

FIRST LARGE-SCALE PROVENANCE STUDY OF PIGMENTS REVEALS NEW COMPLEX BEHAVIOURAL PATTERNS DURING THE UPPER PALAEOLITHIC OF SOUTH-WESTERN GERMANY*

E. C. VELLIKY† 

Institut für Naturwissenschaftliche Archäologie, Mathematisch-Naturwissenschaftlichen Fakultät, University of Tübingen, Tübingen, Germany and Archaeology/Centre for Rock-Art Research and Management, M257, Faculty of Arts, Business, Law and Education, School of Social Sciences, The University of Western Australia, Crawley, WA, Australia and SFF Centre for Early Sapiens Behaviour (SapienCE), University of Bergen, Bergen, Norway

B. L. MACDONALD 

Archaeometry Laboratory, University of Missouri Research Reactor (MURR), Columbia, MO 65211, USA

M. PORR 

Archaeology/Centre for Rock-Art Research and Management, M257, Faculty of Arts, Business, Law and Education, School of Social Sciences, The University of Western Australia, Crawley, WA, Australia and Institut für Ur- und Frühgeschichte und Archäologie des Mittelalters, ROCEEH—The Role of Culture in Early Expansions of Humans, University of Tübingen, Tübingen, Germany

N. J. CONARD

Institut für Naturwissenschaftliche Archäologie, Mathematisch-Naturwissenschaftlichen Fakultät, University of Tübingen, Tübingen, Germany and Department of Early Prehistory and Quaternary Ecology & Senckenberg Centre for Human Evolution and Quaternary Ecology, University of Tübingen, Tübingen, Germany

The use of red iron-based earth pigments, or ochre, is a key component of early symbolic behaviours for anatomically modern humans and possibly Neanderthals. We present the first ochre provenance study in Central Europe showing long-term selection strategies by inhabitants of cave sites in south-western Germany during the Upper Palaeolithic (43–14.5ka). Ochre artefacts from Hohle Fels, Geißenklösterle and Vogelherd, and local and extra-local sources, were investigated using neutron activation analysis (NAA), X-ray diffraction (XRD) and scanning electron microscopy (SEM). The results show that local ochre sources were continuously and systematically accessed for c.29500years, with periodic events of long-distance (about > 300km) ochre acquisition during the Aurignacian (c.35–43ka), suggesting higher mobility than previously suspected. The results reveal previously unknown long-term, complex spatio-temporal behavioural patterns during the earliest presence of Homo sapiens in Europe.

KEYWORDS: MINERAL PIGMENTS, OCHRE PROVENANCE, GEOCHEMISTRY, EUROPEAN UPPER PALAEOLITHIC, SYMBOLIC BEHAVIOUR

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†Corresponding author: email elizabeth.velliky@uib.no

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INTRODUCTION

The use of mineral pigments by hominins is generally accepted as an important element in the early material expressions of complex cognitive thought, syntactical language and mediation of symbolic communication (McBrearty and Brooks 2000; D’Errico 2003; Henshilwood and Marean 2003; Watts 2009). A common mineral pigment used by hominins is a material colloquially referred to as ochre, which encompasses a range of materials containing mineral phases of Fe oxides/oxyhydroxides. The habitual use of ochre emerged as early as 300–500 ka in Sub-Saharan Africa (Watts *et al.* 2016; Brooks *et al.* 2018), 90 ka in the Levant (Hovers *et al.* 2003; Salomon *et al.* 2012), and roughly 45–50 ka in Europe (Salomon *et al.* 2008; Zilhão *et al.* 2010; Bodu *et al.* 2014), although some older European examples (*c.*100–200 ka) exist (Zilhão *et al.* 2010; Roebroeks *et al.* 2012; Hoffmann *et al.* 2018). Though these reports present compelling evidence for the collection and manipulation of ochre pigments, the conceptualization of these processes and the identification of symbolic items remains a highly contested field (Wadley 2003; D’Errico and Henshilwood 2011; Mithen 2014). Much debate exists about whether ochre was used primarily in symbolic or functional contexts, as some have reported ochre-based residues on lithics (Lombard 2006; Wojcieszak and Wadley 2018), and recent experiments have shown ochre to be a useful hafting mastic (Wadley *et al.* 2004; Wadley 2005; Lombard 2006), insect repellent (Rifkin 2015a), sunscreen (Rifkin *et al.* 2015), hide-tanning ingredient (Audouin and Plisson 1982; Rifkin 2011), in various compound mixtures (Henshilwood *et al.* 2011; Villa *et al.* 2015) and for medicinal purposes (Velo 1984).

With the onset of the Upper Palaeolithic (UP) in Europe and the migration of anatomically modern humans (AMHs) into the continent, several forms of material culture, including painted and engraved cave art (Clottes 2008; White *et al.* 2017), personal ornaments (White 1995; Vanhaeren and D’Errico 2006), and complex lithic technology (Bataille and Conard 2018; Dinnis *et al.* 2019), suggest forms of complex and well-established symbolically mediated behaviours. The use of ochre and pigments is included in this palimpsest, yet much reporting focuses on Western European contexts. Though some previous research explored geochemical aspects of ochre use (Sajó *et al.* 2015), long-term diachronic developments and social and environmental interplay between AMHs and ochre use in Central Europe remains comparatively underdeveloped. This is perhaps due to the lack of painted cave art in this region, and a prevalence of portable art where pigments were not used, have not survived or have not been the focus of intensive analyses.

The Swabian Jura region in south-western Germany is of crucial importance for exploring early behavioural complexity in Europe. Cave sites here (Fig. 1) offer early examples of human occupation in the continent, dating to *c.*43 ka (Conard 2003; Conard and Bolus 2006, 2008), and document the Middle to UP transition (Conard and Bolus 2008; Conard 2011). Two tributary valleys of the Danube, the Ach and Lone valleys, contain several of such sites. Hohle Fels (HF) is well known for its assortment of carved figurines (Conard 2009), personal ornamentation (Wolf 2015), musical instruments (Conard *et al.* 2009) and lithic technology (Conard and Bolus 2006). A recent assessment of the HF ochre assemblage revealed temporal changes in the types of ochre that were collected and how they were used (Velliky *et al.* 2018). The neighbouring cave sites of Geißenklösterle (GK) and Vogelherd (VH) also yielded ochre artefacts and examples of figurative art (Hahn 1988; Conard 2003; Wolf 2015; Dutkiewicz *et al.* 2018). Additionally, several Fe oxide-bearing outcrops lie near HF and in surrounding regions, which would have been accessible during the late Pleistocene. However, what are the temporal relationships and patterns regarding the qualitative ochre data and the

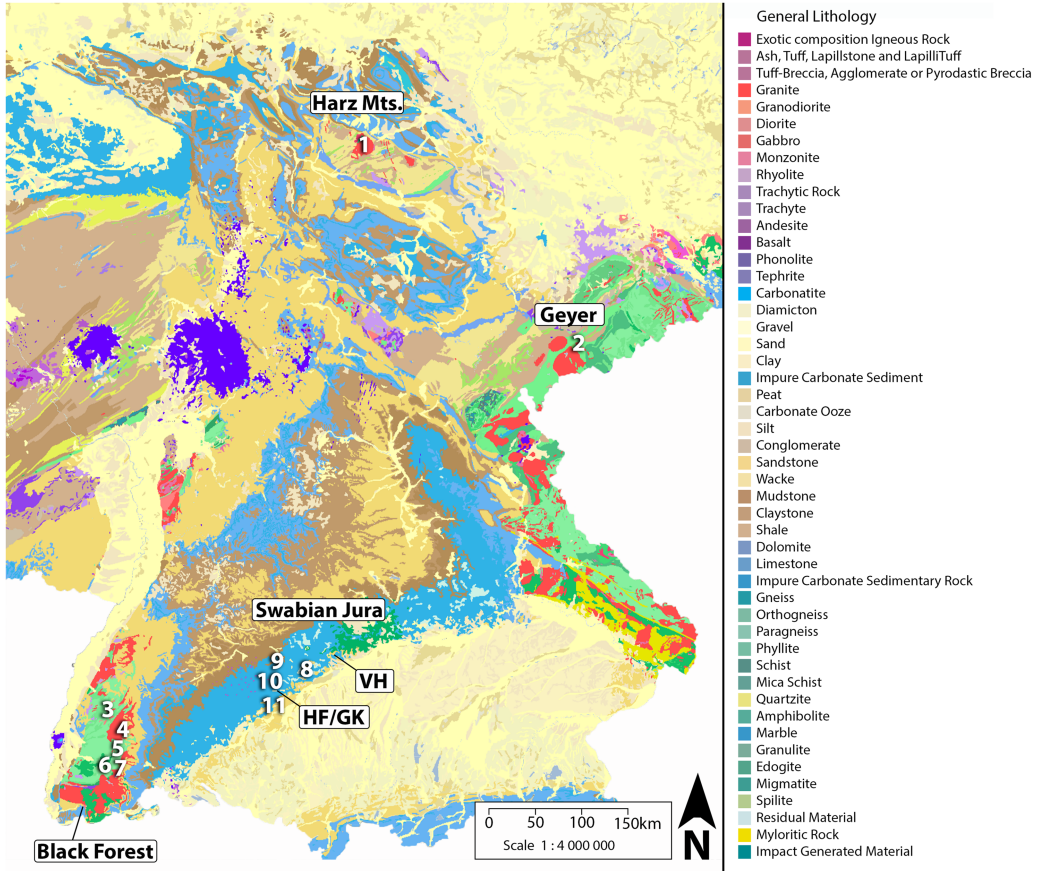


FIGURE 1 Map of the study area, general lithology and three cave sites: Hohle Fels (HF), Geißenklösterle (GK) and Vogelherd (VH). Numbers correspond to ochre outcrops: (1) Harz, (2) Geyer-Erzgebirge, (3) Hechtsberg, (4) Nussbach, (5) Schollach, (6) Rappenloch, (7) Zindelstein, (8) Tormerdingen, (9) Radelstetten, (10) Bohnerz, Bohn-B, Herz-Jesu Berg, Gerhausen and Schelklingen and (11) Allmendingen, Altheim, Ringingen and Kirchbierlingen. Map generated using data from Landesamt für Bergbau, Energie und Geologie (LBEG), Geozentrum Hannover, Niedersachsen. [Colour figure can be viewed at wileyonlinelibrary.com]

presence of the outcrops in the region, and how does this inform us about human and ochre interactions during the UP?

Here we present a provenance study on UP ochre assemblages in Central Europe. We investigate the compositional variation of ochre artefacts from HF and compare them with a database of previously characterized ochre outcrops in Germany (Velliky *et al.* 2019), including local (Swabian Jura, < 80km), extra-local (Black Forest, 80–300km) and distant (Thuringia and Saxony, > 300km) sources, with the goal to test whether the archaeological ochres were gathered from these formations. We use a combination of qualitative data (Velliky *et al.* 2018), trace element geochemistry (neutron activation analysis—NAA), scanning electron microscopy (SEM) and X-ray diffraction (XRD) to make our provenance interpretations. We interpret how the exploitation of different ochre outcrops changed over time at HF based on a combination of qualitative and quantitative data, and apply these data to assemblages from GK and VH to explore regional patterns between the cave sites. The results are compared with environmental data from the UP

(Barbieri *et al.* 2018), which we use to infer landscape knowledge and adaptations to changing environmental and climatic conditions. The data are also compared with previously published behavioural patterns related to lithic (Hahn 1987; Burkert and Floss 1999; Scheer 2000; Floss and Kieselbach 2004) and osseous (Münzel 2001; Barth *et al.* 2009) resources to investigate possible relationships in resource-gathering strategies. Lastly, we identify how changes in the material-acquisition strategies and patterns in ochre behaviour changed or persisted through time.

Introduction to Hohle Fels, Geißenklösterle and Vogelherd

HF is an Upper Jurassic limestone cave situated in the eastern extension of the Swabian Jura, located in the Ach Valley, a tributary of the Danube (Fig. 1). This region of the Swabian Jura is known for its caves with abundant Middle and UP sites. HF is the largest cave with a surface area of 500 m². The site has been of interest since the late 19th century (Fraas 1872) and has yielded one of the deepest, most intact archaeological sequences in the region, spanning from the Middle Palaeolithic (> 44 ka) to the Magdalenian (16.5–14.5 ka) (Conard 2011). The sediments are composed of calcareous clay and locally phosphatic clay, with infrequent inclusions of quartz, phosphatic grains and organic material. Varying amounts of bone, lithic and charcoal fragments are intermixed in the sediments throughout the sequence (Miller 2015).

Red and yellow ochres have been documented at HF since its resurgent excavations during the 1970s by Hahn (Hahn 1977; Blumentritt and Hahn 1978). Numerous artefacts painted or stained with red residues are also reported, including marine and freshwater shells and faunal elements with discreet traces of ochre and limestones bearing distinct geometric patterns of red pigment (Wolf *et al.* 2018). Ochre artefacts with traces of anthropogenic modification (e.g., grinding striations, engravings) and pigment grinding stones attest to its use for various purposes at the site. A re-evaluation of the HF assemblage uncovered a total of 869 ochre artefacts, including 27 anthropogenically modified pieces and 21 suspected modified pieces (Velliky *et al.* 2018).

Ochre is also reported from both GK and VH, though to date there are no systematic studies on the assemblages. Gollnisch (1988) describes some 124 pieces of hematite and other mineral pigments from GK, with total masses of 77 g in the Aurignacian (*c.*35–43 ka) and 138.45 g in the Gravettian (*c.*30.5–34 ka) sequences. In addition, a so-called ‘ochre layer’, or *Rötelschicht*, was reported in the Aurignacian horizon IIIa (Hahn 1988). The recent excavations at VH are of the back-dirt from the original excavations in the 1930s by Riek (1934); therefore, stratigraphic integrity is not assured. Even so, 129 ochre artefacts are recorded from the site, which correspond to Aurignacian and Magdalenian (*c.*14.5–16.5 ka) deposits based on the original chronostratigraphy (Riek 1934) as well as artefact typology (Conard and Bolus 2006). Fig. S1 in the additional supporting information shows examples of ochre artefacts from the three cave sites; Fig. S2 also shows the stratigraphic context from HF and GK.

MATERIALS AND METHODS

Ochre artefacts were selected for geochemical analysis from archived excavation collections housed at the University of Tübingen. All artefacts were macroscopically examined, photographed and catalogued. A detailed summary of the HF ochre assemblage typology is presented by Velliky *et al.* (2018). The descriptive variables used for GK and VH are consistent with those used for the HF ochres (see Table S1). In total, 183 pieces were selected from HF, 18 from VH and nine from GK. Artefacts from HF were chosen based on size, typology and with regard for even sampling distribution amongst the UP cultural periods (Table 1). From HF, 62 pieces

from the Magdalenian, 61 from the Gravettian and 60 from the Aurignacian were chosen. Systematic assessments have not yet been conducted for the GK and VH ochre assemblages, and sample selection was limited by the low numbers of available identified material. Their inclusion in this study serves as a preliminary comparison with the HF ochres. All ochre artefacts were analysed and compared with a geochemical database of 20 source outcrops reported by Velliky *et al.* (2019).

Neutron activation analysis, X-ray diffraction and scanning electron microscopy

NAA was conducted at the Archaeometry Laboratory at the University of Missouri Research Reactor (MURR) using standard procedures described elsewhere (Velliky *et al.* 2019). In ochre provenance studies, it is common to apply multiple transformations to elemental concentration data before statistical testing, including Fe normalization and \log_{10} (Popelka-Filcoff *et al.* 2007; Macdonald *et al.* 2011, 2013, 2018). \log_{10} transformation compensates for the variation in magnitude between major and trace elements, and is necessary for scale-dependent, multivariate statistics (e.g., principal component analysis—PCA or canonical discriminant analysis—CDA). Moreover, because the Fe content can vary significantly, which can artificially amplify or dilute the presence of other trace elements, it is often advantageous to normalize all elemental concentrations to the Fe concentration (Fe normalization). However, it is important for the data transformations to be assessed for their efficacy. In our routine statistical exploration, including iterative bivariate plotting (element concentrations, \log_{10} Fe-normalized ratios), PCA and CDA, we consistently found that using Fe-normalized, \log_{10} values produced the clearest visual separation of compositional groups.

Subsamples of selected ochre powders were submitted for X-ray diffraction (XRD) at the Department of Chemistry, University of Missouri. The XRD patterns were collected using a Scintag X2 powder diffractometer equipped with a Peltier-cooled energy-sensitive detector operating at 40kV and 50mA, using Cu-K α radiation (1.54060 Å). A monochromatic X-ray beam was oriented at each target sample and scanned from 5 to 80° 2 Θ at a step size of 0.02°, and dwell time of 2.0s. The peak patterns were matched with crystallography reference libraries using

Table 1 *Temporal distribution of ochre artefacts at Hohle Fels, Geißenklösterle and Vogelherd*

<i>Period</i>	<i>Hohle Fels</i>		<i>Geißenklösterle</i>		<i>Vogelherd</i>	
	<i>Total</i>	<i>NAA</i>	<i>Total</i>	<i>NAA</i>	<i>Total</i>	<i>NAA</i>
Holocene	21					
Magdalenian	164	67				
Gravettian	278	57	39	1		
A/G transition	35	12				
Aurignacian	371	47	149	8	234	18
Total	869	183	188	9	234	18

Notes: Comprehensive analyses have not yet been conducted on the ochre assemblages from Geißenklösterle and Vogelherd, therefore numbers come from existing excavation databases.

NAA, neutron activation analysis.

FullProf and Match software, and comparison with the Crystallography Open Database (Grażulis *et al.* 2009) and RRUFF database (Lafuente *et al.* 2015).

We used scanning electron microscopy (SEM) to investigate the microfabric of selected samples. The instrument is a Phenom Pharos desktop SEM (Thermo Fisher Scientific) located at the Senckenberg Centre for Human Evolution and Palaeoenvironment (HEP) Tübingen. It is equipped with a field-emission gun source and was operated in low-vacuum mode (60Pa) at 15 KeV.

RESULTS

Characterization by NAA produced concentrations for 33 elements in the assemblage of 183 ochre artefacts. Using multivariate statistical tests, we identified nine compositional groups within the data set. Figure 2 (a, b) are Fe-normalized, element-pair scatterplots showing the distribution of the groups. Similar clustering patterns were observed in scatterplots using combinations of rare earth (La, Ce) and transition metal elements (Cr, As), which is consistent

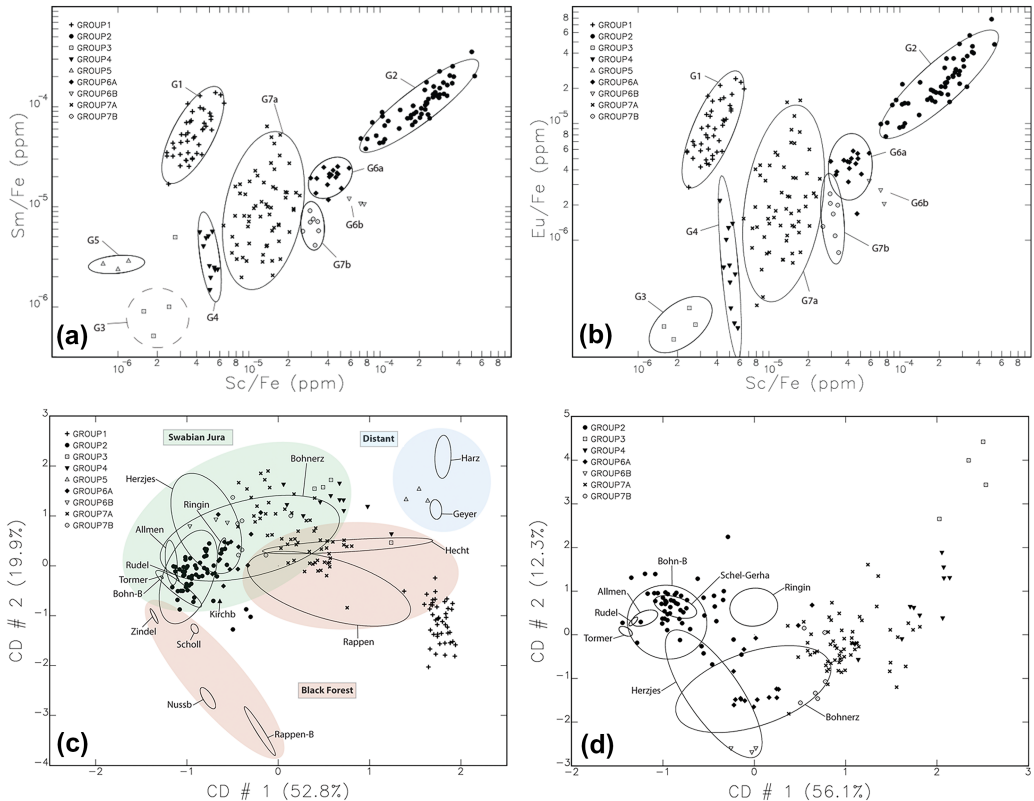


FIGURE 2 Multivariate analyses comparing sources and artefacts: (a) scatterplot of $\log_{10} \text{Sc/Fe}$ versus $\log_{10} \text{Sm/Fe}$; (b) the scatterplot of $\log_{10} \text{Sc/Fe}$ versus $\log_{10} \text{Eu/Fe}$. G5 was excluded due to Eu values below the detection limit; (c) canonical discriminant analysis (CDA) plot including all sources and all artefact samples sorted by group; and (d) CDA plot of selected Swabian Jura sources and compositional groups. In (c) and (d) ellipses represent sources and icons indicate the artefact samples per group. Ellipses are drawn at 90% confidence interval for all except for the dashed line G3 in (a). [Colour figure can be viewed at wileyonlinelibrary.com]

with elements that were diagnostic for the differentiation of regional ochre sources (Velliky *et al.* 2019). Concentrations of those elements were also positively correlated to Fe, suggesting they were not greatly impacted by weathering (Popelka-Filcoff *et al.* 2007). Table 2 shows group totals separated by time periods and sites. Most samples (about 84%) fall into one of three groups (G1, G2, G7a), with six smaller groups (G3, G4, G5, G6a, G6b, G7b) accounting for the remainder (about 16%). A representative sample from each group is shown in Figure 3. The means and standard deviations for each group are shown in Table S2, as well as the PCA results (Fig. S3 and Table S3). Group membership probabilities validated by Mahalanobis distance calculations are shown in Tables S6–S10.

The ochre samples from HF represent a diachronic and lateral distribution in 40 of the 71 excavated quadrants (see Figs S4–S5). Ochres from G2 have the highest representation in the Aurignacian period (about 39% of the total sampled Aurignacian assemblage). The Aurignacian-aged ochres constitute about 53% of the total population of G2, and are present in all groups except G1 and G3. The ochre sources inferred by G5 and G6b were exclusively accessed during the Aurignacian, and ochre from G6a was predominantly collected in the Aurignacian (about 87% of G6a is Aurignacian aged). During the Gravettian (34–30.5kcal. BP), almost all of the groups, except for G5 and G6b, are represented. The most abundant group represented in the Gravettian period is G1 (about 45% of all Gravettian-aged samples).

Table 2 *Archaeological group totals and subtotals for periods per site*

	Magdalenian			Gravettian			Aurignacian			Total
	HF	GK	VH	HF	GK	VH	HF	GK	VH	
G1	16			24						40
G2	13			10			26			49
G3	1			2						3
G4	2			3					5	10
G5							3			3
G6a	1			1			9	2	2	15
G6b								3		3
G7a	26			11	1		5	2	5	50
G7b				1			2	1	1	5
Total	59			52	1		45	8	13	

Notes: GK, Geißenklösterle; HF, Hohle Fels; VH, Vogelherd; G, group; Mag., Magdalenian; Grav., Gravettian; Aurig., Aurignacian. The bar graph shows in percentages the total sampled group populations from each site, organized by period.



FIGURE 3 Compositional group ochres: Examples highlighting the visual characteristics of ochre from each compositional group. Scales = 5 mm. [Colour figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

XRD and SEM

Mineralogical analysis by XRD was conducted on eight selected samples representing most groups. Samples from G1 and G3 were excluded as they were highly distinctive based on compositional data, and mineral data were deemed unlikely to advance the provenance interpretation any further. Instead, we opted to analyse two samples from G7a to assess within-group variation. All samples showed peak pattern matches for one or more Fe oxides, primarily hematite (Fe_2O_3), proto-hematite, goethite ($\text{FeO}(\text{OH})$), or Fe phosphate (see Table S4 and Figs S6–S7). Proto-hematite is a metastable phase with a unique structure, and may indicate high temperature thermal exposure (Burgina *et al.* 2000).

A representative artefact from each compositional group was selected for analysis by SEM to examine their micro-fabrics. The results showed distinctive fabrics, indicating different geological textures between the groups that may have played a role in their selection (see Fig. S8). Specimens from G1 and G5 contained platy micaceous particles. A summary of the key differences in chemical and physical characteristics between each group is shown in Table 3.

Comparison with the ochre source database

We compared the compositional groups to the ochre source database to determine if the artefact assemblage would uphold the *provenance postulate* (Weigand *et al.* 1977). The artefact and source databases were combined and re-examined using PCA, CDA, and Mahalanobis distance equations to calculate group membership probability (see Table S8). Results from CDA (Fig. 2, c) show that artefacts in G1 are distinctive and trend similarly to some Black Forest sources, however this is not consistent with projections of CD2 and CD3 (see Fig. S9). G5 plots with distant sources, which is consistent in other CDA projections (see Fig. S9). The majority of artefacts in the remaining compositional groups plot within the same clusters representing the local Swabian Jura region sources. This local cluster was investigated further (Fig. 2, d, and see Fig. S10), and showed G2 clustering with Fe oxide sources within 10km of the cave sites of HF and GK.

The results show five noteworthy observations: 1) G1 ochres were probably not acquired locally from the Swabian Jura; 2) G2 ochres are likely from a source in the immediate vicinity (about 20km) of HF and GK; 3) Ochre from G3, G4, G6a, G6b, G7a, and G7b could be from unsampled local or extra-local (< 80km) sources, but not the same outcrop as G2; 4) Ochres in G5 are not local and are likely from a distant (about 300km) source, and; 5) Local sources are present throughout the UP sequence, with some different sources accessed (G1, G5) before and after the LGM.

DISCUSSION

We can infer several aspects of ochre collection behaviours at HF, and some preliminary observations about behaviours at GK and VH. Ochres in G1 are likely not local to the Swabian Jura as their characteristics are significantly different from all identified local Swabian Jura sources, though there is the potential for an insofar unidentified local source. However, this is unlikely, as its characteristics are different from all identified local sources (i.e. lack of CaCO₃, enriched in actinides). G2 ochres are composed of iron-enriched limestone and likely originate from the Jurassic limestone bedrock outcrops near HF and GK (Fig. 1). Since GK is located only ~5km from HF, it is surprising that no G2 ochres were identified at GK. This could be influenced by the low number of ochres from GK (n=9), and additional sampling may offer more insight. Mahalanobis distance probabilities (see Table S8) and CDA (Fig. 2, c, and see Fig. S9) indicate that ochres in G5 trend with the distant sources of Geyer and the Harz Mountains, located about 300km from HF. Similar geological formations are found about 200km away in the far eastern region of Bavaria, still signalling long-distance transport of ochre.

The results show that G6a, G6b, G7a, and G7b are present at GK. The CDA projections (Fig 2, c, d) suggest that inhabitants at GK were accessing local Swabian Jura sources, though not the same source as G2. Ochres from G4, G6a, G7a, and G7b are present at VH in the Lone Valley. Those groups are distinct from the cluster of G2 (see Fig. S5, B), and some Swabian Jura sources (Torner, Rudel, Schel-Gerha, Allmen, Kirchb, Bohn-B), also suggesting the use of sources in an

Table 3 Summary of observations of the chemical and physical characteristics of the compositional groups

Group	n	Low variation (RSD about < 45%)	High variation (RSD about > 45%)	XRD	SEM observations	Macroscopic observations	Possible Provenance	Notes
G1	40	As, La, Nd, Sm, U, Yb, Ce, Co, Cr, Cs, Eu, Fe, Rb, Sb, Se, Tb, Th, Zr, Al, Ba, Ca, Dy, K, Mn, V	Lu, Hf, Ni, Sr, Ta, Zn, Na, Ti	n.a.	Laminated texture, platy particles	Purple to dark purple Fine-grained, mica inclusions Dark red streaks	Extra-local	Enriched in most rare earth elements (e.g., Nd, Sm, U, Th), visually and chemically homogeneous
G2	49		As, La, Lu, Nd, Sm, U, Yb, Ce, Co, Cr, Cs, Eu, Fe, Hf, Ni, Rb, Sb, Sc, Sr, Ta, Tb, Th, Zn, Zr, Al, Ba, Ca, Dy, K, Mn, Na, Ti, V	Calcite Fe phosphate	Granular with platy particles	Light to dark red Fine and coarse-grained sandstones Red to light red streaks	Local	Greatest internal variation, yet distinctive in Fe-normalized scatterplots
G3	3	Fe, Sc	As, La, Lu, Nd, Sm, U, Yb, Ce, Co, Eu, Hf, Ni, Sb, Th, Zn, Zr, Al, Ba, Ca, Dy, K, Mn, Na, Ti, V	n.a.	Porous, radiating fibrous Fe crystals	Light red to purple Fine-grained sand Light red streaks	Local Extra-local	High degree of internal consistency
G4	10	Sm, Fe, Sc, Al	As, La, Lu, Ns, U, Yb, Ce, Co, Cr, Cs, Eu, Hf, Ni, Rb, Sb, Sr, Ta, Tb, Th, Zn, Zr, Ba, Ca, Dy, K, Mn, Na, Ti, V	Hematite Quartz	Porous with oolitic Fe spheres	Dark red and purple Silty/sandy, mica inclusions Red to dark red streaks	Local Extra-local	Visually heterogeneous, chemically homogeneous

(Continues)

Table 3 (Continued)

Group	n	Low variation (RSD about < 45%)	High variation (RSD about > 45%)	XRD	SEM observations	Macroscopic observations	Possible Provenance	Notes
G5	3	As, La, Lu, Nb, Sm, U, Yb, Ce, Cr, Cs, Eu, Fe, Hf, Ni, Sb, Sc, Sr, Ta, Tb, Th, Zn, Al, Ba, K, Mn, Na, Ti, V	Rb, Zr, Ca, Dy	Hematite	Granular with platy particles	Dark purple/grey Silty Dark red streaks	Distant	Visually and chemically homogeneous
G6a	15	Sm, Eu, Fe, Sc	As, La, Lu, Nd, U, Yb, Ce, Co, Cr, Cs, Hf, Ni, Rb, Sb, Sr, Ta, Tb, Th, Zn, Zr, Al, Ba, Ca, Dy, K, Mn, Na, Ti, V	Hematite Proto-hematite	Granular with platy particles	Mixed purple/red Silty, mica inclusions Red to dark red streaks	Local Extra-local	Visually homogeneous
G6b	3	As, Lu, Nb, Sm, U, Cr, Eu, Fe, Rb, Sb, Sc, Sr, Al, Ba, Ca, Dy, Mn, Na, V	La, Yb, Ce, Co, Cs, Hf, Ni, Ta, Tb, Th, Zn, Zr, K, Ti	Goethite Hematite Calcite	Massive, botryoidal	Dark red/purple Silty/sandy Light to dark red streaks	Local Extra-local	Similar geochemical trends to 6a
G7a	50	Fe, Sc	As, La, Lu, Nd, Sm, U, Yb, Ce, Co, Cr, Cs, Eu, Hf, Ni, Rb, Sb, Sc, Sr, Ta, Tb, Th, Zn, Zr, Al, Ba, Ca, Dy, K, Mn, Na, Ti, V	Hematite Quartz	Granular with platy particles	Light to dark red and purple Silty/clayey Red to brown streaks	Local Extra-local	Possible collective 'local' ochre signature for Hohle Fels
G7b	5	Sm, Ce, Eu, Fe, Hf, Sc, Na	As, La, Lu, Nd, U, Yb, Ce, Co, Cr, Cs, Ni, Rb, Sb, Sr, Ta, Tb, Th, Zn, Zr, Al, Ba, Ca, Dy, K, Mn, Ti, V	Goethite	Granular with platy particles	Orange to brown Fine-grained sand Orange to red streaks	Local Extra-local	Potential subgroup of 7a

area different from G2 yet local to the region. Future research will include robust sampling of these and other cave sites in the Ach and Lone Valleys to explore these trends.

Patterns in the temporal distribution of artefacts in G1, G2, and G7a signal the long-term collection strategies by the inhabitants of HF. Ochres in G1 are mostly from the Gravettian (30.5–34ka; n=24, about 60%) and Magdalenian (14.5–16.5ka; n=16, about 40%) periods, and are the most chemically, physically, and visually homogenous (Table 3). The source that this group represents, though its location yet unidentified, was well-known to HF inhabitants after its discovery, and was increasingly favoured during and after the Gravettian, coinciding with the narrowing of ochre preferences at HF to silty and purple ochres producing a dark red powder (Velliky *et al.* 2018).

G7a is the largest and most varied of the compositional groups. Inhabitants from all three caves collected ochres from this source throughout the UP. Two artefacts from GK and five from VH, all Aurignacian, were collected from the G7a source (Table 2). Moreover, ochres in G7b are chemically similar to G7a and may represent a sub-outcrop or spatial variation within that source deposit. This scenario could suggest that G7a was a larger, spatially widespread source as intra-source variability can be moderate in exposed and weathered environments (Popelka-Filcoff *et al.* 2007; Macdonald *et al.* 2011).

As the sample selection is larger for HF compared to the other caves, compositional groups unique to the HF ochre assemblage are to be expected, and is realized, in G1, G2, G3 and G5. The current data also show that G6b ochres came from a source that was accessed exclusively by Aurignacian groups at GK. The distribution of samples at all sites suggests that ochre collection strategies during the Aurignacian were different than later time periods, and that either opportunistic acquisition strategies, individualistic collection preferences, or genuine differences in cultural expression during the Aurignacian existed. We cannot exclude the possibility that certain landscape or environmental features may have impacted accessibility, affecting the availability of sources. For instance, environmental fluctuations during and after the LGM caused a drop in water table and increased hillside erosion in the Ach and Lone valleys (Barbieri *et al.* 2018), which may have increased the surface accessibility of ochre outcrops.

Following the Gravettian, aside from landscape changes affecting collection opportunities, another hypothesis is that sources with specific ochre qualities were collected preferentially. These qualities need not be limited to physical attributes, though this is seen with the increase of purple, fine-grained micaceous ochres in the Gravettian and Magdalenian (Velliky *et al.* 2018). Desirable qualities can also rest in physical places and the association of certain areas or landscape features to symbolic or ritual aspects. In ethnographic examples, specific raw materials are sought after not because they are of a superior quality (however, see Rifkin 2015b) but because they are from a symbolically charged place (Bradley 2013), even if those places are located at a great distance. This scenario is plausible in the context of the symbolic associations of ochre throughout widespread temporal and geographic contexts, and coincides with the increase in symbolic expression found during the Aurignacian (Hovers *et al.* 2003; Salomon 2009; Watts 2009; Macdonald *et al.* 2011; Roebroeks *et al.* 2012).

In the assemblage from HF, the Gravettian and Magdalenian sequences contain ochre with traces of anthropogenic modification (n=27), an assortment of artefacts containing traces of red residues including an ochre grindstone (Velliky *et al.* 2018), and limestone fragments with red painted dot designs (Wolf *et al.* 2018). Currently, the HF Aurignacian has no definitive examples of anthropogenically modified red ochre pieces, though there is one yellow ochre piece with incisions (Velliky *et al.* 2018). The comparative lack in modified pieces during the Aurignacian can be attributed to post-depositional processes, cave-specific activities, or differences in ochre

behaviours between the time periods. However, the number of ochre pieces and ochre-related artefacts throughout the UP sequence attest to the sustained significance of ochre to the cave inhabitants. Ochre is found in the earliest Aurignacian layer at HF (layer Vb) and is subsequently present in every anthropogenic horizon, suggesting that ochre use was a well-established behavioural practice that was engrained in daily life. Those practices likely played varying roles over time, as evidenced by the change in ochre types collected throughout the UP. Though its specific meaning may have fluctuated, the provisioning of ochre by UP populations at HF remained constant over 29 500 years.

Comparison with other raw material-acquisition behaviours

Other material culture forms illustrate behavioural changes throughout the UP in the Swabian Jura. Figure 4 shows proposed regional movement networks based on, lithic, and osseous resources. At HF, the lithic assemblage is dominated by local resources during the Aurignacian (Hahn 1987), with 96% of the assemblage being a local chert called *Jurahornstein* (Scheer 2000). During the Gravettian, only 56% of the lithic material is made from *Jurahornstein*, with a corresponding increase in non-local lithic materials indicating expanding resource acquisition areas after the Aurignacian (Hahn 1987; Burkert and Floss 1999; Scheer 2000; Floss and Kieselbach 2004). Most of the proposed migration routes for the Swabian Jura sites are oriented along an east–west axis following major waterways like the Danube (Hahn 1987; Burkert and Floss 1999). The association of G5 to a distant source originating northeast of the Swabian Jura diverges from these hypotheses; however, similar geological formations also occur along the eastern border of Bavaria (about 200 km east), and may have yielded accessible ochre formations during the late Pleistocene. Though the possibility of ochre from that region would be more consistent with lithic acquisition patterns during the Magdalenian, it still indicates a wider area of movement, or perhaps broader social networks, during the Aurignacian of Central Europe, which is earlier than was previously proposed.

Ochres at HF that were collected during the Aurignacian came from local and extra-local sources exhibiting higher diversity in ochre type and texture. During the Gravettian and Magdalenian, preference shifted toward purplish ochres with silty textures from either local or extra-local areas. This pattern contrasts with the lithic assemblage, which is dominated by a homogenous set of local materials in the earlier Aurignacian, followed by an increase in extra-local and greater diversity of raw materials in later time periods. These inverse patterns indicate that people maintained collection strategies unique to different material types, suggesting that they played different roles within the fabric of society (Porr 2010a).

The Gravettian period saw expanded resource acquisition over Western Eurasia as well as in the Swabian Jura (Scheer 2000; Tallér *et al.* 2019), with social networks spanning greater distances and increased diversification in ochre behaviours throughout Europe (Hahn 1987; Pettitt *et al.* 2003). Material culture styles and forms became increasingly standardized and ubiquitous compared to the Aurignacian (Soffer *et al.* 2000; Porr 2010b). Though the presence of Mediterranean and Atlantic mollusc shells during the Gravettian occupation of HF and GK suggest a broader resource collection area (Hahn 1988; Scheer 2000), evidence for ochre provision from distant sources during the Aurignacian indicates that trade and/or movement networks were already in place before the onset of the LGM. This was previously suspected based on the presence of an ammonite fossil from the Black Forest at VH (Hahn 1987). Our results provide additional evidence for the presence of long-distance interactions during the Aurignacian, which

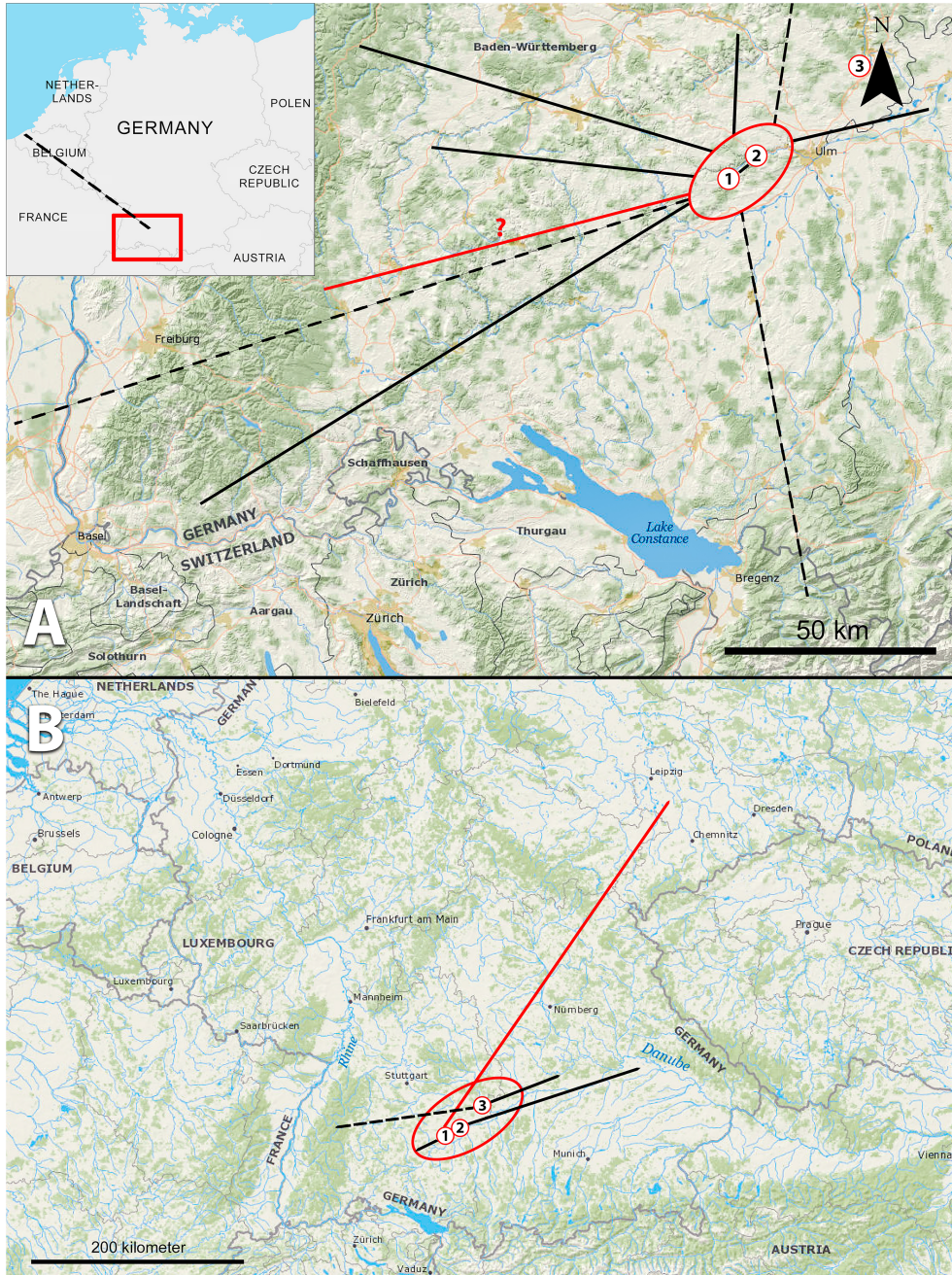


FIGURE 4 Comparison of the proposed transportation networks during the Upper Palaeolithic: (1) Hohle Fels, (2) Geißenklösterle and (3) Vogelherd. Red lines and circles are possible ochre procurement mobility or exchange networks. (a) Proposed movement routes for the Gravettian and Magdalenian. Solid black lines represent lithic transportation (Scheer 2000); and dashed lines represent shell pendant transportation, including long-distance, left inset (Rähle 1994). (b) Proposed routes for the Aurignacian. Solid black lines represent lithic transportation; and dashed lines correspond to movement of notched shell fossil (Hahn 1987). [Colour figure can be viewed at wileyonlinelibrary.com]

is in agreement with recent studies suggesting early Aurignacian populations were more widespread over Europe than previously suspected (Cortés-Sánchez *et al.* 2019).

The ochre procurement patterns observed between the cave sites corroborate other evidence that communication and cohesion existed between groups in the Ach Valley throughout the UP. Studies on the lithic assemblages show the refitting of some Gravettian and Magdalenian artefacts between HF, GK, Sirgenstein and Brillenhöhle in the Ach Valley (Scheer 2000, Taller *et al.* 2019). The presence of certain lithic technological stages suggests that HF and Brillenhöhle operated as base camps, and GK and Sirgenstein functioned as temporary satellite occupations (Taller *et al.* 2019). Another scenario is that smaller groups inhabited specific caves at alternate points in time, or that certain activities were exclusive to specific caves (Scheer 1990). It is probable that the caves were primarily occupied during the winter, indicated by faunal evidence (Münzel and Conard 2004), with migrations to smaller seasonal camps during warmer periods. However, this hypothesis is difficult to test as few intact open-air sites have been found in this region.

Cultural exchange between caves in the Ach and Lone Valleys is also seen during the Aurignacian. Figurines and personal ornaments made from mammoth ivory are found at HF, GK, VH, and other caves (Conard 2003; Conard 2009; Dutkiewicz *et al.* 2018). While there are subtle differences in the aesthetic arrangement of patterns at each site, the similarities are overarching and suggest shared cultural elements (Dutkiewicz *et al.* 2018). Ochre provenance results also indicate that these groups were not isolated; while there was some level of cultural exchange (e.g., ivory ornaments, figurines), the patterns of ochre collection suggest groups kept to certain areas and the resources contained within them (for instance, G6a ochres exclusively collected by GK inhabitants). This indicates a new dimension of variability related to material culture, in this case ochre, which emerges through an interplay of collective (social) and individual behaviours.

The results regarding ochre use at HF present four aspects: 1) Local (< 80km) ochre outcrops were used throughout the UP, and distant (about 300km) sources were accessed earlier than previously suspected; 2) An intimate knowledge of the landscape surrounding the caves as evidenced by raw material acquisition strategies and the longevity of ochre acquisition patterns; and, 3) Ochre-related behavioural patterns that were shared yet kept unique, as seen in the presence of ochre from specific sources throughout the UP.

CONCLUSIONS

This paper advances our understanding of the behavioural complexities of AMHs during the European UP, a pivotal period of symbolic, cognitive, and cultural evolution. Although evidence for ochre use in the Swabian Jura is not as abundant as in other sites in Europe (Salomon *et al.* 2008; Salomon 2009) and Africa (Watts 2009; Hodgskiss 2014; Rosso *et al.* 2016; Hodgskiss 2020), it is possible to illuminate the nuances of ochre selection strategies of AMHs as they migrated into the continent and adapted to subsequently changing landscapes. It is evident that AMHs had established symbolic behaviours once they arrived in Central Europe, and included in this palimpsest is the recognition of ochre as an important facet of individual and cultural expression.

The results show that ochre behaviours were part of an intricate network of source locations and selective preference, possibly reaching distances up to 300km, some of which were maintained over millennia. Ochre was evidently an important cultural item, as reflected in the loyalty to certain local sources and maintained throughout generations, despite dramatic shifts in mobility patterns, and climatic and environmental conditions. Though there was likely exchange of ochre, among other resources, between the Swabian Jura caves, specific ochre locations were exclusively accessed by people from certain sites, as shown by some compositional groups unique

to HF and GK. Whether the locations were part of established territories or ownerships maintained through socio-political means is possible, although difficult to discern. What we can infer is a new dimension of behavioural complexity during the European UP via the application of scientific techniques to a so-far underappreciated artefact category.

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PEER REVIEW

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

Table S1. Descriptive information on Geißenklösterle (GK) and Vogelherd (VH) ochres. Note that since VH was an excavation of the back-dirt and ochre artefacts were not systematically collected, no stratigraphic or spatial information is provided. Data on HF ochres can be found in (Velliky et al., 2018).

Table S2. Means and standard deviations for raw elemental values on ochre artefact samples. All values are represented in ppm unless otherwise stated. BDL = below limit of detection.

Table S3. Percentage variation and cumulative percentage variation for each principal component.

Table S4. Summary of XRD results showing samples, chemical groups, and major phases present.

Table S5. Summary of outlier CDA group membership probabilities using Mahalanobis distance for archaeological compositional groups. The results are based on the first component, explaining 79.8% of the variance. The bivariate elemental pair projections of the outliers against the compositional groups is shown in SI Figure 10.

Figure S1. Selected ochre artefacts from Vogelherd, Geißenklösterle and Hohle Fels, showing the visual and textural variety in ochre types and sizes. Scale = 1 cm.

Figure S2. Stratigraphy of Hohle Fels and Geißenklösterle organized by archaeological horizons (AH).

Figure S3. Biplot from RQ-mode PCA showing PC1 versus PC2. Compositional groups surrounded by 90% confidence ellipses. Principal component analysis was performed on the Fe-normalized \log_{10} -transformed data set, including artefact samples and all possible elements: Na/Fe, V/Fe, Co/Fe, Zr/Fe, La/Fe, Dy/Fe, Al/Fe, Cr/Fe, Zn/Fe, Sb/Fe, Ce/Fe, Th/Fe, Ca/Fe, Mn/Fe, As/Fe, Cs/Fe, Sm/Fe, U/Fe and Sc/Fe (excluding K/Fe, Ti/Fe, Ni/Fe, Rb/Fe, Sr/Fe, Ba/Fe, Nd/Fe, Eu/Fe, Tb/Fe, Yb/Fe, Lu/Fe, Hf/Fe, and Ta/Fe due to excessive values below the limit of detection).

Figure S4. Outline of Hohle Fels cave excavation area showing the distribution of ochre artefacts characterized with NAA, labelled by time period. Group 6b is not shown as this group is purely GK ochres.

Figure S5. Outline of Hohle Fels cave excavation area showing the distribution of ochre artefacts characterized with NAA, labelled by compositional group.

Figure S6. XRD spectra with identified peak locations. Analyzed artefacts are shown in bottom right corner. All scales are set to 1cm.

Figure S7. XRD spectra with identified peak locations. Analyzed artefacts shown in bottom right corner. All scales are set to 1cm.

Figure S8. SEM BSE images of the surface of some representative examples of the identified compositional groups. Numbers in the top left correspond to compositional groups, and are as follows: 1) HF95.57.IIb.449, laminated texture with platy particles, 2) HF91.78.IIa.201, granular with platy particles, 3) HF10.110.IIb.1166, porous with radiating fibrous iron crystals 4) HF94.67.IIa.208, porous with spherical aggregates of iron, 5) VHC004, granular with platy micaceous particles, 6a) HF13.54.IIIa.363, granular with platy particles, 6b) GK01.76.IIIa.1222, massive, botryoidal 7a) VHC001, granular with radiating fibrous iron crystals, 7b) HF99.77.IIc.770, granular with platy particles.

Figure S9. Bivariate plot of CD#2 versus CD#3 of all sampled sources and compositional groups. Ellipses indicate the projection of sources, while icons indicate the individual samples in each compositional group. Ellipses encircling the source groups are at 90% confidence.

Figure S10. Bivariate plot of CD1 versus CD2 of selected Swabian Jura sources and compositional groups. Projection includes only Swabian Jura sources and all artefact samples sorted by group, except for Groups 1 and 5. Ellipses indicate the projection of sources, while icons indicate the individual samples in each compositional group. Note the exclusion of the Ringingen source. Ellipses encircling the source groups are at 90% confidence.

Figure S11. Scatterplot of $\log_{10} \text{Sc/Fe}$ versus $\log_{10} \text{Sm/Fe}$ for ochre artefact Groups 1, 6a, 6b, 7a, and 7b, showing the distribution of groups. Ellipses around the clusters represent 90% confidence levels for group membership. This plot further illustrates the separation of Groups 6a, 6b, 7a, and 7b.

Figure S12. Scatterplot of $\log_{10} \text{Sc/Fe}$ versus $\log_{10} \text{Eu/Fe}$ for all ochre artefact samples, showing the distribution of compositional groups and the placement of outliers in compositional hyperspace. Ellipses around the clusters represent 90% confidence levels for membership in those groups. Note the omission of G5 as that chemical group did not report levels of Eu above the limit of detection.

Table S6. Group membership probabilities on all groups using Mahalanobis Distance calculation. Results are based on the first component, explaining 79.8% of the variance.

Table S7. Group membership probabilities on only large ($n > 10$) using Mahalanobis Distance calculation. Results are based on the first eight components, explaining 100% of the variance. Best group is based on highest membership probability $> 0.001\%$

Table S8. Group membership probabilities using Mahalanobis Distance calculation. Results are based on the first two components, explaining 72.7% of the variance. Best group is based on highest membership probability $> 0.001\%$

Table S9. Group membership probabilities using Mahalanobis Distance calculation. Results are based on the two discriminant functions, explaining 67.3% of the variance.

Table S10. Group membership probabilities using Mahalanobis Distance calculation. Results are based on the first two discriminant functions, explaining 67.3% of the variance.