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MULTIPARAMETRIC CHARACTERIZATION OF SOUTHWESTERN GERMAN CHERTS
Application to the study of raw material circulation during the Upper Paleolithic period

Céline BRESSY[1] **& Harald FLOSS**[2]

1. Max-Planck Institut für Chemie, Germany; UMR 6636, 5 rue du Château de l'Horloge, 13094 Aix-en-Provence, France. celine.bressy@gmail.com
2. Eberhard-Karls-Universität Tübingen, Institut für Ur- und Frühgeschichte und Archäologie des Mittelalter Abteilung Ältere Urgeschichte und Quartärökologie, Schloß, Burgsteige 11, D-72070 Tübingen, Germany. harald.floss@uni-tuebingen.de

Abstract. This paper presents the first results of a methodological approach for the characterization of cherts in Southwest Germany. Many of the chert varieties found in the study area resemble each other macroscopically, which makes reconstructing raw material circulation patterns difficult, and affects our ability to make inferences about patterns of prehistoric human mobility based on raw material transfers. Jurassic cherts, which are assumed to have been imported over great distances into the Swabian Alb, are a good example of the need for more reliable criteria with which to distinguish local and foreign raw material. We have initiated a multiparametric approach, combining petrography and geochemistry using non-invasive techniques, on 12 geological samples of Jurassic flint. Petrographic microfacies were studied by stereomicroscope. Trace element analyses were performed by laser (LA)-ICP-MS (Inductively Coupled Plasma-Mass spectrometer). Combining criteria from petrographic and trace element analysis we propose discrimination patterns for each of the considered sources on the basis of microfossil occurences and mainly of Rb, Zr, Nb, Hf, Pb, U and rare earth elements (REE) contents for the later. The results are encouraging and we suggest that our approach can be used to analyse artefacts for archaeological applications.

Résumé. Ce papier expose les premiers résultats d'une approche méthodologique menée sur la caractérisation de silex du Sud-Ouest de l'Allemagne. Parmi les variétés de ce secteur géographique, certaines peuvent présenter des ressemblances macroscopiques susceptibles de fausser la reconstitution des circulations de matière première, ayant des implications sur la connaissance de la mobilité humaine au cours de la préhistoire. C'est le cas de plusieurs variétés de silex jurassiques qui sont supposés avoir été importés sur de longues distances dans les sites paléolithiques du Jura souabe. Afin de disposer de critères fiables pour distinguer les silex jurassiques souabes de ceux allochtones, nous avons entrepris sur 12 échantillons géologiques une approche multiparamétrique combinant la pétrographie et la géochimie faisant appel à des techniques non-destructives. Les microfaciès pétrographiques ont été étudiés par stéréomicroscope. Les analyses d'éléments traces ont été réalisées par ICP-MS (Inductively Coupled Plasma-Mass spectrometer) à ablation laser. En combinant ces deux approches, nous proposons des critères de discrimination pour chaque source considérée, plus particulièrement basés sur la présence de certains microfossiles et les teneurs en Rb, Zr, Nb, Hf, Pb, U et terres rares. Ces résultats nous encouragent à poursuivre notre démarche sur des artefacts, pour développer les applications archéologiques.

Introduction

The Swabian Jura constitutes a key area for understanding the evolution of the Early Upper Paleolithic in Central Europe (Conard & Bolus 2003). The numerous sites discovered in this area, including Hohlenstein-Stadel, Hohle Fels, Geissenklösterle and Vogelherd, and their dates provide a wealth of information on the first stages of Central European colonization by modern humans. Among the artefacts recovered, a set of mammoth ivory figurines stands out, and appears as an expression of cultural innovation and human evolution (Hahn 1986; Conard 2003; Floss 2004). In such a context, raw materials analyses are a powerful tool to trace modern human mobility patterns and raw material procurement strategies, and to study interregional contacts. It can emphasize both economic and cultural features of these hunter-gatherer societies.

Recent studies (Burkert 1998; Burkert & Floss 2005; Floss & Kieselbach 2006) have dealt with raw material procurement. They provide data on flint outcrop distribution in Southwestern Germany and draw an outline of procurement patterns and their evolution throughout the Upper Paleolithic. Benefiting from these advances, the present work is focused on one aspect of raw material supply: the identification of long distance circulation patterns. To enable archaeological reconstruction of far-flung movements, a reliable means of characterizing cherts is essential. We propose a multiparametric approach for the characterization of cherts in Southwestern Germany.

Geological and archaeological context

The Swabian Jura is a highland located in Southwest Germany, bounded in the west by the Black Forest and to the east by the Franconian Alb. Its southern border is marked by the Danube. Consisting of sedimentary formations, the Swabian Jura and its borders offer numerous chert outcrops in Triassic, Jurassic and Tertiary formations (Burkert & Floss 2005).

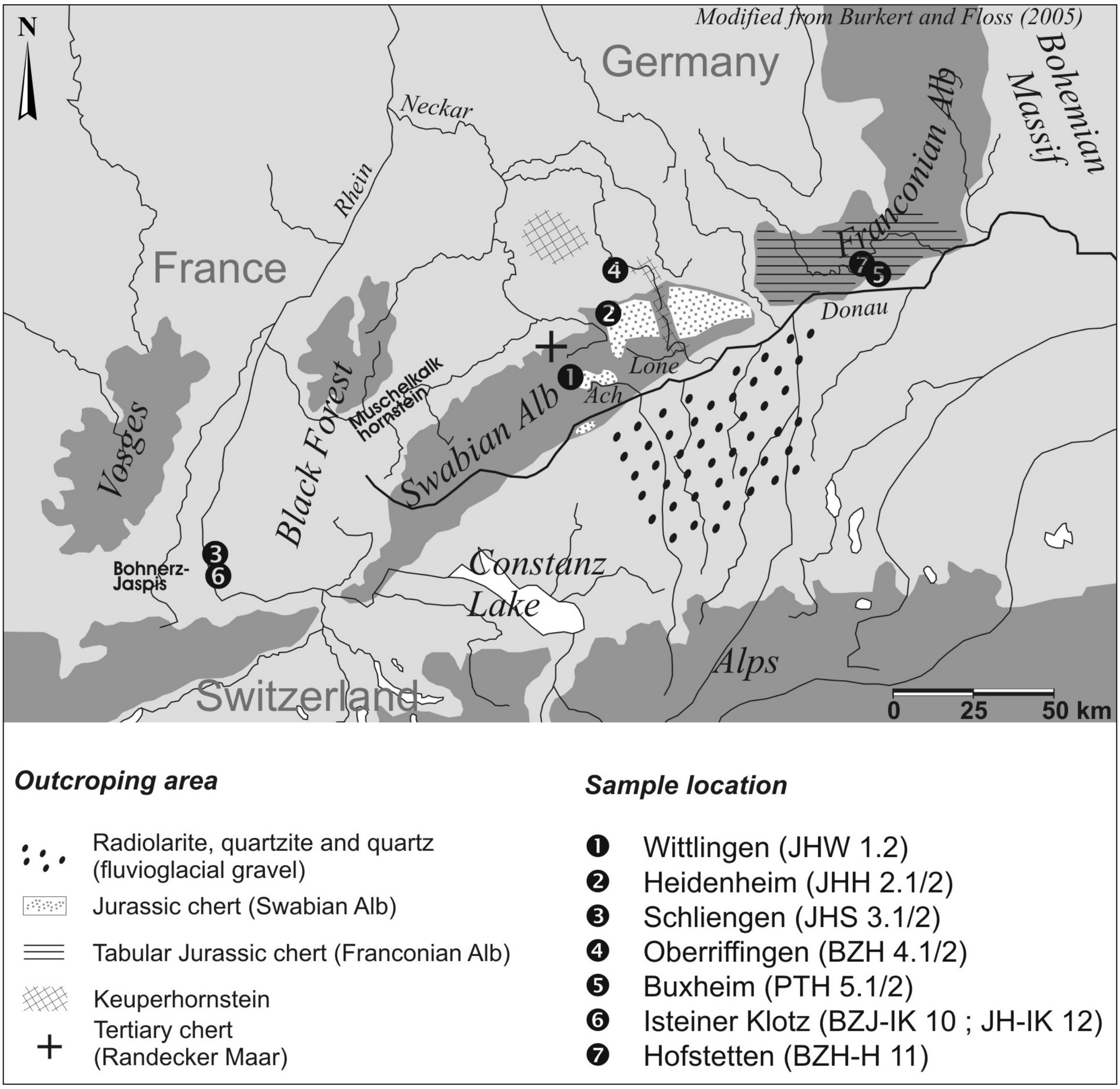

Figure 1. Siliceous resources in Southwestern Germany and location of the samples.

As described by Burkert (Burkert 1998), each deposit may contain different types of chert distributed in specific areas (fig. 1). While Triassic cherts (Muschelkalkhornstein and Keuperhornstein) are to be found in the regions North of the Swabian Jura, Jurassic cherts are widespread throughout the Swabian and Franconian Jura. Bohnerzhornstein corresponds to Jurassic chert tinted by oxydes during the Cenozoic. As secondary occurences, it occurs in river deposits across the uplands and along the Southern edge of the Swabian Alb. Finally, Tertiary cherts are encountered in the Randecker Maar and are linked to volcanic activity. Beside cherts, a range of other siliceous materials are available in alluvial deposits: radiolarite, quartzite and quartz.

Based on this knowledge of regional siliceous resources, Burkert (Burkert 1998; Burkert & Floss 2005) carried out a qualitative raw material provenance study on 16 Aurignacian, Gravettian and Magdalenian Swabian lithic assemblages. Until then, the considered area was lacking of such study (Féblot-Augustins 1997). The results show that Jurassic cherts, occurring locally, dominate the raw material spectrum. Bohnerzhornstein and radiolarite are of secondary importance. In addition to these materials, Triassic and Tertiary cherts and quartzite complete the lithic spectra. As regard to foreign materials, Burkert points out the recurrent presence, in Aurignacian to Magdalenian settlements, of a fine grained tabular chert (Plattenhornstein). On the basis of macroscopic examinations, it is assumed to originate from Jurassic formations of Franconian Alb and/or Abensberg area, that is, 120 km away on average from the Swabian settlements, indicating possible eastward connections. Beside the Plattenhornstein, the so-called Bohnerzjaspis has been noted occasionnally, for example in Gönnersdorf in Magdalenian levels (Rhineland, Floss 1994). Bohnerzjaspis outcrops occur in the Oberrhein area, on the Western slope of The Black Forest, in a distance of 300 km from the site.

All of Burkert's identifications were based on macroscopic examination. However, some of the cherts that are assumed to be of distant origin closely ressemble local Swabian varieties. This is mainly the case for Jurassic cherts: for example, on one hand, Swabian Jurahornstein resembles Franconian Plattenhorstein and on the other hand, Swabian Bohnerzhornstein resembles Upper Rhine Bohnerzjaspis. Thus, confusions between these cherts are possible and may lead to mistaken assignments. Accurate discrimination of foreign raw materials are of primary importance when discussing intergroup relationships, long distance eastward circulation patterns, and the part played by the Danube Valley in the populating of Central Europe by first modern humans. Therefore, we have initiated petrographical and geochemical research aimed at the proper characterization of Jurassic cherts from the Upper Rhine area, the Swabian Alb and the Franconian Alb. Since our research is still in an exploratory stage, the first step of our study has consisted in the analysis of 12 geological samples (tabl. 1) in order to fingerprint Jura- and Bohnerzhornstein chert varieties from Southern Germany.

Methodology

Our analyses were performed at the Max-Planck-Institut für Chemie of Mainz (Germany). Non-destructive petrographic analyses were undertaken on bulk samples under the stereomicroscope. They are based on the observation of the frequency, dimension and nature of carbonate elements, oxydes, organic matter and micropalaeontological contents (Bressy 2003).

Trace element composition was measured by laser ablation (LA)-ICP-MS (Inductively Coupled Plasma Mass Spectrometry) allowing us to determine element composition for up to 30 elements: Rb, Sr, Y, Zr, Nb, Cs, Ba, Hf, Ta, Pb, Th, U, Ni, Co, Ge, Sn, including the Rare Earth Elements (REE): La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu. Through the action of a high intensity ultra-violet laser beam, a small volume of the sample is removed and analysed using the ICP-MS. This relatively non-invasive method produces a 120 µm diameter crater which is invisible to naked eye. On each sample, six to nine ablation craters were made to provide measurements in different parts of the sample. Composition data are thus the result of the average of these different points of analysis. The international standard NIST 612 (Pearce *et al*. 1997) was used as external standard to calibrate the analyses. An assumed content of Si, chosen as the internal standard, has been used to calculate the approximate element concentrations. Therefore, we can not directly compare the calculated content of each element but only ratios of one element to another.

Results

Petrographic approach

Petrographic descriptions are reported in table 2. For each variety considered, Jurassic cherts and Bohnerzhornstein, we provide criteria useful to distinguish the material from that coming from other sources.

A variety of facies of Jurassic chert are identified, consistent with the different outcroping areas sampled. For the Swabian Jurahornstein, the Wittlingen sample is characterized by the abundant occurrence of sponge spicules. The Heidenheim samples show a quantity of lithoclasts and no microfossil content. Moreover, the two Heidenheim outcrop samples reveal slight differences in microfacies, highlighting intra-source petrographic heterogeneity. Thus, Swabian Jurahornstein offers hemipelagic facies in the former outcrop, and "breccia"

Type	Laboratory reference	Area	Location	Geology	Outcrop	Morphology	Quality	Samples
Jurahornstein	*JHW 1.2*	Swabian Jura	Wittlingen	Oberer Jura (Malm)	Primary position	Nodules	+	
	JHH 2.1	Swabian Jura	Heidenheim, Brenzschotter	Oberer Jura (Malm)	Brenz alluvial deposits	Eroded nodules	+	Distinct blocks
	JHH 2.2							
	JHS 3.1	West of the Black Forest	Schliengen (Markgräfler Hügelland)	Oberer Jura (Malm, Rauracien)	Primary position	Round to ovoïd nodules	+++	Same block
	JHS 3.2							
Bohnerzhornstein	*BZH 4.1*	Swabian Jura	Oberriffingen (Bopfingen)	Oberer Jura (Malm)	Secondary position	Pebbles and nodules	++	Same block
	BZH 4.2							
Plattenhornstein	*PTH 5.1*	Franconian Alb	Buxheim (bei Ingolstadt)	Oberer Jura (Malm)	Primary position	Tablets	+++	Distinct blocks
	PTH 5.2							
Bohnerzjaspis	*BZJ-IK 10*	West of the Black Forest	Isteiner Klotz	Oberer Jura (Malm)	Secondary position	Nodules	+++	
Bohnerzhornstein	*BZH-H 11*	Franconian Alb	Hofstetten (Eichstätt)	Oberer Jura (Malm)	Secondary position	Irregular nodules	+	
Jurahornstein	*JH-IK 12*	West of the Black Forest	Isteiner Klotz	Oberer Jura (Malm, Rauracien)	Primary position	Nodules	+++	

Table 1. Descriptive list of the analyzed samples.

Type	Laboratory reference	Macroscopic description		Stereomicroscope observation		
		Cortex	Appearance	Texture	Elements	Microfossil content
Jurahornstein	JHW 1.2	1-5 mm, compact	Light grey, fine-grained, opaque	Packstone - 20 %	Round calcareous elements	Numerous Sponge spicules
	JHH 2.1	Siliceous eroded natural surfaces	Light grey, heterogeneous, coarse-grained inclusions, fractured, opaque	Packstone - 25 %	Many heterometric calcareous spheroïds (up to 1 mm), lithoclasts : "breccia facies"	Not observed
	JHH 2.2		White to greyish to brownish orange, heterogeneous, spoted, coarse-grained, opaque	Packstone - 20 %	Heterometric calcareous spheroïds (up to 1 mm), lithoclasts, less abundant	Not observed
	JHS 3.1, JHS 3.2	Natural flat oxidized surfaces	Light grey, homogeneous, with banded parts (off-white to brown-grey), fine-grained, opaque	Mudstone - 2-5 %	Scarce oxides, round calcareous heterometric elements (biogenic ?) concentrated in white beds, organic matter	Few Sponge spicules, fragmented, unidentified fossils
	JH-IK 12	Thick, white, compact, siliceous, yellowish in surface	Light grey, banded, fine grained, slightly translucent	Mudstone - 5-8 %	Not visible	Numerous unidentified fossils as previous sample
Plattenhornstein	PTH 5.1	1 mm, grey, compact	Grey, slightly homogenous, lightly banded, fine grained, opaque	Mudstone to wackestone - 8-18 %	Orange iron oxydes, calcareous elements, filaments, gravels among which some are impregnated with oxides	Few Sponge spicules
	PTH 5.2	Pellicular, ochre	Light grey to grey, banded, fine grained, opaque	Mudstone to wackestone - 8-18 %	Small calcareous particules, lithoclastes (up to 1 mm), micro-laminations	Not observed
Bohnerzhornstein	BZH 4.1, BZH 4.2	2-3 mm thick, white in section	Greyish to orange, fine-grained with coarse-grained inclusions, fractured, opaque	Mudstone - recrystallization	Round calcareous elements impregnated with yellow oxides, scarce fine calcareous particules (<100 µm), lithoclastes	Scarce Sponge spicules
	BZH-H 11	Thin eroded and porous cortex	Red to mustard-yellow, mid-grained, opaque	Packstone - 40 %	Quartz, vacuoles, rounded calcareous elements	Monaxone and triaxone spicules, Foraminifers
Bohnerzjaspis	BZJ-IK 10	Siliceous eroded natural surface	Bright red with yellow veins, fine grained, opaque	Recrystallisation	Oxides impregnations modified the original texture, unidentified elements	Not observed

Table 2. Petrographic description of the samples.

or detrital facies in the latter one. West of the Black forest, Jurahornstein outcrops offer characteristic banded mudstone facies. Their bioclastic content is also very specific although the nature of the fossils observed still has to be identified. Finally, Franconian Alb Plattenhornstein can be distinguished by its tabular morphology, banded aspect and the presence of filaments, iron oxides and the scarcity of microfossils.

Bohnerzhornstein and Bohnerzjaspis are discriminated based on flimsier criteria. As we are dealing here with Jurassic cherts subjected to oxidation, the transformation of their initial structure is more or less complete depending on the chert. The primary structure and texture of Bohnerzjaspis (West of the Black Forest) is totally hidden by oxide impregnation, which can be considered as a peculiar discriminating feature. Bohnerzhornstein from the Swabian Jura and the Black Forest are distinguished by their respective microfossil contents. Furthermore, the latter shows more pronounced colouring. At last, Bohnerzhornstein from Franconian Alb microfacies are close to those of Swabian Jura. However the former can be discriminated on the basis of the occurrence of Foraminifera.

Geochemical analyses

Since several measurements are made on each sample, we get an accurate overview of intrasample heterogenity. Thus, the variation coefficient of elemental contents within a sample can vary between 1.5 and 89%. Between samples from a single source, compositional variations can also be important. Some elements were often at or just below instrumental detection limits: for example, REE, Ta. Others, like Sn, showed little or no compositional variation between the samples.

Qualitative results

Even if the calculated abundances can not be considered directly (cf. § 3), they at least point out some tendencies that may be useful for the discrimination of Jurahornstein sources

Type	Laboratory reference	Specific composition	
		High contents	*Low contents*
Jurahornstein	*JHW 1.2*		
	JHH 2.1	Ba Zr	Rb
	JHH 2.2	Sr, Y, REE	
	JHS 3.1		REE
	JHS 3.2		U
	JH-IK 12	Rb, Cs	REE
	PTH 5.1		
	PTH 5.2		REE, Pb
Bohnerzhornstein	*BZH 4.1*		
	BZH 4.2		
	BZH-H 11	Y, Zr, REE, Hf, Pb	
	BZJ-IK 10	REE, Pb	

Table 3. Main discriminant elements for Jurahornstein and Bohnerzhornstein.

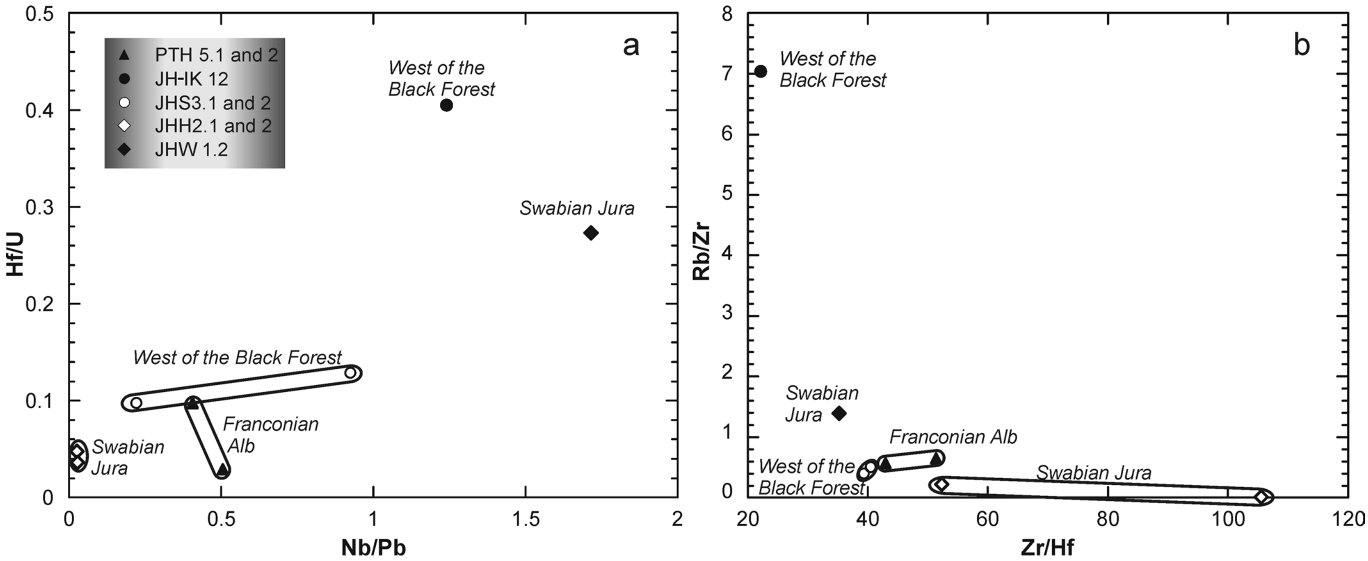

Figure 2. Binary diagrams Nb/Pb vs Hf/U (a) and Zr/Hf vs Rb/Zr (b) for the Jurahornstein analyzed.

on the one hand, and Bohnerzhorstein outcrops on the other.
Some sources appear to be characterized by particularly high or low contents of different elements (tabl. 3). Samples JHW 1.2, PTH 5.1, BHZ 4.1, BHZ 4.2 show average composition in most elements. Samples JHH 2.1/2, JH-IK 12, BZJ-IK 10, BZH-H 11 display high content in specific elements. These occurrences may be explained by the presence of specific minerals or impurities relating to chert formation or diagenetic history (Luedtke 1992). Therefore, they act as reliable fingerprinting patterns. Based on the association between high Y, Zr, REE, Hf, and Pb levels, clays must have played a part in the genesis of BZH-H 11. The same explanation could be proposed for other samples showing high Pb, Rb, Cs and REE contents. Ba could be associated to clays as well as to organic matter.

Quantitative results

Here, we investigate the potential of element ratios as a means of discriminating between raw material sources.

Jurahornstein – The Hf/U, Nb/Pb, Rb/Zr, Zr/Hf ratios allow us to discriminate Jurahornstein samples (fig. 2). They present the further advantage of being relatively homogenous within a given source.

As they display the highest Hf/U, Nb/Pb, Rb/Zr ratios, the sources of Isteiner Klotz (JH IK 12 - West of the Black Forest) on one hand, and of Wittlingen (JHW 1.2 - Swabian Jura) on the other hand, can be clearly distinguished from the others (fig. 2a and b).

PTH 5.1/2 and JHS 3.1/2 show closer Hf/U and Nb/Pb ratios, being slighty heterogeneous within a given source (fig. 2a). However, the Zr/Hf vs Rb/Zr binary diagram (fig. 2b) displays greater ratio homogeneity for Schliengen samples (JHS 3.1 and 2 - West of the Black Forest) while emphasizing their close resemblance to Buxheim cherts (PTH 5.1 and 2 - Franconian Alb).

The Heidenheim samples (JHH 2.1/2 – Swabian Jura) offer the lowest Hf/U, Nb/Pb, Rb/Zr with variable chemical homogeneity.

These results highlight the possibility of distinguishing between the Jurassic outcrops of two given areas: the Swabian Jura on one hand and outcrops west of the Black Forest on the other hand.

Bohnerzhorstein – Sr/Ba and Pb/Th ratios allow us to distinguish markedly the three sources analyzed for Bohnerzhorstein (fig. 3) with Isteiner Klotz Bohnerzjaspis (BZJ-IK10 - West of the Black Forest) offering the highest values. Furthermore, the two samples from Oberriffingen (BZH 4.1/2 - Swabian Jura) have very similar ratios, showing the homogeneity of the source.

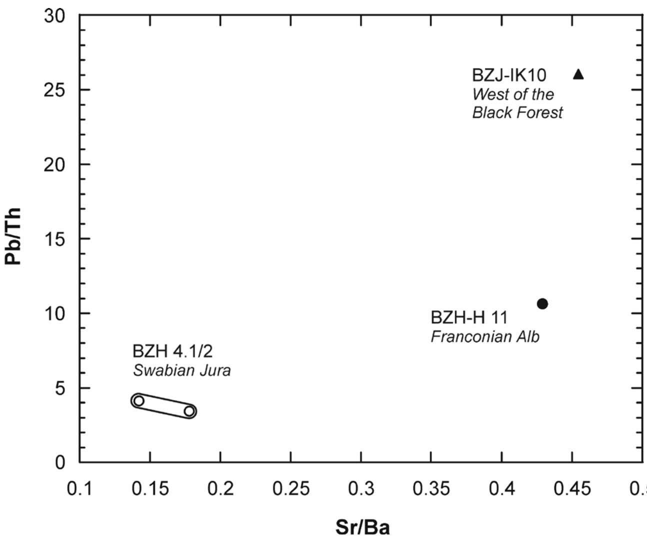

Figure 3. Binary diagram Sr/Ba vs Pb/Th for the Bohnerzhornstein analyzed.

Discussion and conclusion

This work provides a preliminary overview of Jurassic cherts from outcrops lying between the Rhine and the Bohemian massif, and is the first attempt for a geochemical characterization of Southwest Germany cherts. Further samples from each of the areas considered are needed to fully discuss the petrographic and chemical variability of these cherts.

Nevertheless, in the current state of progress, the combination of both geochemical and petrographic criteria allows us to distinguish each of the chert types taken into account in this study. Characterizations are mainly based on the presence or absence of microfossils (Foraminifers, Sponge spicules), REE abundances and some elemental ratios. In the case of Bohnerzhornstein, geochemical criteria seem to be more reliable than petrographic ones.

The perspectives for the further development of this study are two-fold. First, the petrographic examinations will be deepened on the basis of thin sections. In particular, we will aim to accurately identify fossil inclusions to species, for example in the Jurassic cherts occuring west of Black forest (Schliengen). Paleoenvironnemental conditions prevailing at the time of chert formation could be clarified in the process. Such information would be useful for establishing a geographical zonation of Jurassic chert facies, with direct applications in lithic artefact sourcing. Secondly, geochemical analyses are currently being complemented with Sr isotope analyses. This is a novel approach which has only been attempted on Paris Basin and Auvergne flints, where it provided interesting discriminating patterns and a better understanding of silicification processes (Poidevin 2002).

At last, the first geochemical data obtained on the Southwestern German sources in the framework of this study open new perspectives regarding flint sourcing. A set of archaeological samples has been analyzed to argue, on the basis of quantitative data, raw material long distance circulations during Upper Paleolithic.

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