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The earliest evidence for mechanically delivered projectile weapons in Europe

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# 1 **The earliest evidence for mechanically delivered projectile weapons in Europe**

2  
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16  
17 **Microscopic analysis of backed lithic pieces from the Uluzzian technocomplex (45–40**  
18 **thousand yr ago) at Grotta del Cavallo (southern Italy) reveals their use as mechanically**  
19 **delivered projectile weapons, attributed to anatomically modern humans. Use-wear and**  
20 **residue analyses indicate that the lithics were hunting armatures hafted with complex**  
21 **adhesives, while experimental and ethnographic comparisons support their use as projectiles.**  
22 **The use of projectiles conferred a hunting strategy with a higher impact energy and a**  
23 **potential subsistence advantage over other populations and species.**  
24

25 The Uluzzian was traditionally recognized as one of the Middle to Upper Palaeolithic transitional  
26 cultures in southern Europe (that is, Italy and Greece), but has been recently redefined as an Early  
27 Upper Palaeolithic culture<sup>1</sup>. Grotta del Cavallo (Fig. 1), excavated by A. Palma di Cesnola and P.  
28 Gambassini between 1963 and 1986, is a pivotal site for the Uluzzian because its stratigraphic  
29 sequence includes three main Uluzzian layers, EIII (archaic Uluzzian), EII-I (evolved Uluzzian) and  
30 D (final Uluzzian)<sup>1</sup> (Supplementary Fig. 1), sandwiched by the tephra Y-6 at  $45.5 \pm 1.0$  thousand  
31 years ago (ka)<sup>2</sup> and Y-5 (Campanian Ignimbrite) at  $39.85 \pm 0.14$  ka (refs.<sup>2,3</sup>). The Uluzzian  
32 technocomplex exhibits features that are typically associated with modern human assemblages  
33 (Supplementary Information 2) and characterized by the presence of ornaments, bone implements<sup>4</sup>,  
34 colouring substances<sup>5</sup> and crescent-shaped backed pieces made on small blades or bladelets<sup>1</sup>. These  
35 crescent shaped backed pieces (also referred to as lunates or segments) are a hallmark<sup>1,6</sup> of the  
36 Uluzzian and exhibit no techno-morphological link to the Mousterian or Initial Upper Palaeolithic  
37 assemblages in Europe before the Uluzzian. Similar backed pieces on bladelets have been observed  
38 in East Africa, although there is no archaeological evidence indicating a route from East Africa into  
39 Europe<sup>5</sup>. To better understand the differences between the Uluzzian and earlier lithic traditions, as  
40 well as the importance of the emergence of this new technocomplex in Europe, it is crucial to  
41 identify the function of the backed pieces.

42 The excavations of Grotta del Cavallo unearthed numerous backed pieces<sup>6</sup>, and we undertook a  
43 systematic use-wear analysis of a total of 146 of them from the three Uluzzian layers. This analysis  
44 indicates that the major function of the Uluzzian backed pieces was hunting (Supplementary Table

1). Only seven pieces were used for functions other than hunting (cutting and scraping). Out of the 146 backed pieces, 26 show 55 diagnostic impact fractures (DIFs), which form only when stone tips hit an animal target (Fig. 2). Among them, 9 backed pieces (34.6%) bear DIFs only at a single location, while 17 (65.4%) yield multiple DIF types (Supplementary Table 2 and Supplementary Fig. 2). As several projectile trials resulted in no fractures or only non-diagnostic ones<sup>7,8</sup>, the number of DIFs indicates the minimum number of specimens used as hunting weapons. Six pieces showed microscopic linear impact traces (MLITs) as well (Fig. 2a,f), proving that they were securely used as hunting armatures. Most of the Uluzzian backed pieces showed residues on the back, suggesting that this portion was covered by a type of adhesive (Supplementary Fig. 3). We therefore performed Fourier transform infrared (FTIR) spectromicroscopy on these pieces to characterize the chemical nature of the residues and identified them as a mixture of both organic and inorganic components, mainly ochre, a plant/tree gum and beeswax. The main absorption bands attributed to the organic fraction are highlighted by the grey shaded areas in Fig. 2o (see Methods for more details). In addition, FTIR spectroscopy analyses of several red deposit and soil samples recovered from Grotta del Cavallo enabled us to rule out the presence of organic contaminants from the burial environment and to confirm the presence of ochre as a mixture of silicate and iron oxides by correlative scanning electron microscopy/energy dispersive X-ray (SEM/EDX) measurements (see Supplementary Figs. 4 and 5). Together, the results allowed us to postulate that the three adhesive components had been intentionally mixed, as known in the middle Upper Palaeolithic context<sup>9</sup>. To reconstruct the hafting modes of Uluzzian backed pieces, the frequency of the DIF types (Supplementary Fig. 2) was compared with those obtained by projectile experiments with backed piece replicas<sup>10,11</sup>. The projectile experiments indicated that hafting as barbs resulted less often in multiple DIFs, compared with when the pieces were hafted as tips. Among the multiple DIF types, the type a2m (flute-like, burin-like or transverse fractures from bidirectional ends) was dominant in the Cavallo backed pieces (Fig. 2b–f) and occurred only in experiments with tip hafting (straight/oblique hafting). We do not rule out the possibility that some Uluzzian backed pieces were hafted as barbs because of the relatively high frequency of type a2 (burin-like fracture from steep angle) (Fig. 2a), which occurred in barb hafting as well. However, the frequency of the DIF types suggests that several Uluzzian backed pieces were attached on the tip of a wooden shaft. Uluzzian backed pieces are notably small: complete or almost complete backed pieces with DIFs measured an average of 27.1 mm in length, 10.5 mm in width and 4.6 mm in thickness (Supplementary Fig. 6a). The tip cross-sectional area (TCSA) and tip cross-sectional perimeter (TCSP) of Cavallo backed pieces with DIFs were compared with those of ethnographic North American dart tips and arrowheads<sup>12,13</sup>. The box plots of the TCSA and TCSP of the Uluzzian backed pieces with DIFs fell within the range of those of North American ethnographic arrowheads, but were concentrated on a smaller range (Supplementary Fig. 6b,c). The Uluzzian backed pieces are significantly smaller than the ethnographic dart tips in terms of TCSA and TCSP (TCSA:  $t = -9.414$ ,  $P < 0.05$ ; TCSP:  $t = -13.650$ ,  $P < 0.05$ ), and even smaller than the ethnographic arrowheads (TCSA:  $t = -2.773$ ,  $P < 0.05$ ; TCSP:  $t = -5.709$ ,  $P < 0.05$ ). The extremely small dimensions of the Uluzzian backed pieces suggest that they are suitable for neither thrusting nor throwing spear tips (Supplementary Fig. 7a,b). Despite the small size, the DIFs found on Cavallo backed pieces are relatively large: the largest DIF measures 24.7 mm in length, and 9 DIFs are larger than 10 mm. Several pieces show a significant eduction in the body due to impact damage (Fig. 2b,d,e). Even if specimens retain almost their original length, they often bear elongated DIFs along the side or on

90 the surface. The lengths of several elongated DIFs (flute- and burin-like fractures) exceed 20% of  
91 the entire length of the backed pieces, and four DIFs have a length greater than half the entire length  
92 of the specimens (Supplementary Table 3). The relatively large dimensions of DIFs suggest that  
93 the backed pieces were delivered at high impact velocities. As several Uluzzian backed pieces were  
94 hafted on the tip of a wooden shaft, the small dimensions of the backed pieces must reflect the small  
95 diameter of the shaft. If a thinner shaft is used, the total size of the hunting weapon is smaller.  
96 Therefore, large DIFs, as well as multiple DIF types, occur only when the impact velocity is as high  
97 as is found for mechanical delivery, such as by a spearthrower or bow<sup>8</sup>. Although the TCSA and  
98 TCSP values indicate that the projectile capability of the Uluzzian backed pieces is closer to that of  
99 the North American arrowheads than to that of dart tips, we do not have sufficient information to  
100 discriminate between them. Nonetheless, because of the assumed velocity based on the DIF pattern,  
101 it is more plausible that the Uluzzian backed pieces were projected  
102 using either a spearthrower or a bow. A higher impact energy, however, requires more stable  
103 hafting, since otherwise, stone tips can easily be displaced. A complex mixture, characterized by the  
104 addition of beeswax and ochre, increases the mechanical properties of the adhesive, making it less  
105 brittle<sup>14</sup>. The use of the complex adhesive demonstrated by FTIR spectroscopy in this study  
106 suggests that hunters at Grotta del Cavallo used advanced hafting technology for projectiles with a  
107 higher impact velocity. While the mechanical projectile system enables a higher impact velocity and  
108 long-range shooting, fletching to the base of the shaft is necessary to propel armatures in a straight  
109 trajectory. The discovery of cut marks due to the removal of feathers from bird remains at the  
110 Uluzzian site of Castelcivita (southern Italy) (Supplementary Information 3) indicates that the  
111 fletching technology was also practiced by the Uluzzian people. The multiple findings, such as use-  
112 wear patterns, significant smallness of the Uluzzian backed pieces and complex adhesives, for  
113 Grotta del Cavallo samples dated between 45 ka and 40 ka constitute the earliest evidence for the  
114 use of mechanically delivered projectile weapons in Europe, which is more than 20,000 years  
115 earlier than previously thought. In Europe, the earliest direct evidence for spearthrowers was found  
116 from a Solutrean layer at Combe Saunière, France, dated between ~23 ka and ~20 ka (ref. <sup>15</sup>), and  
117 for bows and arrows preserved in peat bogs at an Ahrensburgian site of Stellmoor, Germany, at  
118 12.9–11.7 ka (ref. <sup>16</sup>). Taking into account that most of the ethnographic spearthrowers are made of  
119 perishable materials, such as wood<sup>17</sup>, it is no wonder that we have only much younger  
120 archaeological remains of spearthrowers and bows and arrows. Neanderthals used wooden spears<sup>18</sup>  
121 and might also have used stone-tipped ones<sup>19</sup>. Their possible stone spear tips, including Levallois  
122 and Mousterian points, are overall much larger than the Upper Palaeolithic points<sup>20</sup>. Although  
123 micropoints recovered from layer E (Neronian) of Grotte Madrin, France, that might be ~5,000  
124 years older than the Uluzzian appearance in Europe are significantly small<sup>21,22</sup>, a systematic use-  
125 wear analysis is required to detect their function. Based on the current state of studies on  
126 Neanderthal hunting<sup>23</sup>, their spears were basically hand delivered (thrusting or throwing), but not  
127 mechanically projected. Conversely, evidence from Africa suggests that modern humans innovated  
128 mechanically delivered projectile weapons before they expanded out of Africa<sup>20,24</sup>.  
129 Although the association between the Uluzzian technocomplex and modern humans has been  
130 challenged<sup>25</sup>, the information currently available from Grotta del Cavallo links the Uluzzian to  
131 modern humans. In particular, the two deciduous teeth retrieved from the Uluzzian layers of Grotta  
132 del Cavallo were attributed to modern humans<sup>26</sup>, and their association with the Uluzzian materials  
133 has been recently confirmed by excavation field notes<sup>1</sup> (Supplementary Information 1) and the  
134 stratigraphic sequence<sup>2</sup>. If further studies confirm the attribution of the Uluzzian to modern humans,

135 we suggest that modern humans equipped themselves with new projectile technology when they  
136 migrated into Europe at around 45 ka. Zooarchaeological data on faunal remains from Grotta del  
137 Cavallo indicate more intensive exploitation of young horses at the Uluzzian levels than that seen at  
138 the late Mousterian (Supplementary Information 4). Considering the fact that young  
139 horses are protected by stallions<sup>27</sup>, the intensive hunting of young horses may reflect skilled long-  
140 range hunting in the Uluzzian. As mechanically delivered armatures allow more accurate hunting<sup>28</sup>  
141 while keeping a greater distance from potentially dangerous prey than hand-delivered hunting (but  
142 see ref. <sup>29</sup>), this new projectile technology could have offered modern humans an advantage in  
143 subsistence strategies.

144

## 145 **Methods**

146 **Functional analysis.** A use-wear analysis was undertaken via a low-power approach<sup>30–33</sup> and a  
147 high-power approach<sup>34–37</sup>. Out of the 146 backed pieces, 34 pieces were recovered from layer EIII,  
148 60 pieces from layer EII-I, 30 pieces from spit E-D and 22 pieces from layer D. Traces were  
149 observed using a Hirox KH7700 digital microscope at magnifications ranging from  $\times 20$  to  $\times 50$  for  
150 macrotraces and from  $\times 140$  to  $\times 480$  for microwear traces. DIFs were analysed using projectile  
151 experiments with backed pieces<sup>7,8,38,39</sup>. The DIFs observed on archaeological materials were  
152 recorded using the microscope mode of the Olympus TG-4 digital camera. Besides DIFs, 11 backed  
153 pieces exhibited possible impact fractures, but we cannot rule out the possibility that they formed  
154 accidentally due to knapping, retouching or post-depositional processes<sup>7,39–41</sup>. For instance, pseudo-  
155 impact fractures, including tiny flute- and burin-like fractures smaller than 5 mm, can occur  
156 throughout production and post-depositional processes. We therefore did not define these fractures  
157 as DIFs. The use of the bipolar technique on an anvil in retouching the Uluzzian backed pieces may  
158 create specific pseudo-impact fractures. We therefore conducted an experiment on the production of  
159 Uluzzian backed pieces to avoid the risk of misidentifying bipolar pseudo-impact scars as DIFs.  
160 After the careful observation of experimental backed pieces, we confirmed that although bipolar  
161 retouching sometimes produces mimic DIFs, we can distinguish these from real DIFs using the  
162 presence of a negative bulb of percussion and the position of the fracture initiation (Supplementary  
163 Fig. 8). MLITs are microscopically observable impact scars on lithic surfaces<sup>7,8,42,43</sup>. They comprise  
164 clusters of linear polishes running parallel to one another, exhibiting long shining stripes. Although  
165 little is known about the process of MLIT formation, they probably formed through contact with  
166 fragments detached from stone tips or the bones of animal targets. Similar linear polishes can occur  
167 through knapping by a hammer (Supplementary Fig. 8f) and contact with other stone artefacts  
168 during transport or storage<sup>37</sup>. However, it is possible to distinguish MLITs from the other linear  
169 polishes on the basis of attributes characterized by long, stripe-like linear polishes running in a  
170 specific direction with other linear polishes. The MLITs were recorded using a Hirox microscope at  
171 magnifications between  $\times 140$  and  $\times 480$ .

172 **Residue analysis.** FTIR analyses were performed at the Chemical and Life Sciences branch of the  
173 SISSI beamline at Elettra Sincrotrone, Trieste<sup>44</sup>. Ten backed pieces were analysed by FTIR  
174 spectromicroscopy (100a from layer D; 106 from spit E-D; 75, 1, 34, 64, 45 and 52 from layer EII-  
175 I; and 21 and 23 from layer EIII). A few grains of the adherent residues were gently scraped from  
176 each backed piece using the tip of a needle under a stereomicroscope. Collected grains from each  
177 sample were pressed in a diamond compression cell (Diamond EX press by S.T. Japan, clear  
178 aperture 2 mm) to flatten them to a thickness suitable for FTIR transmission measurements. Owing  
179 to the heterogeneous nature of the samples, 10–15 spectra for each were acquired in transmission

180 mode on half compression cell with a Vis-IR Bruker Hyperion 3000 microscope coupled with the  
181 Vertex 70v interferometer in the MidIR range (MCT-A detector, 4,000–750  $\text{cm}^{-1}$ ). For each  
182 spectrum, 512 scans were averaged at 4  $\text{cm}^{-1}$  spectral resolution, setting the lateral resolution at 50  
183  $\times$  50  $\mu\text{m}^2$  to select the most diagnostic sample regions according to the observable differences in  
184 colour. Spectra of red deposits from layers E and D and soil samples from several stratigraphic units  
185 belonging to Grotta del Cavallo (see Supplementary Fig. 1) were also measured by FTIR  
186 spectroscopy in the sample compartment of the Vertex 70v interferometer, in the closed diamond  
187 compression cell, using a 5 multiplication focusing unit (A524/Q, Bruker Optics) and the Bruker  
188 wide range components (that is, beamsplitter and DTGS detector) for covering FIR (far-infrared)  
189 and MIR (mid-infrared) spectral regions in a single scan. Each spectrum was collected averaging  
190 256 scans at 4  $\text{cm}^{-1}$ . Extending the spectral range from 4,000 to 150  $\text{cm}^{-1}$  allows better  
191 highlighting of the presence of metal-organic spectral features. To identify a specific material  
192 adhered on lithics, all of the acquired FTIR spectra were compared with those reported in the  
193 literature and IR spectral libraries (Kimmel Center for Archaeological Science Infrared Standards  
194 Library and IRUG Spectral Database). In addition, samples 1 and 106 were peeled off with carbon  
195 conductive adhesive tape from the culet of the diamond after FTIR spectromicroscopy analysis and  
196 SEM/EDX measurements were performed. Two red deposits (one from layer D and one from layer  
197 EII-I) and a sample of soil from layer DII were also characterized from a mineralogical perspective.  
198 All measurements were performed using a Zeiss Supra 40 field emission gun, an SEM equipped  
199 with a Gemini column and an in-lens secondary electron detector operated at 10 kV. EDX analyses  
200 were performed using a LN<sub>2</sub>-free X-Act Silicon Drift Detector (Oxford X-ray detection system,  
201 Aztec EDS). SEM/EDX measurements were performed at the IOM-CNR laboratories. Among the  
202 10 backed pieces analysed by FTIR spectromicroscopy, only 6 (1, 34, 64, 106, 100a and 75)  
203 showed clear infrared features indicative of an organic fraction (see Fig. 2o). The organic fraction  
204 was verified by strong absorption peaks in the range 3,000–2,800  $\text{cm}^{-1}$ , which were assigned to  
205 methyl and methylene asymmetric and symmetric stretching modes at  $\sim$ 2,956 and  $\sim$ 2,872  $\text{cm}^{-1}$ ,  
206 and  $\sim$ 2,930 and  $\sim$ 2,860  $\text{cm}^{-1}$ , respectively<sup>45</sup>. At  $\sim$ 1,460 and  $\sim$ 1,378  $\text{cm}^{-1}$ , the bending modes of  
207 the same moieties can be observed. The aforementioned stretching and bending modes are  
208 characteristic of compounds containing long aliphatic chains. In addition, carbonyl (C = O) bands  
209 can be detected at around 1,740  $\text{cm}^{-1}$  for all the selected six samples, and an extra shoulder centred  
210 at about 1,715  $\text{cm}^{-1}$  can be seen for samples 34, 64, 75 and 100a. Typically, carbonyl stretching  
211 modes of esters and carboxylic acids fall in this spectral region<sup>46</sup>. Samples 75, 106 and 100a (Fig.  
212 2o) are characterized by two broad bands in the 1,650–1,550  $\text{cm}^{-1}$  and 1,450–1,350  $\text{cm}^{-1}$  spectral  
213 regions. The two aforementioned contributions may derive from asymmetric and symmetric  
214 stretching of carboxyl groups usually identified as diagnostic of gum (see the next paragraph for  
215 more details)<sup>47</sup>. These contributions are less intense for samples 1, 34 and 64 (Fig. 2o), allowing the  
216 peak centred at about 1,630  $\text{cm}^{-1}$  to arise. All the aforementioned spectral ranges are indicated by  
217 the grey shaded areas in Fig. 2o. The collected data led to postulations that the organic fraction is a  
218 mixture of two main components: tree or plant gum and beeswax. In particular, the broad peaks in  
219 the 1,650–1,550 and 1,450–1,350  $\text{cm}^{-1}$  spectral regions can be associated with carboxylate  
220 fractions from plant or tree gum, a natural biopolymer composed mostly of diverse polysaccharides  
221 and, to a much lesser extent, glycoproteins<sup>45,48</sup>. This hypothesis was proven by the spectral  
222 comparison of samples 75, 106 and 100a with the reference spectrum of tree gum (Fig. 2o) and  
223 several other spectra found in the IR databases (see, for example, spectra IDs ICB00011, ICB00012,  
224 ICB00013 and ICB00038 in the IRUG database). Pure and fresh gum spectra are characterized by

225 narrower bands in the aforementioned spectral regions. Nevertheless, it is well known that the peak  
226 position of both the asymmetric and symmetric modes of carboxyl groups are strongly dependent on  
227 the coordinated cations<sup>44</sup>; therefore, band broadening in our samples reflects the complex  
228 mineral composition of the soil (see the SEM/EDX analysis and Supplementary Fig. 4 for more  
229 details). Reference gum spectra show broad unresolved absorption peaks in the range 3,000–2,800  
230 cm<sup>-1</sup>, which differ from the signals obtained by measuring our samples that exhibited intense and  
231 sharp methyl and methylene stretching modes. This result led to the deduction of the possible  
232 addition of a further organic compound to the adhesive, such as beeswax. This hypothesis can be  
233 tested by comparison of the collected spectra of samples 1, 34 and 64 with beeswax reference  
234 spectra (Fig. 2o). In the literature, the spectra of beeswax (see also IDs IWX00075, IWX00090,  
235 IWX00096 and IWX00099 in the IRUG database) are characterized by well-defined and intense  
236 methyl and methylene bands, as well as by distinctive carbonyl bands centred at about ~1,740 and  
237 ~1,715 cm<sup>-1</sup>, which were also present in our samples. Among the collected spectra, a variability of  
238 the relative intensity of the methylene/methyl/carbonyl bands can be observed, mainly characteristic  
239 of beeswax (Fig. 2o), with respect to the broad bands extending from about 1,650–1,550 cm<sup>-1</sup> and  
240 1,450–1,350 cm<sup>-1</sup>, which are characteristic of tree/plant gum (Fig. 2o). This finding can be  
241 explained by the different percentages of the two organic fractions used to prepare the adhesive  
242 mixture, with further consideration of the different degrees of degradation and aging originating  
243 from long-term interaction of the organic material constituting the adhesives with the burial soil<sup>46</sup>.  
244 The diverse extent of degradation of the samples could have been influenced by differences in soil  
245 composition, pH, humidity or water percolation of the stratigraphic units where the ten backed  
246 pieces were buried for thousands of years. Identification of the gum fraction would have been easier  
247 with access to the ~1,200–900 cm<sup>-1</sup> spectral region, where C–O–C and C–OH stretching modes  
248 diagnostic of polysaccharides are located<sup>46</sup>. In this spectral region, very intense and structured  
249 bands can be seen for all 10 measured backed pieces. This feature, characterized by a main peak at  
250 1,030 cm<sup>-1</sup>, a shoulder at 1,080 cm<sup>-1</sup> and two distinctive peaks at 800 and 780 cm<sup>-1</sup>, can be  
251 attributed to Si–O stretching modes of silicates, which are the main components of clays.  
252 Specifically, the sharp peaks at 3,694 and 3,622 cm<sup>-1</sup> are distinctive vibrational features of well-  
253 crystallized water molecules among the layers of kaolinite<sup>47</sup>.  
254 The red colour of the residues on the backed pieces led us to hypothesize the presence of iron  
255 compounds. To verify this hypothesis, SEM/EDX analyses were performed for a soil sample from  
256 layer DII and samples 106 (from spit E-D) and 1 (from layer EII-I) after FTIR analysis  
257 (Supplementary Fig. 4b,e,h).  
258 EDX measurements of the soil and sample 106 confirmed the presence of silicon, aluminium,  
259 magnesium, sodium, calcium, iron and phosphorus, which are all characteristic of silicates. The  
260 iron-to-silicon ratio increased from  $0.37 \pm 0.01$  to  $4.52 \pm 2.01$  from the soil to sample 106, reaching  
261 a value of  $7.64 \pm 0.45$  in sample 1 (the standard deviation was calculated as the average of three  
262 measurements per sample). The positive trend of the iron-to-silicon ratio from the soil to sample 1  
263 is consistent with a colour transition from light brown to intense red (Supplementary Fig. 4a,d,g),  
264 revealing that the iron content of the samples is much higher than the one of the burial soil and that  
265 it contributes to red pigmentation of the residues on samples 1 and 106, which can be identified as  
266 ochre.  
267 To further verify that ochre (also known as red earth) is the source of the red colour, some red soil  
268 deposits collected from Grotta del Cavallo were analysed by FTIR spectroscopy in the FIR-MIR  
269 region. These deposits belong to the same stratigraphic units (layers E and D) as the analysed



270 backed pieces (see Supplementary Fig. 1). In Supplementary Fig. 5, we report the FIR-MIR spectra  
271 of two of the analysed red deposits. It is possible to identify peaks centred at about 535 and 433  
272  $\text{cm}^{-1}$ , as well as a broad band around 325  $\text{cm}^{-1}$ , that are distinctive of iron oxides. The collected  
273 spectra can be correlated with the IRUG ochre spectrum IMP00365 (red earth made by kaolinite  
274 and hematite). Supplementary Fig. 5 also reports the FIR-MIR spectrum of the soil sample from  
275 layer DII, also analysed by SEM/EDX (Supplementary Fig. 4). This sample does not show the  
276 spectral features characteristic of ochre, accordingly with the minimal iron content revealed by  
277 SEM/EDX analysis; instead, it is mainly characterized by a mixture of silicates and phosphates. As  
278 a matter of fact, the silicate peaks described above can also be recognized in the FTIR spectrum of  
279 the soil, and distinctive features of phosphates can be also identified: two sharp peaks at  $\sim 964$  and  
280  $\sim 870$   $\text{cm}^{-1}$ , a double peak at  $\sim 605$  and  $\sim 564$   $\text{cm}^{-1}$  and a moderate absorption band in the 1,550–  
281 1,300  $\text{cm}^{-1}$  spectral range<sup>49</sup>. The aforementioned phosphate infrared features are still evident in  
282 the spectrum of the red deposit from layer D, whereas they are barely detectable for the red deposit  
283 from layer E II-I. This result implies that the red deposit from layer D is partially contaminated by  
284 the burial soil, while the one from layer E II-I can be considered as a purer ochre. None of the  
285 spectra reported in Supplementary Fig. 5 show absorbance peaks in the region 3,000–2,800  $\text{cm}^{-1}$ ,  
286 which are characteristic of aliphatic chains of organic compounds. This result suggests that, in both  
287 the soil and red deposits, the organic matter content is below the detection limit of the technique,  
288 thereby excluding the possibility that the organic traces on backed pieces are contamination from  
289 the burial environment. Taken together, these results led us to conclude that the residue stuck on the  
290 backed pieces is a mixture of plant/tree gum and beeswax intentionally mixed with ochre and  
291 applied as an adhesive.

292 **Morphometric analysis.** As the Uluzzian backed pieces are extremely small (Supplementary Figs.  
293 6a and 7b), they are not suitable to haft onto the tips of thick wooden spears from Schöningen in  
294 Germany dated  $\sim 300$  ka<sup>50–52</sup>, which were probably used as throwing spears<sup>53,54</sup> (Supplementary Fig.  
295 7a). It has been ethnographically shown that thrusting spears and hand-delivered spears are heavier  
296 than projectile spears launched with a spearthrower or bow<sup>55,56</sup>. Therefore, the Uluzzian backed  
297 pieces do not function well as throwing or thrusting spear tips, which require a massive shaft. If the  
298 Uluzzian backed pieces were inserted into the lateral sides of a shaft as in Magdalenian composite  
299 projectiles<sup>57</sup>, the smallness of the stone artefacts would not necessarily relate to the diameter of the  
300 shaft. However, as the use-wear analysis suggested that a considerable number of Uluzzian pieces  
301 were attached to the tip of a shaft as a hunting armature, the small dimensions must reflect a thin  
302 shaft that is useful only for mechanically delivered spears, such as darts projected by a spearthrower  
303 or arrows shot using a bow. A morphometric analysis using TCSA and TCSP values was therefore  
304 undertaken to evaluate the potential projectile capability of stone tips<sup>20,56,58,59</sup>.

305 TCSA and TCSP values of Uluzzian backed pieces from Grotta del Cavallo were compared with  
306 those of ethnographic North American dart tips and arrowheads<sup>12,13</sup>. Because some Uluzzian backed  
307 pieces were used for cutting and scraping, the TCSA and TCSP analyses were undertaken only for  
308 the backed pieces showing DIFs (Supplementary Fig. 6b,c). The TCSA and TCSP values were  
309 calculated using the equations presented by Sisk and Shea<sup>59</sup>.

310  
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323

## 324 **Author Contributions**

325 A.M. and K.S. conceived and organized the project; S.B. obtained funding and directed the project;  
326 K.S. undertook the use-wear analysis with S.A. as well as the morphometric analysis; C.S., G.B.,  
327 and L.V. performed the residue analysis; D.A. conducted the experiment for producing Uluzzian  
328 backed pieces; I.F., M.G., and A.T. provided data about the exploitation of feathers; F.B., J.C., and  
329 P.B. presented the results of the zooarchaeological analysis; K.S., C.S., V.S., S.R., and I.F. made  
330 figures and illustrations; D.A., F.B., A.R., and A.M. provided permits for the analysis of the  
331 archaeological samples and expertise on site sequences and materials; and K.S., C.S., A.R., A.M.,  
332 and S.B. wrote the manuscript with contributions from all co-authors.

333

334 **Competing interests** The authors declare no competing interests.

335

336 **Correspondence and requests for materials and data** should be addressed to K.S.

337

## 338 *Captions*

339 **Fig. 1** | Locations of the Uluzzian findings in Italy and on the Balkan Peninsula. 1, Klissoura Cave;  
340 2, Kephalaria Cave; 3, Crvena Stijena; 4, Grotta del Cavallo; 5, Grotta di Serra Cicora A; 6, Grotta  
341 Mario Bernardini; 7, Grotta di Uluzzo; 8, Grotta di Uluzzo C/Cosma; 9, Grotta delle Veneri; 10,  
342 Grotta di Castelcivita; 11, Grotta della Cala; 12, Colle Rotondo; 13, Grotta La Fabbrica; 14, Riparo  
343 del Broion; 15, Grotta di Fumane. Sea level is 74 m below the presentdaycoastline (data from ref.  
344 60). The digital elevation model is the European digital elevation model from the GMES RDA  
345 project ([https://www.eea.europa.eu/data-and-maps/data/eu-dem#tab-](https://www.eea.europa.eu/data-and-maps/data/eu-dem#tab-originaldata/eudem_hlsd_3035_europe)  
346 [originaldata/eudem\\_hlsd\\_3035\\_europe](https://www.eea.europa.eu/data-and-maps/data/eu-dem#tab-originaldata/eudem_hlsd_3035_europe)). The bathymetric model is from the European Marine  
347 Observation and Data Network. The map was generated using ArcGIS version 10.5.

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349 **Fig. 2** | Backed pieces from Grotta del Cavallo showing DIFs and MLITs, and sampling of residues  
350 on backed pieces by FTIR spectroscopy and its results. **a**, A simple DIF type a2. **b–f**, Multiple DIF  
351 type a2m. **a(i)**, **c(ii)** and **d(i)** are burin-like fractures; **b(i)**, **c(i)** and **c(iii)** are flute-like fractures; **b(ii)**  
352 is a step-terminating transverse fracture and a spin-off; **e(i)** and **d(ii)** are spin-offs; **e(ii)** is a step-  
353 terminating transverse fracture; **f(ii)** is flute- and burin-like fractures; **f(iii)** is a feather-terminating  
354 transverse fracture. **a(ii)**, **f(i)** and the black lines in **a** and **f** are MLITs. **b**, **c** and **e** are from layer EII-  
355 I; **a** and **d** are from layer E-D; and **f** is from layer D. **g,k**, Optical images at two different angles of  
356 sample 1, layer EII-I (scale bar, 5 mm) and sample 106, spit E-D (scale bar, 5 mm). Sampled areas

357 are highlighted by a black box and magnified in **h** and **i** for sample 1 (scale bars, 1 mm and 0.5 mm)  
358 and in **l** and **m** for sample 106 (scale bars, 2 mm and 1 mm). **j,n**, Optical images of the scraped  
359 residues sitting on the culet of the opened diamond compression cell. **o**, Representative FTIR  
360 spectra of the sampled residues from samples 1, 34, 64, 75, 106 and 100a. Two selected reference  
361 spectra of beeswax and peach tree gum are also plotted using the database from the Kimmel Center  
362 for Archaeological Science Infrared Standards Library ([https://www.weizmann.ac.il/kimmel-arch/  
363 infrared-spectra-library](https://www.weizmann.ac.il/kimmel-arch/infrared-spectra-library)). The grey shaded areas indicate the main absorption bands, characteristic  
364 of the organic fraction. Among them, those relating to beeswax are marked with dagger symbols,  
365 and those relating to plant/tree gum are marked with section symbols. For more details on the band  
366 positions and assignments, refer to the Methods.

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## 369 References

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- 371 1. Moroni, A. et al. *J. Anthr. Sci.* **96**, 125–160 (2018).
- 372 2. Zanchetta, G., Giaccio, B., Bini, M. & Sarti, L. *Quat. Sci. Rev.* **182**, 65–77 (2018).
- 373 3. Giaccio, B., Hajdas, I., Isaia, R., Deino, A. & Nomade, S. *Sci. Rep.* **7**, 45940 (2017).
- 374 4. d’Errico, F., Borgia, V. & Ronchitelli, A. *Quat. Int.* **259**, 59–71 (2012).
- 375 5. Moroni, A., Boscato, P. & Ronchitelli, A. *Quat. Int.* **316**, 27–44 (2013).
- 376 6. Palma di Cesnola, A. *Riv. Sci. Preist.* **21**, 3–59 (1966).
- 377 7. Fischer, A., Hansen, P. V. & Rasmussen, P. *J. Dan. Archaeol.* **3**, 19–46 (1984).
- 378 8. Sano, K. & Oba, M. *J. Archaeol. Sci.* **63**, 13–23 (2015).
- 379 9. Bradtmöller, M., Sarmiento, A., Perales, U. & Zuluaga, M. C. *J. Archaeol. Sci. Rep.* **7**, 1–13  
380 (2016).
- 381 10. Yaroshevich, A., Kaufman, D., Nuzhnyy, D., Bar-Yosef, O. & Weinstein-Evron, M. *J.*  
382 *Archaeol. Sci.* **37**, 368–388 (2010).
- 383 11. Goldstein, S. T. & Shaffer, C. M. *Archaeol. Anthr. Sci.* **9**, 1767–1788 (2017).
- 384 12. Thomas, D. H. *Am. Antiq.* **43**, 461–472 (1978).
- 385 13. Shott, M. J. *Am. Antiq.* **62**, 86–101 (1997).
- 386 14. Wadley, L. *J. Hum. Evol.* **49**, 587–601 (2005).
- 387 15. Cattelain, P. *Bull. Soc. Préhist. Fr.* **86**, 213–216 (1989).
- 388 16. Rust, A. *Die alt- und Mittelsteinzeitlichen Funde von Stellmoor* (Karl Wachholtz, 1943).
- 389 17. Stodiek, U. *Zur Technologie der Jungpaläolithischen Speerschleuder: Eine Studie auf der Basis*  
390 *Archäologischer, Ethnologischer und Experimenteller Erkenntnis* (Archaeologica Venatoria,  
391 Institut für Ur- und Frühgeschichte der Universität Tübingen, 1993).
- 392 18. Thieme, H. & Veil, S. *Ldkr. Verden Kunde* **36**, 11–58 (1985).
- 393 19. Villa, P., Boscato, P., Ranaldo, F. & Ronchitelli, A. *J. Archaeol. Sci.* **36**, 850–859 (2009).
- 394 20. Shea, J. J. *J. Archaeol. Sci.* **33**, 823–846 (2006).
- 395 21. Metz, L. in *Le Troisième Homme Préhistoire de l’Altai* 156–160 (Musée National de la  
396 Préhistoire, 2017).
- 397 22. Slimak, L. *Quat. Sci. Rev.* **217**, 330–339 (2019).
- 398 23. Villa, P. & Soriano, S. *J. Anthropol. Res.* **66**, 5–38 (2010).
- 399 24. Wadley, L. & Mohapi, M. *J. Archaeol. Sci.* **35**, 2594–2605 (2008).
- 400 25. Zilhão, J., Banks, W. E., d’Errico, F. & Gioia, P. *PLoS ONE* **10**, e0131181 (2015).
- 401 26. Benazzi, S. et al. *Nature* **479**, 525–528 (2011).

- 402 27. Heptner, V. G., Nasimovich, A. A. & Bannikov, A. G. *Mammals of the Soviet Union* Vol. 1  
403 (Smithsonian Institution Libraries, National Science Foundation, 1988).
- 404 28. Lew-Levy, S., Reckin, R., Lavi, N., Cristóbal-Azkarate, J. & Ellis-Davies, K. *Hum. Nat.* **28**,  
405 367–394 (2017).
- 406 29. Milks, A., Parker, D. & Pope, M. *Sci. Rep.* **9**, 820 (2019).
- 407 30. Tringham, R., Cooper, G., Odell, G., Voytek, B. & Whitman, A. *J. Field Archaeol.* **1**, 171–196  
408 (1974).
- 409 31. Odell, G. H. & Odell-Vereecken, F. *J. Field Archaeol.* **7**, 87–120 (1980).
- 410 32. Odell, G. H. *J. Field Archaeol.* **8**, 197–209 (1981).
- 411 33. Akoshima, K. in *The Human Uses of Flint and Chert: Proc. Fourth International Flint*  
412 *Symposium Held at Brighton Polytechnic 10-1 April 1983* (eds Sieveking, G. D. & Newcomer, M.  
413 H.) 71–79 (Cambridge Univ. Press, 1987).
- 414 34. Keeley, L. H. *Experimental Determination of Stone Tool Uses: A Microwear Analysis* (Univ.  
415 Chicago Press, 1980).
- 416 35. Vaughan, P. C. *Use-Wear Analysis of Flaked Stone Tools* (Univ. Arizona Press, 1985).
- 417 36. Van Gijn, A. L. *The Wear and Tear of Flint: Principles of Functional Analysis Applied to Dutch*  
418 *Neolithic Assemblages* (Univ. Leiden, 1990).
- 419 37. Sano, K. *Functional Variability in the Late Upper Palaeolithic of North-Western Europe*  
420 (Rudolf Habelt, 2012).
- 421 38. Lombard, M. *J. Archaeol. Sci.* **35**, 26–41 (2008).
- 422 39. Pargeter, J. *J. Archaeol. Sci.* **40**, 4056–4065 (2014).
- 423 40. Sano, K. *Quartär* **56**, 67–86 (2009).
- 424 41. Pargeter, J. *J. Archaeol. Sci.* **38**, 2882–2888 (2011).
- 425 42. Moss, E. H. & Newcomer, M. H. *Stud. Praehist. Belg.* **2**, 289–312 (1982).
- 426 43. Geneste, J. M. & Plisson, H. in *Before Lascaux: The Complex Record of the Early Upper*  
427 *Paleolithic* (eds Knecht, H. et al.) 117–135 (CRC, 1993).
- 428 44. Lupi, S. et al. *J. Opt. Soc. Am. B* **24**, 959–964 (2007).
- 429 45. Petrea, P., Amarioarei, G., Apostolescu, N., Puitel, A. C. & Ciovica, S. *Cell. Chem. Technol.* **47**,  
430 369–375 (2013).
- 431 46. Socrates, G. in *Infrared and Raman Characteristic Group Frequencies Tables and Charts* (ed.  
432 Socrates, G.) 283–340 (Wiley, 2001).
- 433 47. van der Marel, H. W. & Krohmer, P. *Contr. Mineral. Petrol.* **22**, 73–82 (1969).
- 434 48. Espinosa-Andrews, H., Sandoval-Castilla, O., Vazquez-Torres, H., Vernon-Carter, E. J. &  
435 Lobato-Calleros, C. *Carbohydr. Polym.* **79**, 541–546 (2010).
- 436 49. Fleet, M. E. *Biomaterials* **30**, 1473–1481 (2009).
- 437 50. Sierralta, M., Frechen, M. & Urban, B. in *Die Chronologische Einordnung der Paläolithischen*  
438 *Fundstellen von Shöningen: Forschungen zur Urgeschichte aus dem Tagebau von Shöningen Band*  
439 *1* (ed. Behre, K.-E.) 143–154 (Römisch-Germanischen Zentralmuseums, 2012).
- 440 51. Urban, B. & Sierralta, M. in *Die Chronologische Einordnung der Paläolithischen Fundstellen*  
441 *von Shöningen: Forschungen zur Urgeschichte aus dem Tagebau von Shöningen Band 1* (ed.  
442 Behre, K.-E.) 77–96 (Römisch-Germanischen Zentralmuseums, 2012).
- 443 52. Richter, D. & Krbetschek, M. *J. Hum. Evol.* **89**, 46–56 (2015).
- 444 53. Thieme, H. *Nature* **385**, 807–810 (1997).

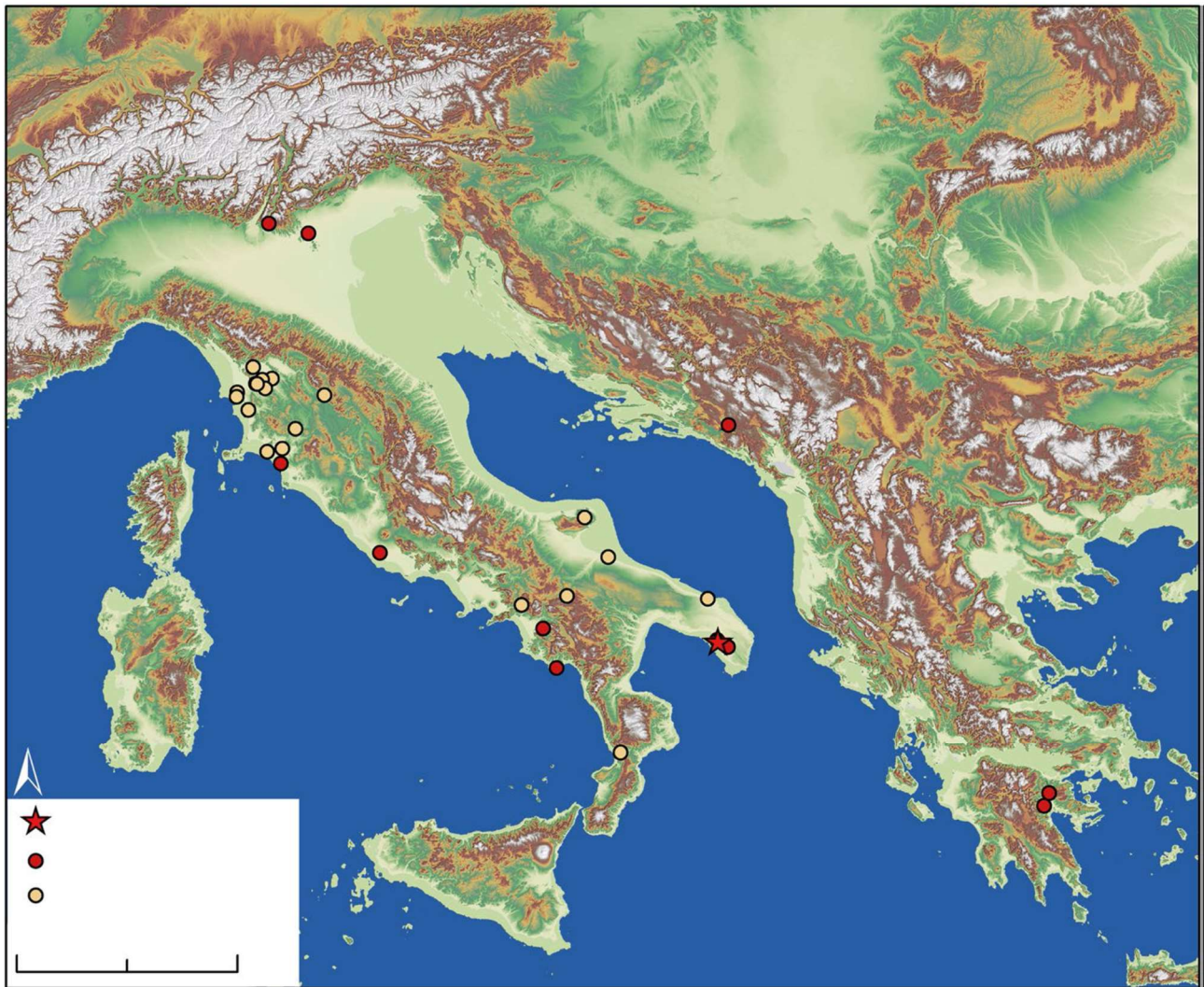
- 445 54. Thieme, H. in *The Hominid Individual in Context: Archaeological Investigations of Lower and*  
446 *Middle Palaeolithic Landscapes, Locales and Artefacts* (eds Gamble, C. & Porr, M.) 115–132  
447 (Routledge, 2005).
- 448 55. Cattelain, P. in *Projectile Technology* (ed. Knecht, H.) 213–240 (Plenum, 1997).
- 449 56. Hughes, S. S. *J. Archaeol. Method Theory* **5**, 345–408 (1998).
- 450 57. Pétilion, J.-M. et al. *J. Archaeol. Sci.* **38**, 1266–1283 (2011).
- 451 58. Shea, J. J. & Sisk, M. L. *Paleoanthropology* **2010**, 100–122 (2010).
- 452 59. Sisk, M. L. & Shea, J. J. *Int. J. Evolut. Biol.* **2011**, 1–8 (2011).
- 453 60. Waelbroeck, C. et al. *Quat. Sci. Rev.* **21**, 295–305 (2002).

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491 **Fig. 1**

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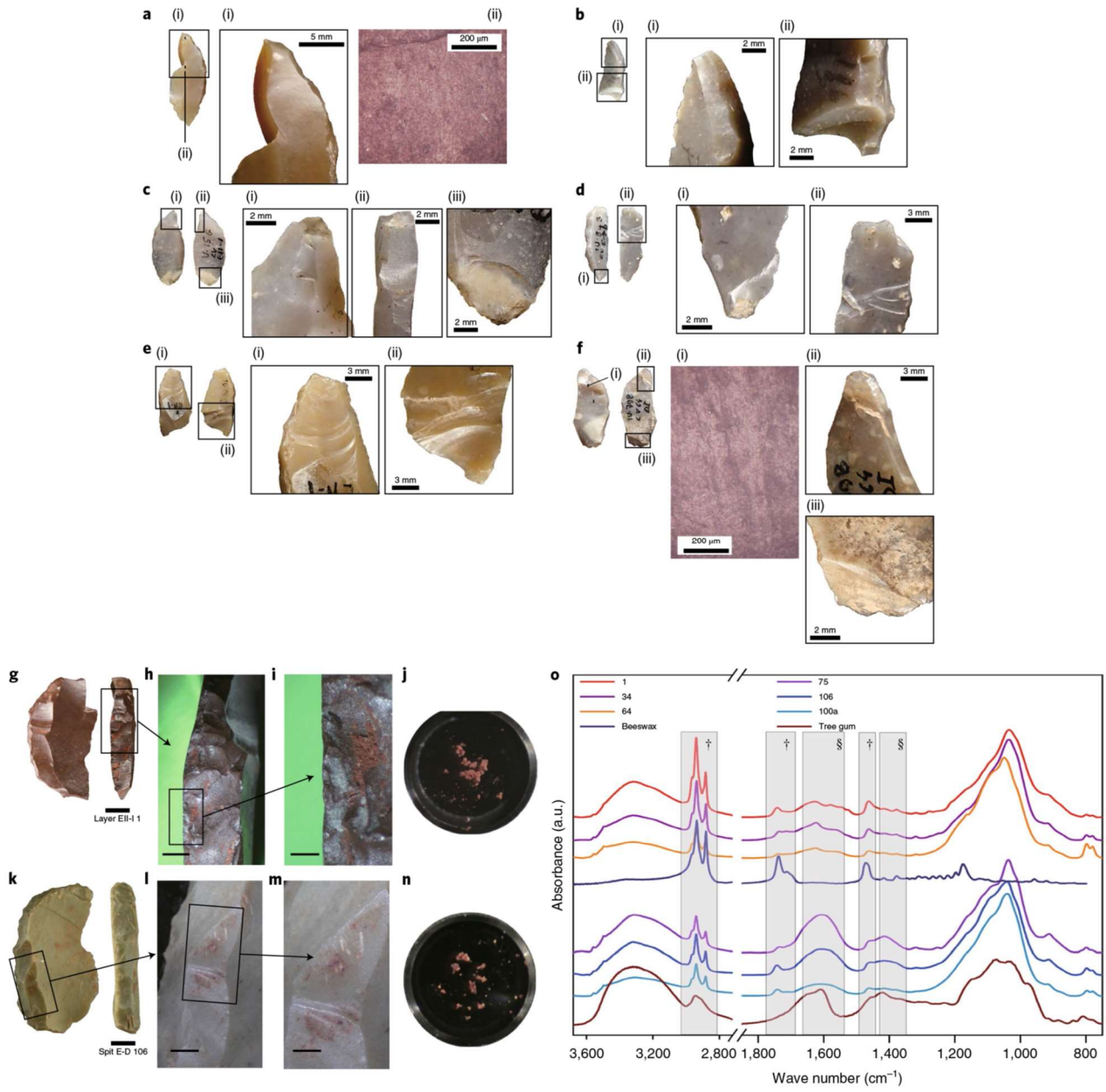
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**Fig. 2**



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