Preliminary archaeometric investigation on Middle Neolithic siliceous tools from Limba-Oarda de Jos (Transylvania, Romania)

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Abstract:

The present archaeometric study focuses on a set of archaeological siliceous lithic tools that are assigned to the early Vinča culture period (Vinča A and Vinča B1). They were found in several pit-houses at Limba-Oarda de Jos (SW Transylvania, Romania), an open settlement that has been dated to 5,405-5,310 cal. BCE, a period in the Middle Neolithic. A total of 322 retouched tools and débitage pieces were typologically and macroscopically investigated. From these, 20 pieces were analysed by polarized light optical microscopy (OM) and 10 pieces were analysed by Fourier-transform infrared spectroscopy (FTIR) in order to identify compositional characteristics, define the petrographic type, and establish the spectral fingerprint of each material.

Four petrographic types were discriminated: radiolarite, chert, fossiliferous chert, and siliceous limestone. Mineralogically, the tools primarily consist of a mass of microquartz and fibrous microquartz (called also ‘chalcedony’) associated with radiolarians remnants (in radiolarites); fossil shell fragments (in the fossiliferous chert); and limestone components, such as ooliths and pellets (in the siliceous limestone). All samples show distinct FTIR bands, most of which are assigned to microquartz, quartz, and fibrous microquartz. The deconvolution of the FTIR spectra in the 950-1300 cm\(^{-1}\) domain reveals the contribution of several other phases, such as calcite and clay minerals.

The results support the assumption that the tools made of chert, fossiliferous chert, and siliceous limestone were produced at the site from nodules that probably originated from the Upper Jurassic chert-bearing limestone that crops out nearby in the Trascău Mts. The tools made of radiolarite were most likely brought to the site as finished products from the Trascău Mts.
Keywords: siliceous tools; Early Neolithic; optical microscopy; FTIR; Vinča culture; Limba-Oarda de Jos; Romania

1. Introduction

In the southwestern part of the Transylvanian Basin, Romania (see Figure 1), remnants of a Neolithic complex settlement were unearthed at Limba-Oarda de Jos (see Figure 2). This settlement was inhabited from the early Neolithic (Pre-Criș and Starčevo-Criș cultures) to the Middle Neolithic (Vinča culture) (Ciută & Ciută 2015, Whittle et al. 2016). Archaeological research has resulted in a large collection of various artefacts that are presently stored at the “Ion Raica” Municipal Museum in Sebeș (Alba county). This pseudo-tell site was emplaced on the Mureș River left bank near the Trascău Mts. and is ca. 4 km (in a straight line) SSE of the present-day city of Alba Iulia (see Figure 1). The location was likely strategically chosen to allow control of the routes connecting the eastern and southern areas of the Transylvanian Basin with Banat, the Pannonian Plain, and the areas beyond the Southern Carpathians (Suciu 2009).

The Vinča culture was widespread in the southeastern part of Europe and occupied a territory that predominantly corresponds to modern-day Serbia, Kosovo, as well as parts of Romania, Croatia, Bulgaria, Bosnia-Herzegovina, Montenegro, the Republic of Northern Macedonia (FTYR), and Greece (Bogosavljević 2017; Suciu 2009). The onset of this culture can be linked with the control of both the salt sources (Nandris 1990) and obsidian trade routes (Suciu 2010). The Vinča culture is characterized by particularly large tell settlements and specific ritual behaviour (Suciu 2009).

For more than twenty years, research and systematic investigations have been carried out in this archaeological site, and these efforts have resulted in the compilation of a collection of various lithic materials, including polished stone tools (over 150 axes and chisels) and more than one thousand retouched and non-retouched blade stones. The seven pieces of alabaster idols and amulets and the two zoomorphic pieces made of marble (Ciută & Ciută 2011) previously found here are regarded as hallmarks with ritual and spiritual significance (Crnobrnja 2011).

Several petroarchaeological studies of Vinča chipped lithic tools and their raw material provenience have been carried out by Bogosavljević (2001; 2016; 2017), Antonović et al. (2005), Biagi et al. (2007), Crandell (2008), and Rey-Solé et al. (2018). They documented the use of a large range of rock types, most of which were identified as quartzite and chert, i.e., “Banat flint” (Biagi et al. 2007; Comșa 1971; Crandell 2008) and “Moldavian flint” (Crandell 2012a; 2012b). Obsidian, radiolarite (Biagi et al. 2007), greenschist, dolerite, and tuff (Antonović et al. 2005) were used as well.

The aim of this investigation is to discriminate and classify 322 siliceous tools and débitage (see Figure 3) that were found at Limba-Oarda de Jos and assigned to the early Vinča culture period (Ciută et al. 2016). A combination of macroscopic and microscopic methods with infrared spectroscopy was applied in order to obtain detailed information on the composition and structure of the materials and to ultimately infer the provenance of the material.

Previous geological and archaeological research has indicated that siliceous rocks such as cherts, radiolarites, cherty limestones, and other knappable materials used in Prehistory originate from the Southern Apuseni Mts., particularly in the Metaliferi and the Trascău mountains (Crandell 2008; 2009; 2014; Herz & Savu 1974; Lupu, 1995; Nicolae 1992).
2. Materials and methods

A set of 322 retouched tools and débitage pieces were washed, photographed, and investigated macroscopically using an SMZ645 Nikon stereomicroscope. Twenty pieces were selected as representative of the main macroscopically defined groups (mainly débitage). From these twenty pieces, thin sliced were cut with a diamond saw and used to prepare thin sections for polarized light optical microscopy (OM). A Zeiss Axio Imager.A2m petrographic microscope (Geology Department at Babeș-Bolyai University in Cluj-Napoca) was used for microscopic and mineralogical studies. The microphotos were captured by an Axio Cam ICc5 video camera.
Fig. 2. Aerial view of the 2016 archaeological excavations at Limba-Oarda de Jos (Photo: M.M. Ciută; image from Ciută et al. 2016). The inset in the lower right shows the position of the area within Romania (modified from D-maps.com (2019)).

Figure 3. Detailed macroscopic images (dorsal and ventral) of some of the studied lithics from Limba-Oarda de Jos. A: flake, sample 4472. B: fragment (débitage), sample 4576(15). C: flake, sample 4446. D: blade, sample 4469 (from Rey-Solé et al. 2018).
Information on the composition and microstructure of the material was obtained by analysing ten of the twenty pieces by Fourier-transform infrared spectroscopy (FTIR) using a JASCO 6200 spectrometer (Nanostructured Materials and Bio-Nano-Interface Center in the Interdisciplinary Research Institute on Bio-Nano-Sciences at Babeş-Bolyai University in Cluj-Napoca). From ~1.2 mg of hand-milled sample mixed with 150 mg KBr, pellets with a 13 mm diameter were prepared under 5 tonnes of pressure. The spectrometer was operated with 256 scans in the spectral range of 4000-400 cm$^{-1}$ and a resolution of 4 cm$^{-1}$. The spectra were analysed using Spectra Analysis software. For detailed compositional information, deconvoluted spectra in the 1300-950 cm$^{-1}$ domain were obtained.

3. Results and discussion

3.1. Macroscopic observation

The macroscopic parameters analysed were based on those described by Luedtke (1992), Crandell (2005; 2006), Tarriño (2006), and Rey-Solé (2016). They include the shape (typology) and size, as well as the fabric, lustre, translucency, grain size, colour (from Munsell 1975), and pattern. The great variability in the appearance of the samples provided the criteria for distinguishing and grouping them. Four different groups were discriminated according to their abundance in the site, as described in the following paragraphs (see Table 1 and Figure 3).

<table>
<thead>
<tr>
<th>Sample no.</th>
<th>Typology</th>
<th>Size (mm)</th>
<th>Munsell colour</th>
<th>Petrographic type</th>
</tr>
</thead>
<tbody>
<tr>
<td>4379</td>
<td>Flake</td>
<td>39x2x18</td>
<td>Dark grey (2.5YR N4)</td>
<td>Radiolarite</td>
</tr>
<tr>
<td>4472</td>
<td>Fragmented blade</td>
<td>25x13x4</td>
<td>Olive yellow (2.5Y 6/8)</td>
<td>Radiolarite</td>
</tr>
<tr>
<td>4350(2)</td>
<td>Fragmented flake</td>
<td>2x2x4</td>
<td>Dark brown (7.5 YR 3/2)</td>
<td>Chert</td>
</tr>
<tr>
<td>4419</td>
<td>Blade</td>
<td>4x16x6</td>
<td>Dark yellowish brown (10YR 4/4)</td>
<td>Chert</td>
</tr>
<tr>
<td>4576(15)</td>
<td>Fragment (débitage)</td>
<td>26x22x8</td>
<td>Very dark greyish brown (2.5Y 3/2)</td>
<td>Chert</td>
</tr>
<tr>
<td>4576(19)</td>
<td>Fragment (débitage)</td>
<td>35x28x14</td>
<td>Very dark greyish brown (2.5Y 3/2)</td>
<td>Chert</td>
</tr>
<tr>
<td>4622(2)</td>
<td>Blade</td>
<td>42x14x5</td>
<td>Brownish yellow (10YR 6/6)</td>
<td>Chert</td>
</tr>
<tr>
<td>4497(7)</td>
<td>Blade</td>
<td>36x14x4</td>
<td>Dark yellowish brown (10YR 3/6)</td>
<td>Fossiliferous chert</td>
</tr>
<tr>
<td>4542(2)</td>
<td>Fragment (débitage)</td>
<td>21x22x4</td>
<td>Dark yellowish brown (10YR 3/6)</td>
<td>Fossiliferous chert</td>
</tr>
<tr>
<td>4576(12)</td>
<td>Fragment (débitage)</td>
<td>38x32x16</td>
<td>Very dark greyish brown (2.5Y 3/2)</td>
<td>Fossiliferous chert</td>
</tr>
<tr>
<td>4576(13)</td>
<td>Fragment (débitage)</td>
<td>27x21x3</td>
<td>Brown/dark brown (10YR 4/3)</td>
<td>Fossiliferous chert</td>
</tr>
<tr>
<td>4446</td>
<td>Fragmented blade</td>
<td>27x19x3</td>
<td>Dark yellowish brown (10YR 3/6)</td>
<td>Fossiliferous chert</td>
</tr>
<tr>
<td>4576(16)</td>
<td>Flake</td>
<td>34x36x3</td>
<td>Black (5Y 2.5/2)</td>
<td>Fossiliferous chert</td>
</tr>
<tr>
<td>4574</td>
<td>Fragment of blade</td>
<td>31x18x3</td>
<td>Dark yellowish brown (10YR 3/6)</td>
<td>Siliceous limestone</td>
</tr>
<tr>
<td>4440</td>
<td>Truncated blade</td>
<td>31x15x3</td>
<td>Light olive brown (2.5Y 5/4)</td>
<td>Siliceous limestone</td>
</tr>
<tr>
<td>4576(18)</td>
<td>Fragment (débitage)</td>
<td>37x2x5</td>
<td>Dark yellowish brown (10YR 3/6)</td>
<td>Siliceous limestone</td>
</tr>
<tr>
<td>4469</td>
<td>Truncated blade</td>
<td>3x12x7</td>
<td>Very dark brown (10YR 2/2)</td>
<td>Siliceous limestone</td>
</tr>
<tr>
<td>4599</td>
<td>Fragment of blade</td>
<td>27x15x7</td>
<td>Light olive brown (2.5 Y 5/4)</td>
<td>Siliceous limestone</td>
</tr>
</tbody>
</table>

The first group consists of 41 samples (12.73% of the total) that are mostly homogeneous in appearance and have a waxy shine. The pieces are opaque to sub-translucent, very fine grained, and olive yellow in colour (2.5Y 6/8)(Munsell 1975). Ten pieces from this group display a mottled and stripped aspect. Macroscopically, the pieces are defined as radiolarites. Typologically, the group is composed of 24 blades and fragmented blades (8 of them are assigned to the stripped subgroups, and 4 pieces are retouched on one side), 4 flakes and fragmented flakes (débitage), 10 fragments (débitage), and 3 cores. The blades range from 30 to 45 mm in length and from 5 to 20 mm in width, and the flakes range from 18 to 15 mm in length and from 12 to 15 mm in width (see Table 1).
The second group is composed of 86 samples (26.71% of the total) with a heterogeneous, dull, opaque, and fine- to medium-grained appearance and a dark greyish brown colour (2.5Y 3/2)(Munsell 1975). Typologically, this group consists of 24 blades and fragmented blades, 18 flakes and fragmented flakes (débitage), 43 fragments (débitage) and 1 core. The length of the blades (most of which are truncated blades) ranges from 15 to 45 mm, and the width ranges from 8 to 25 mm. The flakes range in length from 15 to 45 mm and in width from 15 to 44 mm. Some pieces show remnants of cortex. Macroscopically, the pieces are defined as being made of a type of rough chert.

The third group consists of 90 samples (27.95% of the total) with a homogeneous, shiny, and translucent to sub-translucent appearance. They are very fine to fine grained, and the colour (Munsell 1975) ranges from dark yellowish brown (10YR 3/6) to black (10YR 2/1). Typologically, this group is composed of 40 blades and fragmented blades, 7 of which have only one side retouched. Thirty flakes and fragmented flakes, 19 fragments (débitage), and 1 core also belong to this group. The blades range in length from 10 to 38 mm and in width from 10 to 18 mm. The sizes of the flakes are more variable, ranging from 18 to 45 mm in length and from 10 to 30 mm in width. Fifteen samples in this group have cortex remains. Macroscopically, the pieces are defined as being made of a type of fine chert.

The fourth group consists of 105 samples (32.60% of the total) with a heterogeneous appearance and a greasy shine. They are opaque and medium to coarse grained and the colour (Munsell 1975) ranges from very dark brown (10YR 2/2) to light brownish grey (10YR 6/2). Macroscopically, the pieces are defined as being made of siliceous limestone. Typologically, this group is composed of 45 blades (of which 24 are truncated) and fragments of blades, 33 flakes and fragmented flakes, 25 fragments (débitage) and 2 cores. The blades range from 23 to 53 mm in length and from 9 to 17 mm in width, whereas the flakes range from 11 to 52 mm in length and from 9 to 51 mm in width. Seven pieces (6 flakes and 1 core) from this group show cortex remains.

3.2. Polarized light optical microscopy

On the basis of the mineralogical features (composition and fabric, i.e., structure and texture) observed in the thin sections, four main petrographic types are discriminated: radiolarite, chert, fossiliferous chert, and siliceous limestone (Figure 4 A1-D2). The analysed microscopic parameters, including the description and classification, are based on those described by Dunham (1962), Knauth (1994), Tarriño (1998), Graetsch (1994), Knauth (1994), Bustillo et al. (2009), Přichystal (2010) and Ionescu & Hoeck (2018). Despite having variable macroscopic features, the chert samples are highly homogeneous microscopically.

Radiolarite (samples 4379 and 4472)

The samples studied consist of a microquartzitic mass (less than 100 micrometres in size) and an abundance of whole spherical or fragments of radiolarian tests. The latter range from 50 to 200 micrometers in size (Figure 4 A1 and A2) and most of them are replaced by fibrous microquartz ("chalcedony").

Chert (samples 4350(2), 4576(15), 4576(19), 4622(2), and 4419))

The analysed samples reveal a granular microquartzitic matrix (grains less than 20 micrometers in size) with rare remnants of foraminifera tests and other undetermined bioclasts. Some fossil tests are replaced by fibrous microquartz that grades inward into megaquartz (see Figure 4 B1). The rock also contains clusters of megaquartz crystals (see Figure 4 B2).
Figure 4. Polarized light optical microphotos: A - Radiolarite: general view of sample (A1) and radiolarian test in detail (A2); B - Chert: microfossil replaced by chalcedony that grades inward into megaquartz (B1) and a cavity filled by a mosaic of megaquartz (B2); C - Fossiliferous chert: a shell fragment in a fine-grained mass of microquartz (C1) and monaxon and triaxon sponge spicules in chert (C2); D - Siliceous limestone (Grainstone) with ooliths in siliceous cement made of microquartz (D1) and fibrous microquartz filling spaces among clasts (D2). All images with crossed polarizers, except A1 and C2 (one polarizer). Images A1, B1, C1, and D1 are from Rey-Solé et al. (2018).
Fossiliferous chert (4446, 4497(7), 4542(2), 4576(12), 4576(13), and 4576(16))

The parent rock of these samples was a limestone (wackestone) containing a great diversity of marine microfossils. The carbonate rock is replaced by a microquartzitic matrix. The fossil tests are made of carbonate or are replaced by a mass of fibrous quartz. There are remnants of a wide range of fossil types: bivalves (see Figure 4 C1), distinct sections of calcareous triaxon and monaxon spicules (see Figure 4 C2), vegetal remains, foraminifera, and fragments of undetermined fossils. In some parts, the rock is heterogeneous owing to a concentration of reddish brown iron oxides and organic matter.

Siliceous limestone (samples 4440(1), 4469, 4574, 4576(18), and 4599)

These samples show an oolithic and pelletal texture, which was most likely inherited from a parental grainstone. These particles range from 100 micrometers to almost 1 mm in diameter (see Figure 4 D1). The limestone is intensely silicified and a fine microquartzitic material cements the ooliths and the pellets. There are also abundant fragments of marine organisms, such as foraminifera, thecal plates (grill texture) of echinoids, fragments of bivalves and other fragments of unknown origin. Most of the clasts consist of carbonate. Nevertheless, several bioclasts are replaced by either fibrous microquartz (‘chalcedony’) or by megaquartz (see Figure 4 D2). In addition, opaque minerals are detected.

3.3. Fourier-transform infrared spectroscopy

Fourier-transform infrared spectroscopy (FTIR) is employed for a wide range of materials used in lithic technology. It helps in the identification of microcrystalline and amorphous silica (Banerjee 1993; Handke and Mozgawa 1993), as well as various opals (Adamo et al. 2010). Therefore, this analytical technique can be regarded as essential to the study of siliceous artefact composition and the acquisition of information about the crystallinity of silica minerals (Hlavay et al. 1978).

The spectra obtained for the ten samples (one radiolarite, three cherts, three fossiliferous cherts and three siliceous limestones) are shown in Figure 5. All samples produced Si-O stretching vibrations at 462, a 781-797 doublet, and at ~1089 cm⁻¹, which is characteristic of quartz (Kieffer 1979; Moenke 1974), and an almost indistinct and broad band centred at ~3500 cm⁻¹ that is due to the water content (up to 2 mass %) found in chert (Knauth 1994). This signal can also be linked to H-O-H vibrations of adsorbed water (Madejová 2003) or to OH groups in some clay minerals, which may be present as impurities. The samples made of fossiliferous chert and siliceous limestones have wide bands at 1425 and 878 cm⁻¹ that are generated by CO₃ vibrations (Fabbri et al. 2014; Shoval 2003) and normally assigned to calcite content (Kieffer 1979). The 695 and 464 cm⁻¹ signals are due to Si-O bending vibrations in crystalline materials, i.e., quartz.

To identify the compounds that contribute to the main FTIR signal centred at ~1089 cm⁻¹ (see Figure 5), the spectra were deconvoluted in the 950-1300 cm⁻¹ domain (see Figures 6 A-D and 7 A-F; Table 2). The fitted spectrum (not shown here) overlaps the experimental one. There are three main components in each spectrum, except for one siliceous limestone sample (4576(18)). Radiolarite, chert, fossiliferous chert, and siliceous limestone show similar compositions, which are basically quartzitic (microquartz), with their main signals appearing at 1088-1090 cm⁻¹ (Handke & Mozgawa 1993). Each sample also shows similar amounts of a secondary compound, as evidenced by the 1168-1175 cm⁻¹ signal. This can be assigned to quartz (De Benedetto et al. 2002; Kieffer 1979; Handke & Mozgawa 1993) and fibrous microquartz (Banerjee 1993). The third signal at 1018-1019 cm⁻¹ is weak. It is shifted to a
higher wavenumber (1035 cm\(^{-1}\)) in one chert sample (see Figure 6 D). This FTIR band can be produced by Si-O-Al vibrations in clay minerals (De Benedetto et al. 2002).

The ratio between the intensity (expressed here as the measured area of the signal) of the component signals in the deconvoluted spectra (see Table 2) show that the main signal (centred at \(\sim 1088-1090\) cm\(^{-1}\)) is generally 2-3 times larger than the second one (centred at \(\sim 1168-1176\) cm\(^{-1}\)). The contribution of the third signal is insignificant.

The narrow bands assigned to quartz prove the prevalence of the (micro)crystalline form of quartz in all samples studied. It is both microquartz and fibrous microquartz (\textit{sensu} Graetch 1994). There is no association of specific bands at 1638 + 1102 + 789 (single band) + 475 cm\(^{-1}\) (Caucia et al. 2012); hence, no opal CT occurs in our samples.

![Image of FTIR spectra](https://doi.org/10.2218/jls.3020).

Figure 5. The pattern of the FTIR spectra for radiolarite, chert, fossiliferous chert, and siliceous limestone pieces in the 4000-500 cm\(^{-1}\) region. (Image from Rey-Solé et al. 2018).
Figure 6. Experimental and deconvoluted spectra in the 1300-950 cm\(^{-1}\) range for (A) Radiolarite; sample 4379; (B, C, and D) Chert; samples 4419, 4622(2), and 4350(2). The experimental spectrum is drawn with a dashed line.
Figure 7. Experimental, fitted, and deconvoluted spectra in the 1300-950 cm$^{-1}$ range for (A, B, and C) Fossiliferous chert: samples 4599, 4576(18), and 4469; (D, E and F) Siliceous limestone: samples 4542(2), 4446, and 4576(13). The experimental spectrum is drawn with a dashed line.
Table 2. The wavenumbers for the component signals of deconvoluted spectra and the ratio between the intensity (I) of each signal.

<table>
<thead>
<tr>
<th>Petrographic type</th>
<th>Sample no.</th>
<th>Component (cm(^{-1}))</th>
<th>(I_1 : I_2 : I_3) areas ratio (a.u.)</th>
<th>(I_2/I_1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radolarite</td>
<td>4379</td>
<td>1168 1089 1019</td>
<td>29 : 103 : 11</td>
<td>3.55</td>
</tr>
<tr>
<td>Chert</td>
<td>4419</td>
<td>1171 1088 1023</td>
<td>32 : 95 : 6</td>
<td>2.97</td>
</tr>
<tr>
<td></td>
<td>4622</td>
<td>1174 1088 1018</td>
<td>53 : 100 : 2.2</td>
<td>1.89</td>
</tr>
<tr>
<td></td>
<td>4350</td>
<td>1169 1089 1032</td>
<td>50 : 73 : 17</td>
<td>1.46</td>
</tr>
<tr>
<td>Fossiliferous chert</td>
<td>4599</td>
<td>1169 1089 1025</td>
<td>43 : 60 : 7.7</td>
<td>1.40</td>
</tr>
<tr>
<td></td>
<td>4576(18)</td>
<td>1173 1090 -</td>
<td>32 : 100</td>
<td>3.13</td>
</tr>
<tr>
<td></td>
<td>4469</td>
<td>1171 1088 1019</td>
<td>39 : 97 : 3</td>
<td>2.49</td>
</tr>
<tr>
<td>Siliceous limestone</td>
<td>4542</td>
<td>1174 1088 1019</td>
<td>35 : 92 : 5</td>
<td>2.63</td>
</tr>
<tr>
<td></td>
<td>4446</td>
<td>1173 1088 1019</td>
<td>42 : 101 : 2.5</td>
<td>2.40</td>
</tr>
<tr>
<td></td>
<td>4576(13)</td>
<td>1176 1089 1018</td>
<td>48 : 108 : 2.7</td>
<td>2.25</td>
</tr>
</tbody>
</table>

4. Discussion

Microscopic features facilitate the clear separation of the samples into petrographic groups. Radolarite, chert, fossiliferous chert and siliceous limestone show a primarily quartzitic composition that is represented by microquartz, quartz and fibrous microquartz. The FTIR spectra of the analysed samples show small differences that are largely due to the contribution of other compounds, such as carbonates and occasionally clay minerals. However, the signals are distinct for each petrographic group because of their compositional differences. These FTIR signals may help separate samples containing only small amounts of these compounds, for example, carbonate in radolarite.

However, a clear link is found by comparing the mineralogical-petrographic features of the lithic tools found at Limba-Oarda de Jos with those of suitable rocks from the surroundings. Geologically, the area is located at the contact point between the Trascău Mts. (part of the Southern Apuseni Mts.) to the west and northwest, the Transylvanian Basin to the east, and the Southern Carpathians to the south (as seen in Figure 1). In the Trascău Mts., there are formations of interest to the lithic industry, such as the Upper Jurassic chert-bearing limestones, Upper Jurassic radolarites, as well as Cretaceous conglomerates, sandstones, clays, and limestones (Giușcă et al. 1967; Ionescu & Hoeck 2010; Săndulescu 1984; Săsăran 2006). The mineralogical and petrographic features of the lithic tools fit well with those described for the above-mentioned geological formations. Hence, the Upper Jurassic chert-bearing limestones and the Upper Jurassic radolarites can be envisaged as possible sources of the cherts, siliceous limestones, and radolarites used to produce the Middle Neolithic lithic tools found at Limba-Oarda de Jos.

Biró and Dobosi (1991), Biagi et al. (2007) and Starnini et al. (2015) concluded that two kinds of radolites, the Mecsek and the Úrkút Transdanubian radolarites, which were frequently used to produce stone tools, originate from the Mecsek Mts. and the Bakoni Mts. (Hungary), respectively. In addition, Biagi and Voytek (2006) assumed an exogenous provenance of the Úrkút Transdanubian radolarite pieces found at the Peștera Ungurească (Caprelor Cave) in the Turda Gorges, which is in the northern end of the Trascău Mts., showing the long-distance travel undertaken with this exotic raw material. In our case, the small number of radolarite tools, cores, and débitage found in situ cannot be underestimated; therefore, a comparative study with Transdanubian radolarite could shed light on the provenance and, possibly, trade routes.
5. Conclusions

At the Neolithic site at Limba-Oarda de Jos (Romania), four rock types were used to produce various tools. Among them, chert, fossiliferous chert, and siliceous limestone were the most frequently used, whereas radiolarite was used to a lesser extent. The typological analysis in relation to petrography reveals a preference for the manufacture of blades from all rock types, with some differences; e.g., the blades made of siliceous limestone are larger than the blades made of other rock types.

The large number of cores and débitage made of chert, fossiliferous chert, and siliceous limestone suggests that the lithic raw material was transported as nodules and that the tools were chipped in situ at the settlement. These nodules most likely originate from the Trascău Mts., where Upper Jurassic limestones (Ionescu & Hoeck 2010) crop out (see Figure 1). By contrast, the small number of radiolarite cores and débitage indicates these materials were initially processed at the source and subsequently transported to the settlement as finished products. Although there are other important radiolarite outcrops, such as the Mecsek Mts. and Bakoni Mts. (Hungary) (Biagi et al. 2007; Biró & Dobosi 1991; Biagi & Voytek 2006; Starnini et al. 2015), the Upper Jurassic radiolarites that co-occur with the Upper Jurassic limestones are close to the Limba-Oarda de Jos site. This factor may have been a reason for choosing them instead of other distant radiolarite outcrops in the Southern Apuseni Mts.

The present petroarchaeological study of siliceous lithic tools from Limba-Oarda de Jos is part of a larger outgoing project focused on the provenance of lithic raw materials at different Paleolithic and Neolithic archaeological sites of Transylvania (Romania). Lithic tools and their raw materials are regarded as spatial-temporal markers. Their lithological analysis and geological contextualization allow researchers to define which territories were controlled at a certain time (Mangado 2006).

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