New chronological constraints for Middle Palaeolithic (MIS 6/5-3) cave sequences in Eastern Transylvania, Romania

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ABSTRACT

The Middle to Upper Palaeolithic transition is one of the crucial periods of change in the prehistory of Europe due to the full emergence, continent-wide, of modern human lithic technologies, and detrimental of Neanderthal survival. Knowledge about the transition is growing, however, the evidence for cultural and technological developments for the Middle Palaeolithic in the Carpathian – Lower Danube Basin is still rather sparse. Here we discuss latest findings arising from a chronological investigation of Middle Palaeolithic assemblages within the Varghis karst, Eastern Transylvania, Romania. Combining our first chronological results with information from previous excavations, we can distinguish two main stages of habitation (albeit Middle Palaeolithic lithics and faunal remains appear scattered throughout the investigated profile) within the Abri 122 rock shelter. In order to augment the typological cultural considerations, we applied direct radiocarbon dating on bones and charcoal from within the occupation layers. Radiocarbon dating of bones suggests that the Middle Palaeolithic sequence is older than the upper dating limit of the method, whereas direct luminescence ages on the lowermost productive horizon and immediately above it indicate surprisingly old ages of ca. 106 – 141 ka (OSL – infrared stimulated) or 99 – 174 ka (IRSL – infrared stimulated). Multiple-protocol dating of charcoal found within the two habitation layers produced ages > 38 14C ka BP, also suggesting that the lowermost lithic-rich horizon pertains to the Middle Palaeolithic industries. Overall, the recovered lithics, currently forming one of the most significant collections for Romania, are fully consistent with two main habitation phases connected to Middle Palaeolithic cultural affinities. The occurrence of a volcanic ash layer within Ursului Cave and originating from the Ciumadul volcanic complex (Carpathians) is first reported here. Recently dated to $\geq 43$ (50) ka, it might represent an important marker horizon, providing that it is identified within other Palaeolithic cave assemblages.

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1. Introduction

The dynamics and timing of different phases of modern human dispersal into Eurasia (McBrearty and Brooks, 2000) and the subsequent demise of Neanderthals around the Middle to Upper Palaeolithic transition are matters of intense scientific debate (Higham et al., 2009; Hublin, 2015; Davies et al., 2015; Richter, 2016; Talamo et al., 2016). Site of the Oase Cave (Trinkaus et al., 2003), and centered on the Danube valley and its large tributaries that cross the Carpathian-Balkan ranges (Fig. 1), south-eastern Europe holds significant potential for better understanding of the environment and palaeoclimate that may have influenced the movement of people towards both central-western Europe (Conard, 2003) or eastwards, over the north Pontic area (Iovita et al., 2012). With some notable exceptions, interdisciplinary

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archaeological research (Anghelinu et al., 2012) and especially direct multi-method dating (Onac et al., 2005; Haesaerts et al., 2010; Schmidt et al., 2013; Trandafir et al., 2015; Doboş and Trinkaus, 2012) has seen limited application in the Carpathian — Lower Danube area — particularly for Middle Palaeolithic contexts (Honea, 1981; Mertens, 1996; Cârciumaru, 1999; Paunescu, 2000; Cârciumaru et al., 2000, 2007; Cosac, 2008).

Since a range of discoveries in Carpathian caves provided a significant number of modern human remains (Sofiărău et al., 2006), these discoveries sparked new interdisciplinary research aimed at augmenting the archaeological database and understanding the environment and regional palaeoclimatic climatic (Steguweit et al., 2004; Anghelinu and Niţă, 2014; Silivy et al., 2014; Fitzsimmons et al., 2013; Zeeden et al., 2011; Zeeden et al., 2017; Obreht et al., 2017). However, understanding the spatial distribution and chronological relationships of modern human — Neanderthal techno-complexes in central-eastern Europe (Kozłowski, 2014; Hublin, 2015; Talamo et al., 2016), or even the relative chronological span of the Palaeolithic in Romania remains challenging (Balescu et al., 2015; Iovita et al., 2012; Anghelinu and Niţă, 2014; Doboş and Trinkaus, 2012). Even more compelling, limited chronological control and the paradigm of potentially late persistence in the Carpathian area of the Middle Palaeolithic alongside an unusually late onset of Upper Palaeolithic industries (Cârciumaru, 1999; Paunescu, 1984, 2000, 2001) call for a new approach in data evaluation (Anghelinu and Niţă, 2014).

This contribution presents the initial multi-method dating results (radiocarbon, luminescence, tephrochronology) of a Middle Palaeolithic occupation complex at Abri 122 and Ursului Cave archaeological sequences within the Varghis karst area, in Eastern Transylvania, Romania (Fig. 1). We discuss the chronological assessment of the archaeological contexts, with the overall goal in establishing a chronological framework that would allow for further comparison of the Abri 122 technocomplex, one of the most compelling in the Carpathian area (Cosac et al., 2017) with similar records, on a regional scale.

2. Study area

The Varghis (locally spelled Vârghiş) karst area (46°12′58″ N; 25°32′36″ E, 670 m a.s.l.) is positioned at the northern edge of Persani Mountains (Eastern Carpathians), as the most important karst system of the area that comprises mostly flysch sediments and Quaternary basaltic monogenetic volcanism (Fig. 1). Here, the Varghis river valley crosses a 3.5 km long band of Triassic and Jurassic limestone (Orghidan and Dumitrescu, 1963), thus forming a 56 km² wide karst system, with more than 100 caves (Fig. 1), several with clear evidence for past human occupation (Denes, 2003). The rock shelter identified as Abri 122 is placed on the right side of the valley, in the lower third of the slope, at 625 m a.s.l., and roughly 30 m above the Varghis riverbed (Denes, 2003). It consists of a sheltered limestone platform, apparently continued with an in-filled small cave room (Fig. 2A–B). The rock shelter with a surface of approximately 30 m² opens to the E-SE and it has been the subject of intense archaeological research during the last decades (Cosac et al., 2017 and references therein).

Ursului Cave, also investigated in this work is located on the higher slopes, in a precipitous terrain today. It consists mainly of a large cave room, with three entrances (Fig. 2E inset) and hosts thick clastic deposits (Orghidan and Dumitrescu, 1963). It has only been preliminarily investigated in the past with regards to its archaeological potential (Mottl, 1950).

3. Materials and methods

3.1. Fieldwork and sample collection

The field excavation at Abri 122 is ongoing (Fig. 2A–C), however,
we report here the sampling and dating results produced in order to augment the interpretation of archaeological results of previous excavations (Fig. 2A–B) as discussed in Cosac et al. (2017). The profile within Abri 122 that was left unexcavated (sections S1 and S2 in Fig. 2A–B) by previous campaigns consists of a cave-in-fill of fine-grained matrix supported reddish silty loam, likely wind-blown in origin, with only occasional boulder-sized limestone clasts. There are no obvious layer boundaries, and the sedimentary succession appears rather homogenous. Albeit charcoal and lithic pieces appear scattered throughout the whole section, there are two main horizons of lithics, animal osseous remains and charcoal fragments at roughly 220–230 cm and 260 cm depth, pointing to the main phases of anthropogenic accumulation of material within Abri 122. Samples for radiocarbon dating (both charcoal and bones) were collected from both horizons, whereas luminescence dating focused on the lowermost lithic-rich horizon (Fig. 2C).

The profile in Ursului Cave (Fig. 2D) has been excavated in order to verify the report of Mottl (1950) on the presence of possible Aurignacian lithic finds there. The profile (Fig. 2D) was dug to a depth of 270 cm from the cave floor following the contours of previous excavations (Mottl, 1950). It consists of a recent humic horizon with plant litter and large limestone blocks (0–70 cm depth), very angular sand to gravel sized limestone clasts with almost no fine matrix between 70 and 150 cm depth, within which a 5–15 cm thick dark-grey volcanic ash horizon (sample URS-1.1) is embedded around 110–130 cm depth (Fig. 2D). The profile continues with a unit of very fine yellowish silty clayey loam (130–200 cm), likely wind-blown material, overlying a massive unit of gravel to boulder sized limestone clasts in a reddish carbonate matrix with traces of hydromorphic coatings and manganese staining (200–270 cm). As the investigation of the nature and archaeological potential of the sedimentary infill within Ursului Cave is ongoing, at this stage samples were collected only from the tephra layer in order to report on its chemical composition and infer its origin and tephrostratigraphic potential.

3.2. Radiocarbon dating

For radiocarbon dating, both charcoal and animal bone samples were collected from the freshly excavated profile at Abri 122 during 2014–2015 (Fig. 2A–C). In the field, care was taken for collecting
only well preserved bone remains for radiocarbon dating; however, due to the overall high degree of fragmentation and friability of the osseous remains, only the samples reported in Table 1 passed the laboratory sample selection and methodological thresholds that enabled further dating attempts.

Charcoal samples were processed according to a standard protocol using the acid–base–acid (ABA) method (Jull et al., 2016) that implies treatment with 1N HCl, rinsing in distilled water, 1M NaOH, rinsing, and 1N HCl acid wash at 75 °C, for 1–2 h each step. After the final acid wash, the samples were rinsed with distilled water to neutral pH (4–5) and dried at 60 °C (Molnár et al., 2013a; Ujvari et al., 2016). The chemically treated charcoal samples were then combusted with a two-stepped protocol in pure O2 gas atmosphere, first at 400 °C, and then at 800 °C (ABA-TSC200 and ABA-TSC800, respectively) in a quartz tube. The resulting CO2 gas was purified in a vacuum line, collected after each combustion step from the applied on-line combustion system and later converted to graphite (Rinyu et al., 2013). Bone samples were treated with the ABA protocol in an automated continuous-flow bone cleaning system (Molnár et al., 2013a). Samples were then combusted off-line in sealed tubes with oxidizing reagent. The resultant CO2 was cryogenically purified and converted into graphite, and the graphitized material was measured using a compact radiocarbon AMS system (MICADAS) at the Hertelendi Laboratory, Debrecen, Hungary (Molnár et al., 2013b). The AMS radiocarbon ages are presented in radiocarbon years before present (BP), where BP refers to 1950 AD.

3.3. Luminescence dating

3.3.1. Luminescence sample collection and preparation

Two luminescence samples were also collected from the freshly excavated deposit at Abri 122 (Fig. 2B–C), sample BT1415 from the lowermost productive cluster (260 cm) and BT1416 from the sterile layer above (250 cm). Sample preparation was carried out under inert gas atmosphere. Sieving (200, 90 and 63 µm) were subjected to heavy liquid (sodium polytungstate) density separation to extract either quartz (2.62 g cm⁻³) or K-feldspars (2.10 g cm⁻³). The finest grains were subjected to etching with hydrofluoric acid (HF) to remove any mineral particles except for quartz. For coarse grains, the samples were bleached in a solar simulator for 3 h and then irradiated with a bi-alkali lamp for 3 h and subsequently irradiated with a 254 nm Schott BGF3 glass filter. The dose rate to fine grains of the in-built 90Sr/87Sr activity is 0.06 Gy s⁻¹.

For IRSL measurements, we used a Risø TL/OSL DA-15 reader (Richter et al., 2013) that stimulates at λ = 870 ± 20 nm (IR-LEDs) with a maximum power density of ~130 mW cm⁻² at sample position. Emitted IRSL was recorded at 125 °C measurement temperature with a bi-alkaline Hamamatsu H7360-02 photomultiplier tube and a Delta BP 365/50 EX bandpass filter in combination with a 3 mm Schott BG3 glass filter. The equivalent dose (Dₑ) was determined with a standard single-aliquot regeneration (SAR) technique following Murray and Wintle (2000) for quartz IRSL and with an adjusted SAR protocol for K-feldspar and polymineral IRSL, including an additional pause of 1200 s after regenerative irradiation (cf. Faust et al., 2015). To test the ability of these protocols to accurately measure radiation doses in the investigated samples and to find optimal measurement parameters (cf. Kim et al., 2009), both preheat plateau tests (OSL of quartz fine grains) and combined preheat and dose recovery tests (DRT; quartz and polymineral fine grains and K-feldspar coarse grains) were conducted. For dose recovery tests, part of the sampled material was bleached in a solar simulator at 120 °C for 3 h and subsequently irradiated with a dose comparable in size to the expected burial dose. Suitable preheat temperatures derived from these diagnostic tests were adopted for the main measurements. The cutheat temperature for OSL measurements was set 40 °C lower than the preheat temperature.

In addition, each aliquot was individually checked for dose reproducibility (recycling ratio in the range 0.90–1.10), absence of thermal transfer (recuperation <5%) and feldspar contamination in case of quartz separates (IR depletion ratio in the range 0.95–1.05). Aliquots not meeting these criteria were discarded.

Table 1: Results of radiocarbon AMS dating of charcoal and animal bone remains from Abri 122 sequence. The radiocarbon dates were calibrated using the IntCal13 calibration dataset (Reimer et al., 2013).

<table>
<thead>
<tr>
<th>Lab. ID</th>
<th>Depth (cm)</th>
<th>Dated material</th>
<th>¹⁴C age (yr BP ± 1σ)</th>
<th>Calendar age (cal. yr BP ± 1σ)</th>
<th>Calendar age (cal. yr BP ± 2σ)</th>
<th>Collagen %</th>
<th>Δ%</th>
</tr>
</thead>
<tbody>
<tr>
<td>DeA-7471</td>
<td>207–217</td>
<td>Charcoal</td>
<td>17,461 ± 154</td>
<td>20,867–21,322</td>
<td>20,657–21,542</td>
<td>n.a.</td>
<td>3.74</td>
</tr>
<tr>
<td>n.a.</td>
<td>217–227</td>
<td>Bone fragment</td>
<td>&gt;42,200</td>
<td>–</td>
<td>–</td>
<td>2.5</td>
<td>--</td>
</tr>
<tr>
<td>DeA-8904</td>
<td>227–237</td>
<td>Bison s. bone collagen</td>
<td>&gt;43,400</td>
<td>–</td>
<td>–</td>
<td>38.1</td>
<td>--</td>
</tr>
<tr>
<td>DeA-5868</td>
<td>260</td>
<td>Ursus sp. tooth collagen</td>
<td>&gt;43,400</td>
<td>–</td>
<td>–</td>
<td>36.2</td>
<td>--</td>
</tr>
<tr>
<td>DeA-5957</td>
<td>260</td>
<td>Charcoal</td>
<td>38,900 ± 1100</td>
<td>42,024–43,697</td>
<td>41,244–44,739</td>
<td>n.a.</td>
<td>9.07</td>
</tr>
</tbody>
</table>

(cf. Mauz and Lang, 2004). As quartz coarse grains of both samples still showed a significant IRSL signal even after repeated HF etching, this grain-size fraction was excluded from further analyses.
Feldspars often exhibit anomalous fading, an effect that we tried to reduce by adopting the adjusted IRSL-SAR protocol as outlined above. Nevertheless, we conducted fading tests consisting of constant-dose regeneration cycles, interrupted by pauses of varying duration between irradiation and IRSL measurement (cf. Auclair et al., 2003).

3.3.3. Dose rate assessment

As in-situ measurements of dose rate in $4\pi$ geometry were not possible at the Abri 122, we used a Thermo Fisher RadEye radiation detector to assess the horizontal variation of $\gamma$-dose rate within the lowestmost lithic-rich horizon (roughly 250–260 cm depth; Fig. 2), which amounts to $<25–30\%$ of the average value. In addition, we took representative sediment samples from the vicinity ($\sim 20–30$ cm) of the luminescence samples for determining the K, Th and U concentrations through thick-source $\alpha$-counting and ICP-OES techniques (Table 2). The obtained values were converted into effective dose rates using conversion factors as given in Guerin et al. (2011) and assuming a $\alpha$-value of $0.030 \pm 0.003$ (Mauz et al., 2006) and $0.075 \pm 0.010$ (based on measurements in Kreutzer et al., 2014) for fine grain quartz and polymineral samples, respectively. The internal K concentration of K-feldspar grains was estimated as $12.5 \pm 0.5\%$ (Hunty and Baril, 1997).

The contribution of cosmic radiation to the dose rate was modelled by assuming that due to shielding of the Abri 122 and the overlying limestone cliff only half of the otherwise relevant cosmic dose rate was received by the samples. Based on constant sedimentation rate as well as the longitude, latitude and altitude of the site, the cosmic dose rate was computed with the R package ‘Luminescence’ (v. 0.6.4; Kreutzer et al., 2012a,b, 2014; R Development Core Team, 2016). A moisture content of 15 ± 5 wt% (Table 2) was taken to correct the dose rate for absorption of radiation in pore water.

3.4. Tephra methods

A 5–15 cm thick and laterally continuous dark grey tephra layer was identified at 110–130 cm depth within the stratigraphic profile in Ursului Cave (profile A1; Fig. 2E inset) that was excavated in order to verify the report of Mottl (1950) on the presence of Aurignacian industries within the cave. The tephra is embedded within a clastic unit comprising numerous angular limestone clasts in sand to gravel sizes, likely representing frost-shattered material from the cave walls. The Ursului Cave tephra sample (URS–1.1) was treated with a 15% hydrogen peroxide (H$_2$O$_2$) and a 10% hydrochloric acid (HCl) solution to remove the organic remains and carbonates, and wet-sieved into a 20–100 µm grain size fraction. The material was then embedded on a slide with Aralditeâ®2020 resin, sectioned by hand on silicon paper, polished and carbon coated. The major element composition of glass shards was determined with a JEOL JXA-8230 instrument at the GFZ Potsdam using a 15 kV voltage, 10 mA beam current and beam sizes of 8 µm, respectively. Exposure times were 20 s for the elements Fe, CI, Mn, Ti, Mg and P, as well as 10 s for Si, Al, K, Ca and Na. Instrumental calibration employed the rhyolithic Lipari obsidian glass standard (Hunt and Hill, 1996; Kuehn et al., 2011). Glass major oxide chemical data are reported in Table 3 and further shown in Fig. 6.

4. Results and discussion

The chronological data reported here were obtained on samples collected from the newly excavated profile at Abri 122 whose investigation is ongoing (the interpretation of the most recent archaeological findings are to be presented elsewhere, upon further research). There were two main horizons rich in lithics and faunal

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Table 2: Radiocarbon results (in ka) for the two horizons from Abri 122

<table>
<thead>
<tr>
<th>Sample</th>
<th>Depth (m)</th>
<th>Water content (wt%)</th>
<th>D$_{18}$ (g)</th>
<th>Water content assumed (wt%)</th>
<th>D$_{13}$ (g)</th>
<th>K [wt.%]</th>
<th>Th [ppm]</th>
<th>U [ppm]</th>
<th>Water content measured (wt%)</th>
<th>D$_{0}$ (g)</th>
<th>Recuperation Ratio</th>
<th>K [wt.%]</th>
<th>$\alpha$-value [%/decade]</th>
</tr>
</thead>
<tbody>
<tr>
<td>BT1416</td>
<td>1.30</td>
<td>1.03</td>
<td>1.30</td>
<td>1.03</td>
<td>1.30</td>
<td>0.99</td>
<td>0.01</td>
<td>0.03</td>
<td>0.99</td>
<td>1.30</td>
<td>1.03</td>
<td>1.03</td>
<td>0.01</td>
</tr>
<tr>
<td>BT1415</td>
<td>1.40</td>
<td>1.03</td>
<td>1.40</td>
<td>1.03</td>
<td>1.40</td>
<td>0.99</td>
<td>0.01</td>
<td>0.03</td>
<td>0.99</td>
<td>1.40</td>
<td>1.03</td>
<td>1.03</td>
<td>0.01</td>
</tr>
</tbody>
</table>

*Minimum age estimate (see main text for further details).
remains, at around 220–230 cm and respectively 260 cm depth, both representing clear Middle Palaeolithic typological affinities (Cosac et al., 2017). Moreover, the spread of lithics along the vertical profile as well as the clear correspondence between previous finds and our recent survey suggest that the chronological estimates discussed here are representative for the entire Middle Palaeolithic archaeological sequence at Abri 122 (Cosac et al., 2017).

4.1. Radiocarbon dating results

Radiocarbon dating results are summarized in Table 1. The results show that the lowermost productive horizon at 260 cm depth has a chronological span most likely beyond the upper limit of radiocarbon dating, as indicated by the inclusions of charcoal at 237 cm depth that produced an age of 42,200 ± 14C yr BP (see Table 1) as beyond the upper limit of radiocarbon dating. However, one shall take into account the significant uncertainties related to the dating of old bone remains subjected to contamination and degradation, as well as the limitations arising from laboratory pre-treatment issues (Brock et al., 2010; Talamo and Richards, 2011).

The uppermost dated charcoal sample at 207–217 cm depth (treated through the ABA-TSC400 protocol) consisted of scattered small fragments of charcoal, and is thus, perhaps more prone to contamination (cf. Újvári et al., 2016). The 20,657–21,542 cal yr BP range for this sample must be treated with caution, albeit the dated material was not directly associated with Middle Palaeolithic lithics and could also suggest that the upper part of the sedimentary infill at Abri 122 is much younger than previously considered.

4.2. Luminescence data evaluation

Luminescence measurements were analyzed with the Analyst software (v.4.319; Duller, 2015) to calculate the Dn values of individual aliquots. Dose response curves were constructed with signals resulting from integration of the first 0.5 s (OSL) or 5 s (IRSL) of the decay curves from which a background averaged from the last 0.5 s (OSL) or 20 s (IRSL) was subtracted. Dose response curves were fitted with an exponential-plus-linear function. A representative IRSL dose response curve of sample BT1415 is shown in Fig. 3.

The final Dn was estimated from the arithmetic mean of individual contributing aliquots for all samples. As was just one dose population resulting from coarse grain measurements of K-feldspar, no age model was applied. Furthermore, recent investigations have shown that in settings with potential spatial heterogeneity in β-radiation — as is often the case in caves or wall-
sheltered archaeological contexts — the arithmetic mean of individual \( D_e \) values appears to best approximate the true burial dose provided that all contributing grains represent the same OSL/IRSL age (Guérin et al., 2013).

Fading correction according to the method developed by Huntley and Lamothe (2001) was performed using the R package ‘Luminescence’ (v. 0.6.4; Kreutzer et al., 2012a,b, 2014; R Development Core Team, 2016). Ages were computed using the software Adele (v. 2015 0.21a). The systematic error of \( b \)-source calibration was not propagated, but an assumed relative uncertainty of 4% was added in quadrature to the overall error on \( D_e \). All results from analytical and luminescence measurements are compiled in Table 2. The \( D_e \) distribution of sample BT1415 (coarse grained K-feldspar) is shown as Abanico plot in Fig. 4.

Results of the preheat plateau and dose recovery tests indicate suitable preheat temperatures of 240–260 °C for OSL of fine-grained quartz and of 250 °C for IRSL of polymineral fine grains (see Fig. 5). Dose reproducibility was better than 8% for OSL of sample BT1415, better than 1% for polymineral fine grain IRSL of sample BT1416 and better than 9% for K-feldspar coarse grain IRSL of sample BT1415 for preheat temperatures of 240 °C, 250 °C, and 270 °C, respectively. Fading tests yielded consistent fading rates of \( -3.2 \pm 1.0 \)% per decade for the two polymineral fine grain samples. The measured fading rate of polymineral sample BT1415 was adopted for the coarse grain K-feldspar fraction of this sample.

However, as the natural OSL signal of sample BT1415 was very close to the saturation level of the laboratory dose response curve, the determined \( D_e \) for this sample (Table 2) and hence the OSL age must be regarded as a minimum estimate. This is a known observation for quartz OSL SAR in the dose range approaching saturation (e.g., Murray et al., 2007).

As the luminescence signal of fine grain separates is averaged from the order of \( 10^6 \) grains per measurement, such analyses do not provide information on the degree of complete signal resetting prior to burial. We therefore investigated coarse grain K-feldspars of sample BT1415 (Table 2) while reducing aliquot size to the minimum to evaluate the distribution of \( D_e \) values (cf. Duller, 2008).

Due to low light levels, it was not possible to measure less than 70–80 grains at once so that signal averaging might still have played a role. Although the number of measured aliquots is not sufficient to develop a conclusive statement (Rodnight, 2008), it appears that there is just one dose population (Fig. 4). Calculating the arithmetic mean of all \( D_e \) values results in an age identical to
that of polynuclear fine grains of this sample (Table 2). Excluding the highest $D_e$ value from the coarse grain measurements still gives a $\pm 4\%$ younger age not significantly different from the polynuclear fine grain age (Table 2). Increasing the number of measured aliquots is not expected to yield a bimodal or polymodal $D_e$ distribution demanding the application of age models. Given the geomorphological setting at Abri 122, fluvial transport of material can be excluded. Furthermore, the preliminary microfaunal analyses (Cosac et al., 2017) indicate that the sediment at Abri 122 has been limited (or even no) remobilization; it is likely that the investigated materials were mainly delivered to the site by wind activity, thus favoring good bleaching conditions.

4.3. Chronological considerations at abri 122; radiocarbon versus luminescence ages

The luminescence dating of the Middle Palaeolithic technocomplex at Abri 122 (Cosac et al., 2017) thus indicates that material within the lowermost productive horizon was deposited around $141 \pm 12$ ka based on OSL dating of quartz (BT1415). IRSL dating of the same sample provided closely matched ages of $173 \pm 41$ ka for feldspar grains, or $174 \pm 37$ ka on polynuclear grains (Table 2). Radiocarbon dating of animal bones from the same level indicated infinite ages (Table 1), whereas charcoal yielded contrasting results with age estimates in the range of 42010–41567 cal yr BP for bulk charcoal (multiple small charcoal pieces scattered around 260 cm depth). Although this radiocarbon age is close to the upper limit of the method, the age difference between bone collagen and charcoal fragments could also be explained by the sensitivity of sedimentary charcoal to chemical contamination by soluble humic substances (Alon et al., 2002; Rebollo et al., 2011; Wild et al., 2013), as well as chemical oxidation and microbial degradation (Alon et al., 2002; Cohen-Ofri et al., 2006; Ascough et al., 2011). Given the sedimentary context at Abri 122 (exposed to rainwater percolation, and rich plant litter and animal remains) and the small amounts of sample that likely reflects in-situ charcoal fragmentation (Cohen-Ofri et al., 2006), the charcoal dating results shall be regarded as minimum age estimates. As special care was taken in removing exogenous carbon through the acid—base—acid (ABA) treatment, this also resulted in a mass loss prior to analysis (the use of the chemical ABA treatment has been shown that in some circumstances (Higham et al., 2009; Douka et al., 2010; Wood et al., 2012) may result in slightly younger ages than other pre-treatment methods).

The difference between radiocarbon and luminescence ages of the same level (Fig. 2) is however significant. Nonetheless, it could be stated that the luminescence dating provided a maximum age for the Middle Palaeolithic assemblage at Abri 122, that is, within or prior to Marine Isotope Stage (MIS) 5. This age range is further constrained by more luminescence results obtained on sample BT1415, 10 cm above and dated to $99 \pm 17$ ka (IRSL age on polynuclear grains) or $106 \pm 11$ ka on quartz grains (Table 2) that could be considered a likely age for the lower productive horizon (Fig. 2).

The radiocarbon dating of herbivore bone collagen from the upper main horizon of tools/bones at around 227–237 cm depth also yielded an age $>42200 ^{14}$C yr BP, and thus beyond the upper limit of the method. Another radiocarbon date $10$ cm above it on scattered pieces of charcoal (over $20$ cm depth interval) yielded an intriguing young age of $17461 \pm 154 ^{14}$C yr BP (the validity of this age determination not directly associated with Middle Palaeolithic lithics must be verified through further work).

Overall, the dating approach provides clear estimates for the long timespan of accumulation of human-related material within the Middle Palaeolithic assemblage at Abri 122, starting likely from or before MIS 5 and extending throughout MIS 3, as indicated also by the microfaunal analysis (Cosac et al., 2017).

4.4. Glass-shard geochemistry and tephrochronological considerations

The URS-1.1 tephra is a medium to fine sand-sized pyroclastic fall dominated by lithic clasts representing mainly dacitic rock fragments and microlite-rich (feldspars, pyroxenes) pumices and blocky, vesicular glass shards (Fig. 2E). Other phenocrysts include plagioclase, green pyroxene and biotites. The major element glass composition (normalized, volatile-free data) indicates calc-alkaline rhyolitic affinities with $SiO_2$ concentrations of 75.0–77.9 wt%, $Al_2O_3$ at 12.3–14.4 wt%, $FeO$ at 0.4–0.8 wt% and $CaO$ around 0.6–1.1 wt%. The alkali ratios ($K_2O/Na_2O$) vary between 0.9 and 1.8 wt%, and total alkali ($K_2O + Na_2O$) at 71.7–91.1 wt% (Table 3; Fig. 6). This glass composition is typical for Late Pleistocene tephras of the nearby Ciomadul volcanic complex in the East Carpathians (Figs. 1 and 6).

A detailed tephrostratigraphic record of the latest explosive phases of Ciomadul for the $>51–29.6$ ka time interval has recently been published (Karatzon et al., 2016, and references therein). Three major eruptive phases were identified, based on glass-shard major oxide compositions, stratigraphic considerations and detailed multi-proxy dating of pyroclastic deposits outcropping around Ciomadul volcano (Fig. 6). These include an early EPPA phase (Early Phreatomagmatic and Plinian Activity), with a succession of phreatomagmatic events at $>51$ ka, called Turia eruption(s) (Karatzon et al., 2016), which produced widespread tephra with highly evolved rhyolitic glass compositions (78–79 wt% $SiO_2$). The EPPA phase terminated at $<43 (\pm 50$) ka for the so-called BTS (for Baile Tusnad locality; Fig. 1), eruption(s) characterized by pumice fall and pumiceous pyroclastic flow deposits with slightly less evolved and more homogeneous EPPA-type major oxide glass compositions (76–78 wt% $SiO_2$) (Karatzon et al., 2016) (Fig. 6). The latest Ciomadul eruptions at c. 31.5 ka called the MPA (Middle Plinian Activity) stage and at the 29.6 ka LSMA (Latest St. Ana Phreatomagmatic Activity) stage resulted in the formation of SF. Ana crater. These latest major eruptive events produced tephra layers with a less silicic glass composition (mean $SiO_2$ of c. 73 wt% and 76 wt%, respectively), and are distinguishable from the EPPA phases in the major oxide bi-plots (Fig. 6). The data show that the Late Quaternary Carpathian volcanism (Seghedi et al., 2004; Karatzon et al., 2013; Harangi et al., 2015; Szakacs et al., 2015) produced a series of calc-alkaline rhyolitic tephra layers, with a broad areal coverage (Wulf et al., 2016; Karatzon et al., 2016, 2017) and occurrence within a period of extreme environmental changes in the region (Veres et al., 2013; Fitzsimmons et al., 2013; Magyari et al., 2014; Zeeden et al., 2017; Obreht et al., 2017).

With average (normalized) glass shard compositions of 76.63 wt % $SiO_2$, 13.14 wt% $Al_2O_3$, $TiO_2$ of 0.10 wt% and total alkali at 8.58 wt% (Fig. 6), the tephra identified within Ursului Cave might correlate with the final eruptive phase(s) of the early EPPA stage dated at $>43 (\pm 50$) ka (see chronological discussion in Karatzon et al., 2016). However, the correlation of the thick pyroclastic-fall and
pyroclastic-flow sequence (BTS) identified on the western flanks of Cioamadul volcano with the ash bed within Ursului Cave, 30 km to the west suggest that the EPPA eruptive phases spread materials much further than hitherto considered. This raises the possibility of both tracing the Carpathian volcanic products much wider and in establishing their value as isochronous marker horizons (Wulf et al., 2016) for various records, including Palaeolithic cave assemblages.

Mottl (1950), who first performed an archaeological survey of the Varghis karst, provided no stratigraphic information of the excavated profile within Ursului Cave. Albeit we cannot assess whether Mottl (1950) recognized the presence of a volcanic ash bed within Ursului Cave and that no other chronological estimates exist for the archaeological profile there, the mentioning of a few (potentially) Aurignacian lithic finds is intriguing. As our preliminary survey within Ursului Cave returned no discriminatory lithics and only a limited number of faunal remains, it remains to be established by further research whether this volcanic ash bed is widespread within the Varghis karst archaeological sites. Its age at the crucial transition between the Middle and Upper Palaeolithic will enhance its potential as a key marker horizon (Lowe et al., 2015) similar to other tephra reports in the wider region (Constantin et al., 2012; Veres et al., 2013; Fitzsimmons et al., 2013; Anechitei-Deacu et al., 2014; Wulf et al., 2016).

4.5. Regional considerations

In an accompanying paper, Cosac et al. (2017) suggest that the technological assemblage of lithics found so far at Abri 122 unequivocally pertain to Middle Palaeolithic industries but with quite distinctive features compare to other sites in Romania. Its individuality resides in the peculiar technological and typological features identified, clearly exhibiting more than one technological option (i.e., discoid, centripetal Levallois, most likely also Kombeva; see Cosac et al. (2017) for a discussion of archaeological finds). The toolkit identified is also intriguing, with few denticulated, notched and truncated items, alongside numerous types of sidescrapers and bifacial implements in various stages of manufacture and use. As far as we know, such particular mix of typological features remains unprecedented in the larger framework of Middle Palaeolithic in Romania. Albeit such cultural mixtures are usually common within karst archaeological deposits, we refrain from elaborating on the possibility of a combination between two (or more) Middle Palaeolithic technological/typological features until more detailed chronological and archaeological data will become available from other sites within the Varghis karst (Cosac et al., 2017).

On one hand, it is well known that Middle Palaeolithic assemblages with bifacial modified lithic implements found throughout Europe span a broad timeline with markedly different typological signatures (Kot, 2014; Kozlowski, 2014; Demidenko, 2015; Davies et al., 2015; Richter, 2016). On the other hand, understanding the spatial distribution and chronological relationships of the Middle Palaeolithic techno-complexes in Europe (Kozlowski, 2014; Richter, 2016; Talamo et al., 2016), or even the relative chronological span of the Palaeolithic in Romania remains challenging (Honea, 1981; Paunescu, 1894, 2000, 2001; Mertens, 1996; Carciumaru, 1999; Cosac, 2008; Iovita et al., 2012; Anghelnicu and Nitţa, 2014; Dobos and Trinkaus, 2012). Nonetheless, the multi-stratified Middle Palaeolithic open-air sites of Mamaia Sat, as well as several other Middle Palaeolithic sites from the Carpathian area provide some interesting chronological analogies. For example, Dobos and Trinkaus (2012) proposed a minimum age of >45,500 14C yr BP for the Mousterian assemblages at Ricipeni-Izvor, one of the best-studied Middle Palaeolithic sites in the region. Such an upper chronological span for Middle Palaeolithic assemblages would be consistent with other radiocarbon dating reports from other sites across the region (Mertens, 1996; Carciumaru et al., 2000), as well as the upper horizon rich in lithics at Abri 122 (Fig. 2C).

At Mamaia Sat, a site near the Black Sea, the initial stratigraphic overview tentatively assigned the two distinct intraloessic Middle Palaeolithic lithic-rich horizons to the Last Glacial (MIS 5-5.5) and the Early Last Glacial (MIS 5.3–5.1). Balescu et al. (2015) proposed a rather different chronological context for the Mamaia Sat archaeological assemblage based on established pedostratigraphic linking between Dobrogean loess-paleosol sequences, substantiated by luminescence dating. Thus, following Balescu et al. (2015), the Middle Palaeolithic lithic-rich horizons identified at Mamaia Sat likely pertain to MIS 9 (within the S3 paleosol; Markovic et al., 2015) and the early part of the S2 paleosol (i.e., MIS 7). Even though in the absence of direct dating of the Mamaia Sat loess sequence harboring the archaeological finds, the chronological context proposed by Balescu et al. (2015) remains tentative, the direct IRLS dating of Abri 122 indicates that the lowermost lithic-rich horizon is of early MIS 5 age or older, or perhaps of comparable timeline with the uppermost horizon at Mamaia Sat. For the upper horizon at Abri 122 however, radiocarbon results, and especially the microfaunal analysis (Cosac et al., 2017) would indicate faunal assemblages characteristic of environments younger than MIS 5, likely as young as MIS 3. This suggests that Abri 122 likely preserves a record of Middle Palaeolithic occupation over a long timespan, but the limited dating and the complex technological context identified so far requires further research at other sites within the Varghis karst area.

5. Summary

A multi-method chronological investigation of the Middle Palaeolithic archaeological material at Abri 122 within the Varghis karst, Romania was performed. Albeit the investigation of Palaeolithic cultural assemblages within the Varghis karst complex is ongoing, the available data indicate that the technological assemblage of lithics found within the two main clusters of human occupation at Abri 122 pertain to the Middle Palaeolithic industries. This assumption is further supported by the OSL and IRLS dating (both methods applied on the same luminescence samples) that indicates the material within the lowermost productive layer may have been deposited ≥ 100 ka. Radiocarbon dating of animal bones from the same layer returned infinite ages, whereas charcoal remains subjected to different protocols yielded contrasting results. The dating of bones from the upper cluster of lithics produced also infinite radiocarbon ages. In the light of existing data, it is likely the Middle Palaeolithic assemblage documented at Abri 122, one of the most compelling in the Carpathian region, can be assigned to MIS 5 (or most likely beyond) to MIS 3. The identification of a macroscopic volcanic ash bed within Ursului Cave that originated from the Ciomadul volcanic complex (Carpathians) is also first reported here. Recently dated to ~ ≥ 43 (~50) ka, this volcanic ash might form a key marker horizon at the crucial Middle to Upper Palaeolithic transition, providing that it is traced within other Palaeolithic sequences in the wider region.

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