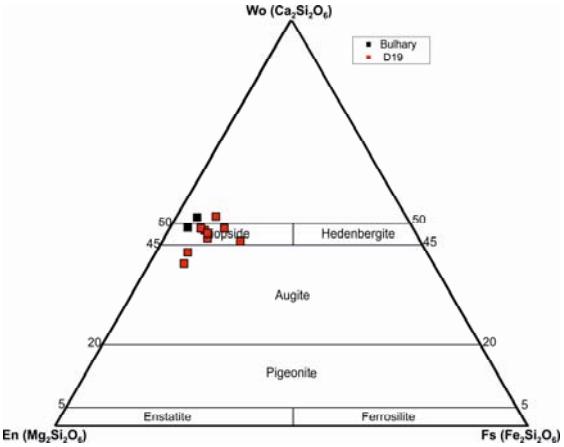

Fig. 5.: Chemical compositions of clinopyroxenes of D19 stone axe and the alkali basalt from Bulhary plotted in the En-Fs-Wo ternary diagram (Morimoto 1989)

5. ábra: Klinopiroxének kémiai összetétele a D19 jelű kőbaltából és a bolgáromi alkáli bazaltból az En-Fs-Wo háromszög diagramon ábrázolva (Morimoto 1989)

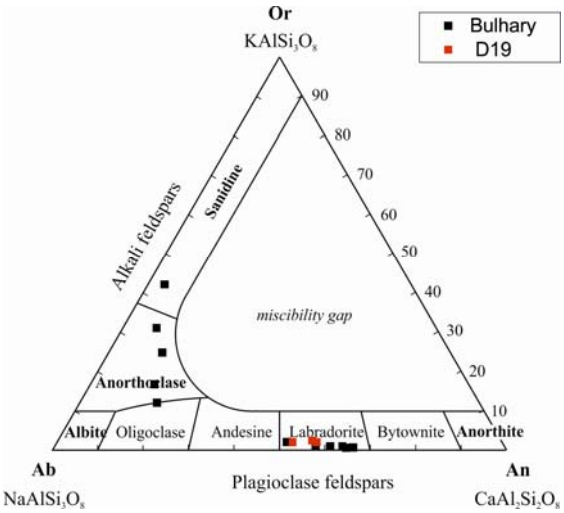

Fig. 6.: Chemical compositions of feldspars of D19 stone axe and the alkali basalt from Bulhary plotted in the Or-Ab-An ternary diagram

6. ábra: Földpátok kémiai összetétele a D19 jelű kőbaltából és a bolgáromi alkali bazaltból az Or-Ab-An háromszög diagramon ábrázolva

The subhedral zoned olivines can reach the 700 µm (**Fig. 4a**). The fayalite-content varies between 24-33% from core to rim. Inside of some olivine crystals formation of smectites can be observed (**Fig. 4b**). The slightly zoned subhedral pyroxenes have diopside composition (**Fig. 5.**). Plagioclases correspond to labradorite (An_{51-67}) composition (**Fig. 6.**), their size is under 100 µm. Alkali feldspars can be observed in the raw material sample (**Fig. 4b**) and according to chemical analyses they fall in anorthoclase and sanidine fields (**Fig. 6.**). The equally distributing Ti-rich

magnetite (TiO_2: 10-14 wt%) forms equant crystals up to 200 µm. Magnetite xenoliths can be observed in pyroxenes and olivines. Ilmenite exsolution can be observed in some magnetites (**Fig. 4c**). Feldspathoids are present as nepheline, leucite and sodalite: nepheline is up to 150 µm, leucite reaches 100 µm (**Fig. 4a**). The largest sodalites occur up to 70 µm and fill the spaces between other crystals (**Fig. 4a**). Apatite has been determined from the sample as an accessory phase (**Fig. 4c**).

XRD analyses

Non-destructive XRD analyses were accomplished both on D19 polished stone tool and the Bulhary basalt. Olivine, diopside, feldspar, sodalite and nepheline were recorded in the D19 stone axe (**Fig. 7.**).Olivine, diopside, feldspar, sodalite, nepheline, leucite and analcime were detected from the alkali basalt (**Fig. 8.**).

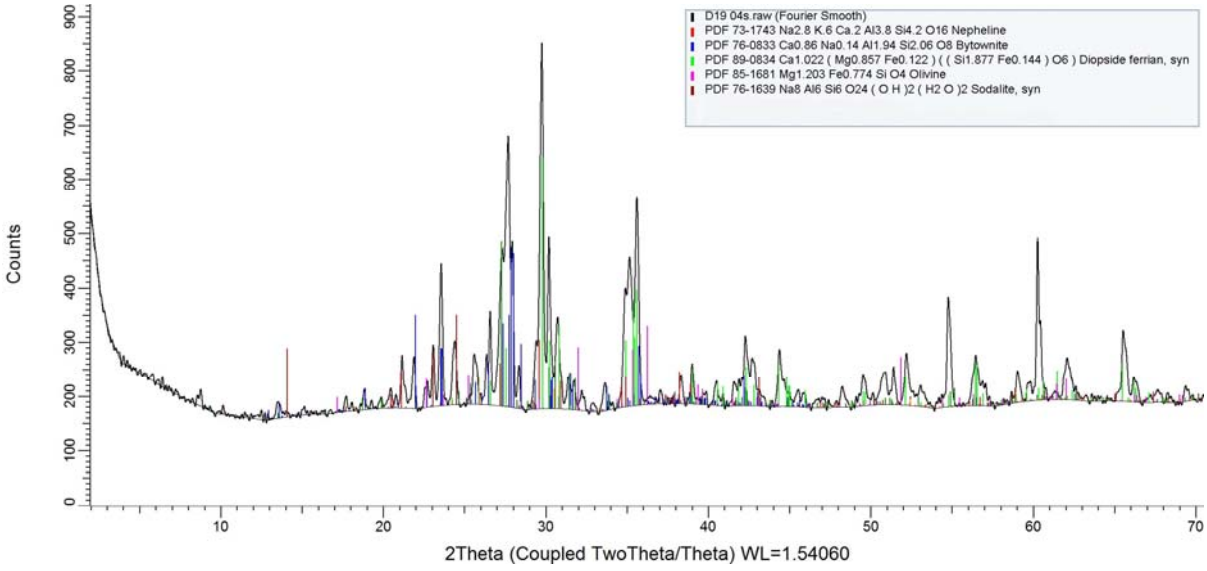

Fig. 7: XRD pattern of D19 stone axe

7. ábra: D19 jelű kőbalta XRD felvétele

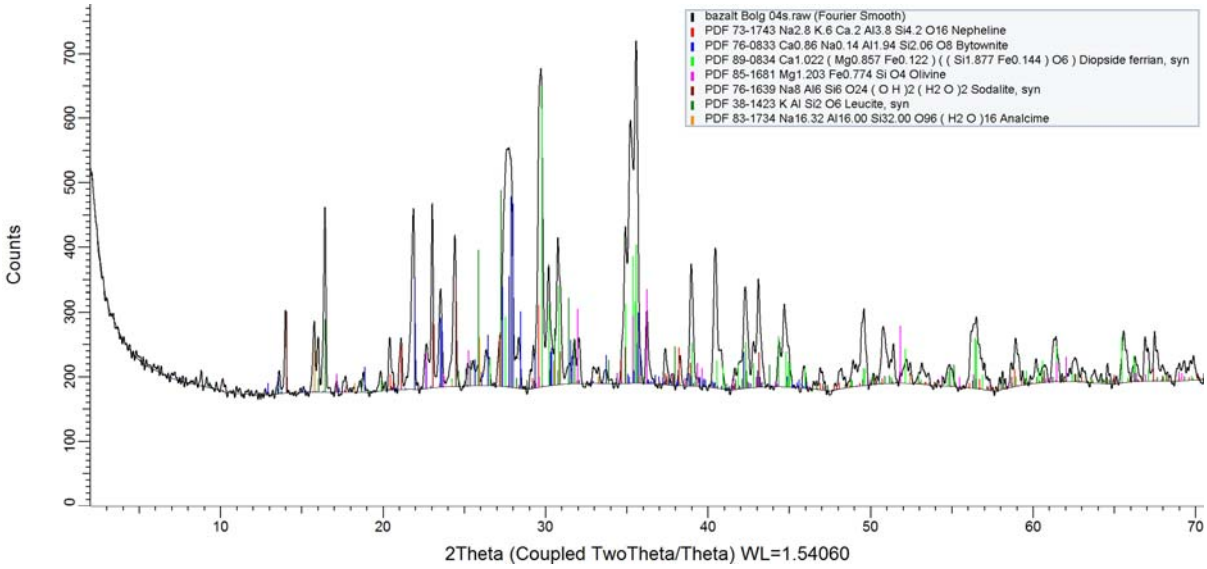

Fig. 8: XRD pattern of Bulhary alkali basalt

8. ábra: Bolgáromi alkali bazalt XRD felvétele

Table 1.: Bulk chemistry results. The major components are given in wt%. The amount of oxides is calculated from the elemental concentration, based on the oxidation numbers

1. D19 stone axe (mixed with glass sand); 2. Bulhary quarry: alkali basalt (mixed with glass sand); 3. Bulhary quarry: alkali basalt; 4. Bulhary: limburgitoide basanite (Forgáč 1970); 5. Konradovce: nepheline basanite (Forgáč 1970) 6. Filakovo: nepheline basanite (Forgáč 1970); 7. Belina: nepheline basanite (Forgáč 1970) 8. Badzovce: nepheline basanite (Forgáč 1970); 9. Borkul-Bagac: nepheline basanite (Forgáč 1970). 10. Average bulk chemistry of basalt fields from North Hungary (Jugovics 1974); 11. Bulhary quarry: basanite (Hakulinová et al. 2011.) 12. Bulhary abandoned quarry: basanite (Hakulinová et al. 2011.)

1. táblázat: Kőzetkémiai eredmények. A főelemek wt%-ban megadva. Az oxidok mennyiségét az elemi koncentrációból számoltuk az oxidációs számok alapján

Sample	1.	2.	3.	4.	5.	6.	7.	8.	9.	10.	11.	12.
SiO_2	no data	no data	46.2	45.53	44.92	44.23	48.53	46.53	42.75	46.62	45.60	46.04
TiO_2	2.25	2.12	1.80	2.70	1.88	2.66	1.88	1.71	1.93	1.75	2.24	2.25
Al_2O_3	no data	no data	15.3	10.76	16.59	15.87	16.03	18.21	14.42	17.53	16.73	16.47
Fe_2O_3*	10.55	9.35	7.36	14.15	10.45	11.44	11.62	8.93	9.82	9.62	9.43	9.05
MnO	0.22	0.21	0.16	0.20	0.24	0.17	0.26	0.32	0.32	no data	0.18	0.17
MgO	5.3	6.85	5.03	8.60	9.03	7.21	7.32	6.34	7.86	5.50	8.55	8.62
CaO	8.75	10.4	9.06	11.07	10.70	11.01	9.04	8.69	12.79	9.51	9.72	9.93
Na_2O	5.75	5.95	5.05	3.27	3.56	3.15	3.64	5.09	2.51	4.14	4.05	4.08
K_2O	2.95	3.25	2.23	1.83	2.10	1.30	1.36	2.90	1.57	2.19	2.42	2.30
P_2O_5	0.51	0.57	0.53	0.00	0.12	0.00	0.17	0.51	0.26	no data	0.60	0.56
S	0.08	0.05	0.01	no data	no data	no data	no data	no data	no data	no data	0.01	0.04

* Total Fe as Fe_2O_3.

Bulk chemistry of the rock samples

WDXRF analyses were carried out both on polished stone tool and Bulhary sample. As 2.4 g analytical purity glass sand was mixed to 0.6 g sample in both cases, the 1:4 ratio is noticed at the evaluation of results. Because of the high content of Si and Al elements of glass sand, those data were ignored from the interpretation. In a controlled analysis the alkali basalt represented results for Si and Al. Regarding the TiO_2, MgO, CaO, Na_2O, K_2O and P_2O_5 content, the stone axe and the alkali basalt show a very good match (**Table 1**., **Fig. 9.**).

Discussion

The EDS/SEM determined mineral association of the stone implement accords with the alkali basalt from Ceres Mountains (Farsang et al. 2014).

EDS/SEM of alkali basalt from Bulhary was accomplished also, because there is no other known occurrence of sodalite-bearing basalt in the Carpathian Basin or its surroundings (Farsang et al. 2014, Fehér et al. 2016). The subhedral olivines can reach 700 µm in both samples, their chemical composition vary very similar from core to rim.

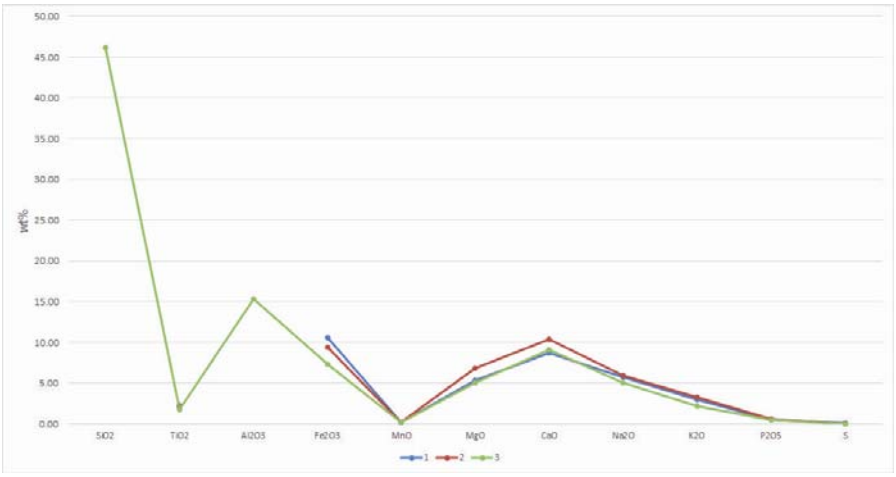

Fig. 9. Bulk chemistry data of D19 stone axe and the alkali basalt from Bulhary.

1. D19 stone axe (mixed with glass sand); 2. Bulhary quarry: alkali basalt (mixed with glass sand); 3. Bulhary quarry: alkali basalt

9. ábra: A D19 jelű kőbalta és a bolgáromi alkáli bazalt kőzetkémiai eredményei.

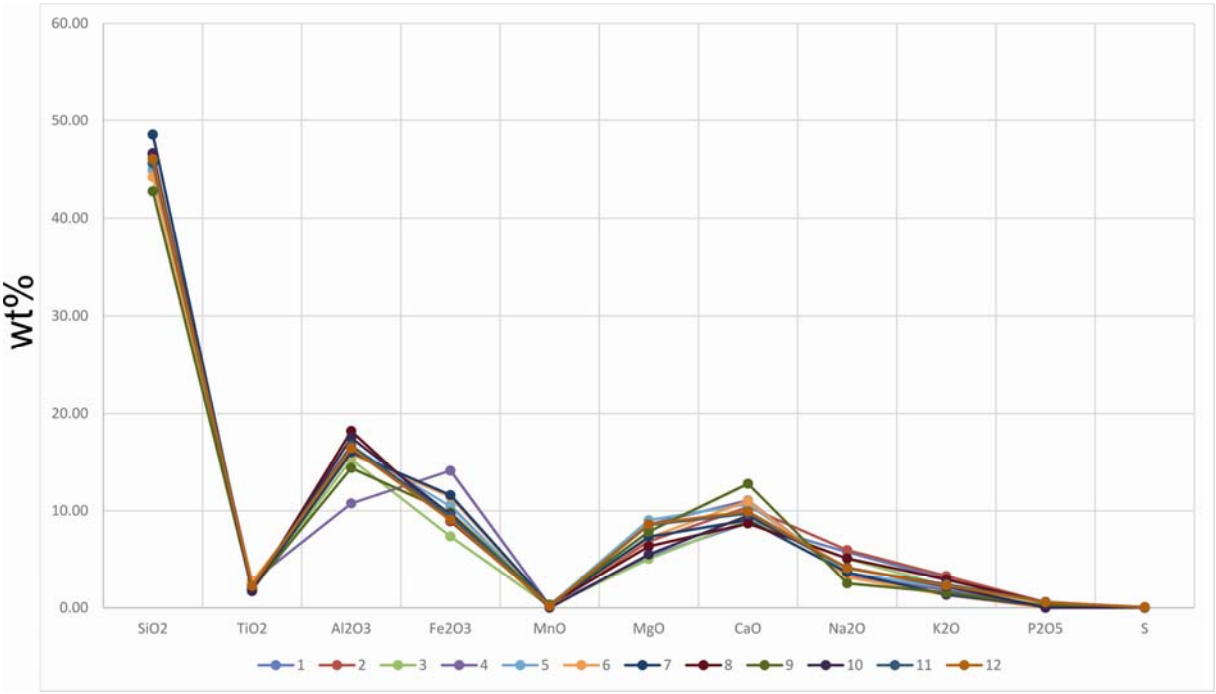

Fig. 10: Bulk chemistry results compared with previously published data

1. D19 stone axe mixed with glass sand; 2. Bulhary quarry-alkali basalt mixed with glass sand; 3. Bulhary quarry-alkali basalt; 4. Bulhary limburgitoide basanite (Forgáč 1970); 5. Konradovce nepheline basanite (Forgáč 1970) 6. Filakovo nepheline basanite (Forgáč 1970); 7. Belina nepheline basanite (Forgáč 1970) 8. Badzovce nepheline basanite (Forgáč 1970); 9. Borkul-Bagac nepheline basanite (Forgáč 1970). 10. Average bulk chemistry of basalt fields from North Hungary (Jugovics 1974); 11. Bulhary quarry basanite (Hakulinová et al. 2011.) 12. Bulhary abandoned quarry basanite (Hakulinová et al. 2011.)

10. ábra: Kőzetkémiai eredményeink összehasonlítva a korábban publikált adatokkal

Fayalite-content overlaps: in the stone implement is 25-34% and in the alkali basalt is 24-33%. In both samples a few olivine crystals contain smectitic alteration zones inside (Figs. 3 and 4). Zoned clinopyroxenes are present both in the stone axe and the alkali basalt. In the stone axe diopside and augite are determined, from the alkali basalt only diopside is described (**Fig. 5.**). Chemical composition of plagioclases corresponds to labradorite, anorthite content of the stone implement and the alkali basalt overlaps (Fig. 6). The size of labradorite can reach 500 µm in the stone axe, but it is smaller (up to 100 µm) in the alkali basalt (**Figs. 3.** and **4.**). Farsang et al. (2014) described plagioclase megacrystals from more locations in the basalt of Ceres Mountains.

Equant spinel-group crystals distribute in each sample evenly (**Figs. 3.** and **4.**). In the case of stone axe ulvospinel up to 40 µm represents the spinel group, however Ti-rich magnetite is observed in the alkali basalt where the largest crystal reached 200 µm. Spinel xenoliths were recorded in olivines and pyroxenes from both samples (Figs. 3, 4)

Feldspathoids are also noticed in both samples, though the mineral species are slightly different. Sodalites fill the spaces between other minerals in both samples. The size of the sodalite crystals are larger in the Bulhary alkali basalt than in the stone axe: 70 µm and 10 µm, respectively (**Figs. 3** and **4.**).

Natrolite is observed in the stone axe. In the Bulhary basalt a zeolite-like mineral is also detected, however, it cannot be specified on the basis of the chemical composition. Nepheline and leucite are present in the Bulhary basalt, but in the stone axe these two minerals are not proved by EDS/SEM, although nepheline is confirmed by XRD.

Alkali feldspars, namely anorthoclase and sanidine, occur in the alkali basalt, however these minerals were not recognized in the stone axe.

The XRD pattern of Bulhary sample shows several similarities with the stone axe, except some accessories (**Figs. 7- 8.**). Glass phase is not detected one of the samples.

Earlier published data from Ceres Mountains and our bulk chemical data are shown in Table 1 and plotted on **Fig. 10.** as confirmation of the Bulhary sodalite-bearing basalt as the possible provenance of D19 Neolithic polished stone tool. Regarding the major elements, the best matching with our data have the Badzovze nepheline basanite (Ceres

Mountains) (Forgáč 1970) and Bulhary-basanite quarry and abandoned pit (Hakulinová et al. 2011).

The stone implement and the Bulhary alkali basalt show several good matches in mineralogical association according to EDS/SEM and XRD. Sodalite was identifed as rock-forming mineral in both samples. According to Farsang et al. (2014), sodalite is originated from high temperature fluids and described as cavity filling mineral. In the stone implement sodalite is defined as rock-forming mineral among the previously crystallized minerals. It could form from residual melt enriched in incompatible elements (Cl) instead of feldspars or by hydrothermal alteration process from feldspars or glass.

Conclusions

Considering the mineralogical assemblage, the textures, and the bulk chemistry of the stone axe, the alkali basalt and noting the previously published data (Forgáč 1970, Hakulinová et al. 2011), the D19 stone axe is very similar to the basanites of Ceres Mountains. Primary sodalite-bearing basalt is only described from Bulhary quarry and its surroundings in the Carpathian Basin (Farsang et al.2014, Fehér et al. 2016). According to this fact, the provenance field of D19 stone axe could be Bulhary or its surroundings (Slovakia) (**Fig. 11.**).

Alkali basalt as stone raw material is known in the Neolithic (Szakmány 2009) although detailed studies have not been published yet. Concluding sodalite-bearing alkali basalt as lithic raw material has not been described from the earlier studied polished stone implements yet.

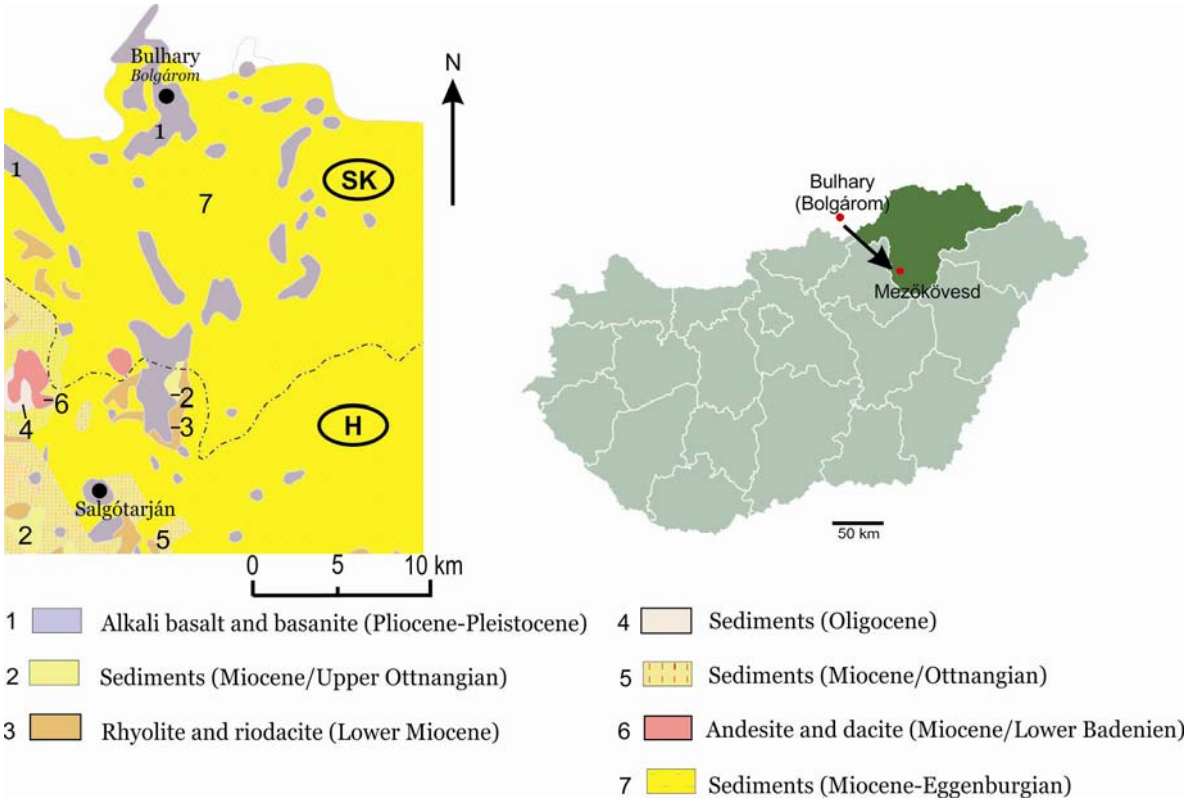

Fig. 11: Archaeological locality and presumed raw material source of the D19 stone axe

11. ábra: A D19 jelű kőbalta régészeti lelőhelye és feltételezett forrásterülete

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