PGAA AS AN ANALYZING TOOL FOR OBSIDIAN ARCHAEOLOGICAL SAMPLES

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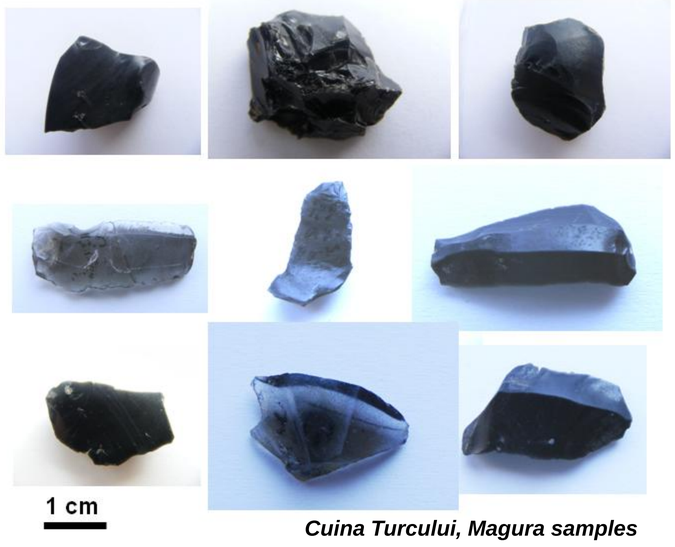
INTRODUCTION: Non-destructive Prompt Gamma Activation Analysis was applied to perform provenance study of Neolithic obsidian artefacts. Elemental compositions of archaeological objects from Romanian sites have been compared with reference measurements of the most important geological sources in Central Europe and in the Mediterranean region. Based on the measured concentrations, especially on B- and Cl content, the samples proved to be either 'Carpathian I' (North of Tokaj mountains, Slovakia) or 'Carpathian II' (South of Tokaj mountains, Hungary) types. However, further methods are recommended to identify more fingerprint-like trace elements in obsidians.

Photo of the investigated Romanian obsidians

Iclod, Tzaga, Silagiu samples

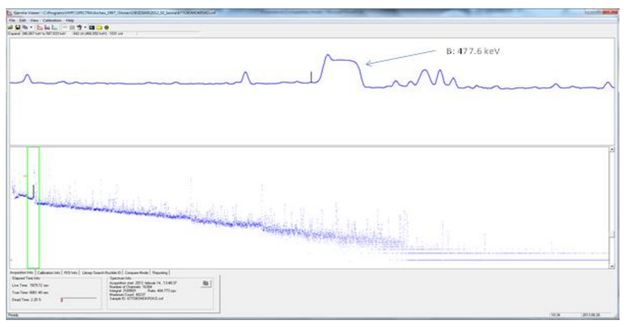


Cuina Turcului, Magura samples

Illustrative map of localities of the investigated samples

METHOD: Prompt Gamma Activation Analysis (PGAA) was applied at the Budapest Neutron Centre to determine the bulk elemental composition [5]. PGAA is based on the detection of characteristic gamma photons emitted in (n,γ) reactions. Contrary to the conventional neutron activation analysis (NAA), the irradiation and the detection happen simultaneously on a horizontal guided neutron beam. The quantitative analysis is based on the k_0 principle [6]. The greatest advantage of PGAA is that it does not require sampling or any preparation of archaeological objects. As a consequence, neither any destruction nor significant induced radioactivity is produced. Based on our previous research, PGAA can be successfully applied in provenance research of obsidians [7].

An obsidian in the sample holder frame



A typical PGAA spectrum with the characteristic Boron peak

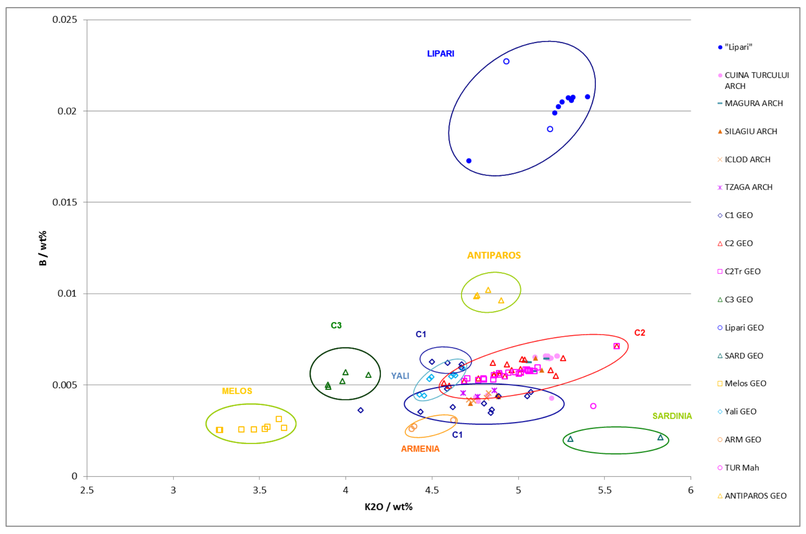
AIMS: Forty-five archaeological samples from different regions of Romania and from different prehistorical periods were analyzed at the PGAA facility of the Budapest Neutron Centre in 2012 and 2017: Iclod, Tzaga, Turda, Seleus, Bocin, Tasnad, Silagiu sites in Cluj and Oradea regions of Transylvania, Neolithic period [1]; Cuina Turcului site at Iron Gates [2] (on Danube border, between Romania and Serbia), Late Mesolithic and Early Neolithic (Neolithization period [3]); Magura site in Teleorman region (South of Muntenia, near Danube), Early Neolithic (Neolithization period).
The aim of the study was to identify obsidian geological sources used in each region and period [4]. Neolithization is the process of transition from hunting-fishing-based society to agriculture, process related to an important populations movement. The most accepted theory is "Ex Oriente Lux", the migration of "Neolithic model" (and population) from Mesopotamia, Anatolia, Greece - through Aegean Islands, Balkans, Central Europe - via Danube.
Two main geological regions are presumed to be the obsidian sources for Romanian territory: Tokaj Mountains (Carpathian I – now in Southern Slovakia and Carpathian II – now in Northern Hungary) and Greek Islands – especially Melos (Aegean Sea).

Obsidian archaeological samples

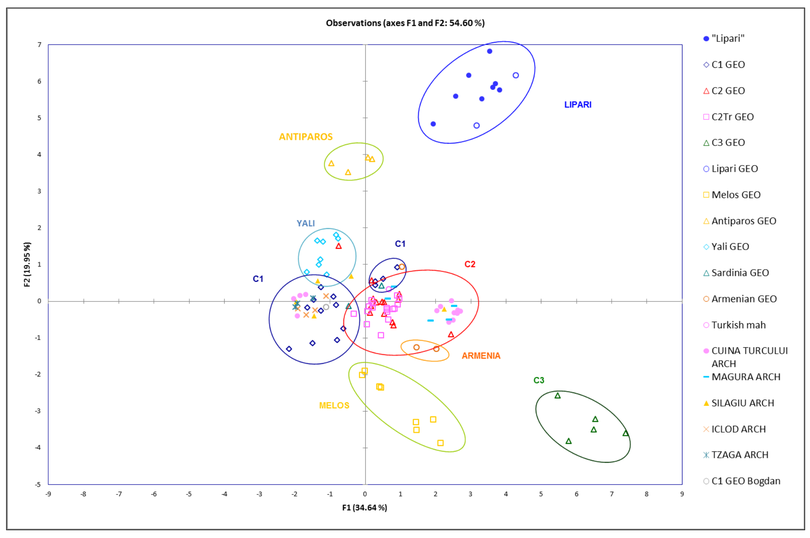
Region	Sites	Period
Cluj (Centre of Transylvania)	Iclod, Tzaga, Turda, Silagiu	Neolithic
Oradea (North-West of Transylvania)	Seleus, Bocin, Tasnad	Neolithic
Iron Gates (on Danube border, between Romania and Serbia)	Cuina Turcului	Late Mesolithic and Early Neolithic (Neolithization period)
Teleorman region (South of Muntenia, near Danube)	Magura	Early Neolithic (Neolithization period)

Obsidian geological souces

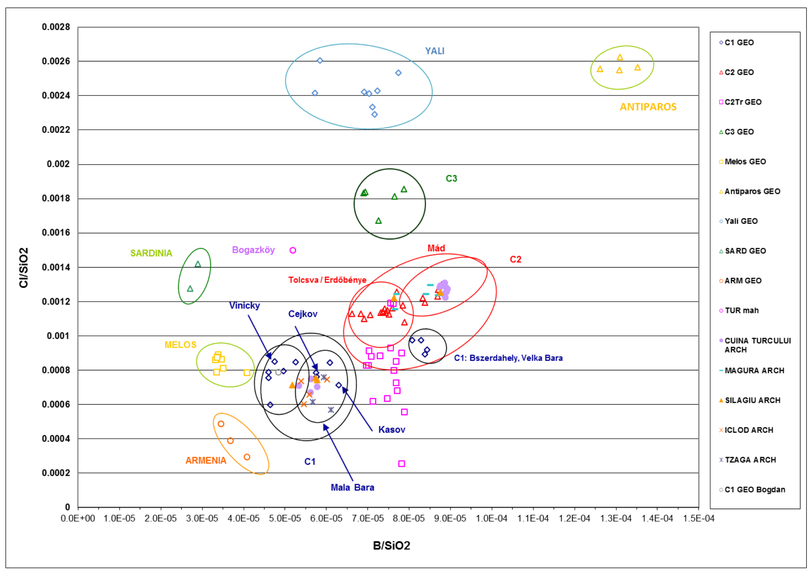
Carpathian I - Hungarian Tokaj Mountains
Carpathian II - Slovak Tokaj Mountains
Melos - Aegean Sea Island
Yali- Aegean Sea Island
Lipari – near Sicily Island
Sardinia - Tyrrhenian Sea Island
Armenia
Eastern Anatolia
Antiparos – Aegean Sea Island
Pantelleria – near Sicily
Palmarola – Tyrrhenian Sea Island



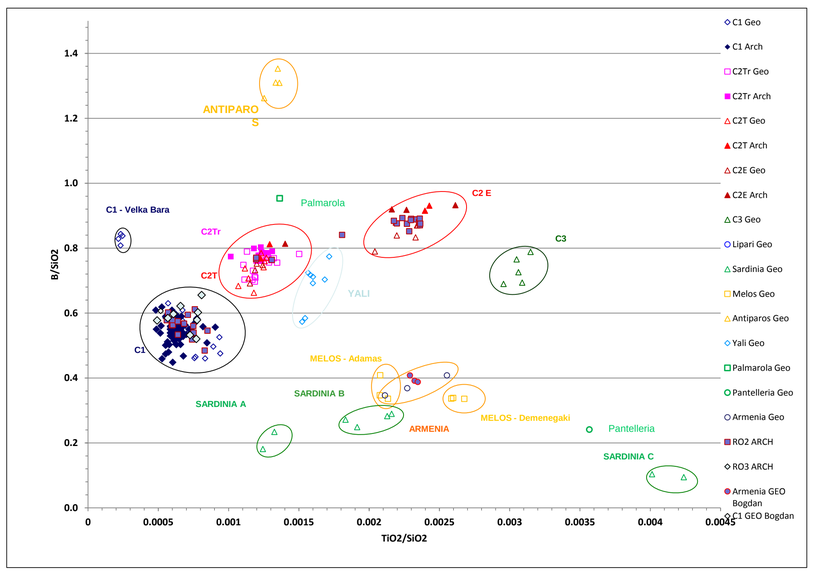
Classification of obsidians according to their K and B content



Classification of obsidians based on the detected components / PCA



Classification of obsidians according to their B and Cl content



Classification of obsidians according to their B and Ti O_2 content
RO2 – 2012 measurements
RO3 – 2017 measurements

RESULTS: PGAA proved to be the most convenient method to quantify the major components and some characteristic trace elements in the bulk material, most of all B and Cl, in a non-invasive way. In order to determine the provenance of the archaeological objects, we have investigated several elements' contents. Compositions of archaeological objects were compared with our own reference database including the major European and Mediterranean samples from the Lithoteka of the Hungarian National Museum. B/SiO_2 vs. Cl/SiO_2 ratios, Principal Components Analysis (PCA) and B/SiO_2 vs. $TiO2/SiO_2$ proved to be the most indicative in determination of different groups.
Our results indicate that all the Transylvanian Neolithic samples fit the Carpathian I pattern. The same pattern can be attributed to Mesolithic Cuina Turcului samples. A special situation is for the Neolithization period, both for Cuina Turcului and Teleorman. These samples fit Carpathian II pattern, however, based on K_2O content, these samples might have been similar to those from Yali Island (Aegean Sea). Since the latters are known to show weak mechanical quality, it has been less probably used for tools production. By increasing the number of fingerprinting elements, using additional analytical methods, one can further confirm or disprove the current theories of Neolithization.

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