

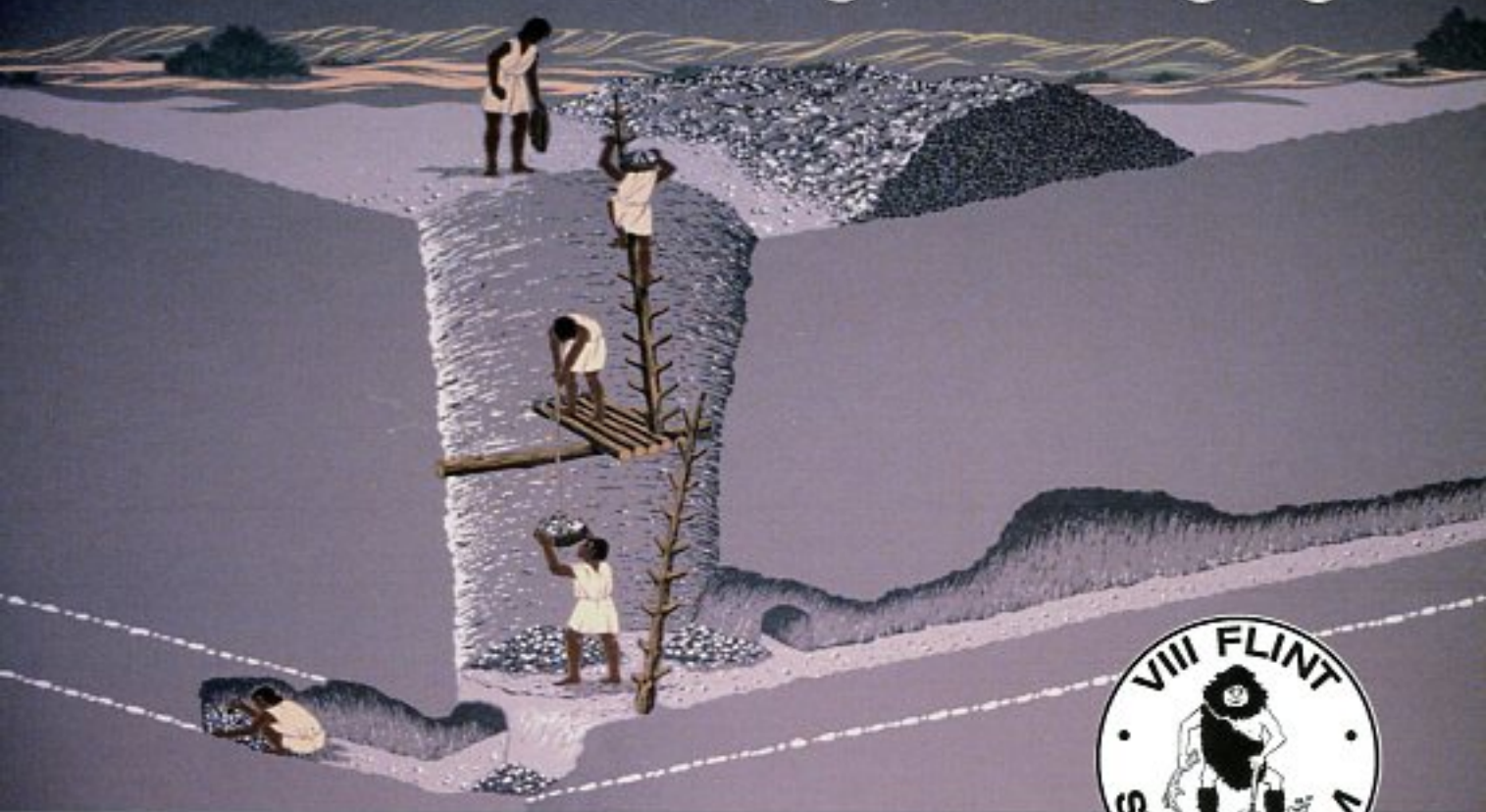
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Petit Spienes, B (Photograph G. Weisgerber)

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# Circulation of archaeological quartz artefacts in the Western Alps

## New evidences from fluid inclusions data

### Abstract

*In the Western Alps, both the Mesolithic and the Neolithic lithic industry is mainly based on flint, some fraction of the stone implements are made of quartz. As main potential quartz sources are distant from the archaeological sites where quartz artefacts have been found, anthropic transportation is needed. To fix the transportation roads implies the ability to fix the geographical provenience of a given quartz.*

*Chemical characterisation of quartz samples is very difficult as quartz has a very constant composition. But in nearly all quartz crystals there are fluid inclusions (FI), remnants of the fluid trapped in the growing crystal or in early sealed microcracks. Contrary to chemical composition, fluid inclusions content – viz. their density and their chemical composition – varies from one quartz to another, depending on the various geological environments during their formation.*

*Study of the fluid inclusions content is easily done using a heating/cooling microscope stage:*

- (i) cooling leads to an estimation of the fluid composition, through its melting temperature,*
- (ii) heating leads to an estimation of the density of the included fluid through its homogenisation temperature.*

*From this two-sided approach it should be possible to infer for each archaeological site the probable source of hyaline quartz. This assumption is validated by a case study in the Western Alps, which indicates that:*

- quartz fluid inclusions study is actually a discriminant tool as fluid inclusions characteristics vary from one site to another,*
- correlation between fluid inclusions data in quartz artefacts and in outcrops of quartz in the External Crystalline massifs tell us what were the source zones for the Epipalaeolithic to Neolithic sites.*

**Keywords:** *Hyaline quartz artefact, fluid inclusions, Western Alps, Epipalaeolithic, Neolithic.*

### Introduction

Throughout recent prehistory, quartz was used like flint, as raw material in many prehistoric sites. Sourcing archaeological quartz is a key problem, as this provenance is a source of precious information in the analysis of the provisioning strategies for each site studied.

Discrimination of archaeological quartz source(s) implies their characterisation, comparison between them, and ability to separate distinct groups for each provenance. As quartz has a constant and simple chemical formula ( $\text{SiO}_2$ ), it is difficult to discriminate the samples on usual chemical and/or mineralogical tests. Only trace elements pattern could change from one quartz to another, but there is a complete lack of such data on the quartz found in the possible source areas. But, quartz crystals often contain tiny fluid inclusions, remnants of the fluids surrounding the crystal when growing (see for example Roedder 1994, who gives many references on the use of fluid inclusions). As quartz crystals have grown from place to place in different P-T and chemical environments, reflected by different geological fluids, it should be possible to discriminate different quartz families and even have some rough idea of their provenance through the fossil fluids trapped in the quartz inclusions.

Although there are numerous methods possible to study fluid inclusions, the microthermometry appears to be the most suited for an exploratory work and methodological testing. Often used in geological studies (metallogenic studies for example), this method is rather inexpensive and easy to use (although fluid inclusions observation under the microscope is time consuming), and seems to have a sufficient precision to allow a typology of archaeological quartz of some prehistoric sites in the Western Alps. Nevertheless it should be noted that this method is destructive

Sites	Geographic location	Altitude	Site type	Periods	Samples number
1 Balme-de-Thuy (Haute Savoie-France) J.P.Ginestet	Bornes massif	620m	Rock-shelter	Middle Epipalaeolithic Middle Mesolithic Neolithic	433
2 La Fru (Saint Christophe-Savoie-France) G.Pion	Chartreuse massif	570m	Rock-shelter	Azilian Middle Mesolithic	26
2 Gerbaix (Saint Christophe-Savoie-France) G.Pion	Chartreuse massif	620m	Open air	Epipalaeolithic	23
3 Aulp-du-Seuil (Saint Bernard du Touvet-Isère-France) P.Bintz	Chartreuse massif	1700m	Rock-shelter	Middle Mesolithic Neolithic	137
4 Vарces (Isère-France) AFAN	Drac and Romanche Valley	310m	Open air	Middle Neolithic	138
5 Grande Rivoire (Sassenage-Isère-France) R.Picavet	Vercors	580m	Rock-shelter	Mesolithic	35
6 Comboire (Claix-Isère-France) R.Picavet	Drac and Romanche Valley	510 to 530m	Sepulture	Last Neolithic	6
7 Terres Blanches (Menglon-Drôme-France) H.Müller	Southern Prealps (Diois)	518 to 525m	Open air	Mesolithic	72
8 Clapier (Recoubreau-Drôme-France) A.Jourdan	Southern Prealps (Diois)	518 to 525m	Open air	Mesolithic Neolithic	66
9 Baume du Rif (La Motte Chalancon-Drôme-France) A.Jourdan	Southern Prealps (Diois)	900m	Rock-shelter and Open air	Cardial	18
10 Alpe Veglia (Alpi Lepontine- Italy) A.Ghiretti and P.Vavassori	Lombard Alps	1750m	Open air	Mesolithic	1055

as the observation of fluid inclusions under the microscope is done in polished thin-sections (thickness: 150 to 300 micrometers). Thus, only not too valuable artefacts could be used.

## Prehistoric sites

The principal characters of the sites where quartz was selected are described in Tab. 1 demonstrating that the presence of quartz artefacts is a common phenomenon from the Northern to the Southern Alps. We have selected the sites studied on several criteria:

- different ages from Epipalaeolithic to Neolithic,
- proximity with crystalline massifs, where possible quartz sources are numerous,
- different types of dwelling: rock-shelter and open air sites.

The diffusion of quartz shows a diachronic evolution (Tab. 1). Actually, it seems that it is not possible to attribute

at a given period in particular the choice of quartz as a raw material.

Quartz artefacts can come from several different geological environments: quartzites (rocks of sedimentary or metamorphic origin, formed almost exclusively of millimetric grains of quartz), or more often single milky or limpid quartz crystals, commonly found in the crystalline rocks, and even in sedimentary rocks (for example bi-pyramidal quartz of goodes in the slaty formations of the "Terres Noires", in the external French Alps). Only limpid quartz veins, or more rarely milky quartz, present fluid inclusions large enough (at least some tens micrometers) to be studied by the optical methods.

The use of fluid inclusions seems adapted in this case study as for all the sites quartz are limpid, little deformed and not recrystallised after their formation. But to be able to make a typology of quartz based on fluid inclusions, we need a database of the fluid inclusions quartz character-

ising possible source areas. From this point of view, the methodological test previously carried out (Cousseran *et al.* 1998) was done in a relatively favourable context:

- probable origin of quartz of the sites partially elucidated by other methods,
- former work in microthermometry FI having brought data on some of the supposed source areas (veins of the crystalline massifs of the External Alps: North to South, Mont-Blanc massif, Poty 1969; Rocheray massif, Ochoa Alencastre 1979; area of Bourg d'Oisans, Jenatton 1981, particularly with the quartz of the Gardette mine, studied by Poty 1969; North of the Pelvoux massif and Haute Romanche, Bernard 1978 and Nziengui 1993),
- existence of significant differences of the fluids observed between one area to another.

In our preliminary study, microthermometry analysis of the fluids included was done on about ten quartz samples for each site. In spite of this restricted sampling, it was possible to highlight the similarities or the differences of sources from one site to another, and to evidence the possible source areas.

## The fluid inclusions tool

### Principles of the method

In a rock crystallising or recrystallising in depth, beside the solid phases (minerals) one or more liquid phases ( $H_2O$ ,  $CO_2$ ...) exist in thermal and barometric equilibrium with these minerals. A small part of these fluids is trapped in the crystal (in the early growing defects or in later sealed microfractures). Remnants of those fluids are preserved as fluid inclusions which can form in the same crystal several distinct families: primary inclusions, formed at the time of crystal growing, or secondary inclusions in the various generations of microfractures.

To use a fluid inclusion as marker of the geological conditions during the formation of the crystal-host, we admit that its content was not modified since its trapping, either considering its chemical composition or its density. That implies that the inclusion behaved like a tiny micro-container of a constant volume. Generally it is admitted that if the mineral-host is quartz such an hypothesis is true.

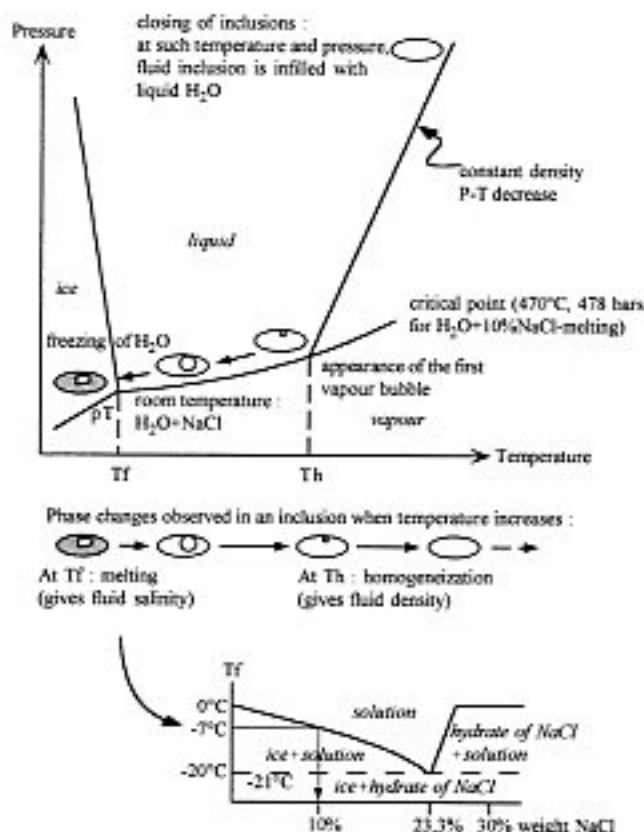
If so we easily can have a good idea of the fluid composition by cryometry (*cf.* chapter Cryometry). In addition, the constant fluid density implies that inclusion (or at least its content) follows, at the time of the exhumation of the crystal-host, a well defined P-T path (Fig. 1), corresponding to an isochor line of the P-V-T phase diagram of the included fluid (Roedder 1984; Weisbrod 1984). This property is exploited to find out the density of the trapped fluid (*cf.* Chapter Microthermometry) by thermometry.

Thus, study of composition and density of the included fluids can be used to discriminate fluid inclusion families, *i.e.* it leads to a typology of the quartz samples based on included fluids (distinction of quartz inside a same site or from one site to another). It can also help to characterise the formation areas, and thus to find sources of quartz artefacts.

### Cryometry

Cryometry is the measurement of the temperatures of phase change (melting) observed in an inclusion at low temperature. After cooling of the sample and freezing of the inclusions, we measure under the microscope the melting points of the various phases formed at low temperature (ice, clathrates,...): from these temperatures we can deduce the chemical components of the trapped fluids. For example, melting of a phase at approximately  $-56.6^\circ C$  indicates the presence of  $CO_2$  (the temperature of the triple point of  $CO_2$  is  $-56.6^\circ C$ ), melting of a phase at approximately  $-182.5^\circ C$  indicates the presence of  $CH_4$ . A  $H_2O + NaCl$  mixture melts at variable temperatures according to

Fig. 1: Fluid inclusion microthermometric study. Example of an inclusion filled with a  $H_2O + 10\text{ wt\% NaCl}$  brine. Top: P-V-T diagram, P-T conditions into the cooling inclusion (pT: triple point; pC: critical point). Below: phase changes observed when heating the inclusion ( $T_f$  melting temperature of last ice crystal,  $T_h$ , bubble disappearance; homogenisation temperature). Bottom: solution salinity determination, using the  $H_2O + NaCl$  system at low temperature. Values are according to Macflincor (Brown and Hageman program, 1994).



the abundance of salt in solution ( $-21^{\circ}\text{C}$  for a brine with 23.3% weight of NaCl, and  $0^{\circ}\text{C}$  for pure water, see Fig. 1). These temperatures could be lower than  $-21^{\circ}\text{C}$  if the brine contains salts of bivalent cations,  $\text{CaCl}_2$  for example.

#### Microthermometry

The microthermometry is based on the assumption that fluid inclusions keep a constant density (volumetric mass) since trapping of the fluid inclusions. Then, during exhumation, the P-T path followed by the fluid inclusion contents must be an isochoric path, whatever is the P-T path followed by the host-rock itself.

During cooling of the sample (Fig. 1), the exhumation path followed by the included fluid in the corresponding P-V-T system can be subdivided in two parts:

- (i) the included fluid first follows the isochor corresponding to its trapping pressure and temperature. During this step, no change of phase is observed.
- (ii) at the point where the isochor joins the ebullition curve, the fluid demixes in two phases (vapor and liquid). Then the vapor+liquid inclusion follows the ebullition curve, down to room temperature, where the two phases can be observed.

When heating the sample from room temperature the fluid follows the same path. That means, the measurement of the homogenisation temperatures gives the density of the fluid included. It corresponds to an isochor, on which must be the trapping P-T couple.

## Fluids in French Alps quartz artefacts

### Fluid inclusions data

#### Fusion temperatures and salinity

Melting points measured highlight significant differences in nature of the fluids observed in our samples. We have observed three types of quartz:

#### *H<sub>2</sub>O+NaCl bearing fluid inclusions:*

This type is encountered in the French Western Alps at sites like Balme-de-Thuy, Gerbaix-La Fru, Aulp-du-Seuil and Varcès, and in the French Southern Alps at sites like Terres-Blanches, Le Clapier and La Baume-du-Rif. Freezing usually occurs around  $-50^{\circ}\text{C}$ , while the measured melting points spread out from  $-4^{\circ}\text{C}$  to  $-16.5^{\circ}\text{C}$ . As we did not know by this method the exact composition of dissolved salt, the salt content is expressed in an "equivalent NaCl", because the NaCl is by far the more frequent natural salt (transformation of the melting point of salty water in % weight of NaCl, using the data for the system  $\text{H}_2\text{O}+\text{NaCl}$ , Potter *et al.* 1978; Bodnar & Vityk 1994). Rather low melt-

ing temperatures obtained (from  $-15^{\circ}\text{C}$  to  $-7^{\circ}\text{C}$ ) lead to rather high equivalent NaCl salinities, ranging from 10 to 18wt%.

#### *CO<sub>2</sub> or CH<sub>4</sub> bearing fluid inclusions:*

This family includes two types of quartz:

- (i)  $\text{CO}_2$  bearing fluid inclusions have three phases at room temperature. In each inclusion, we see  $\text{CO}_2$ -rich gas bubbles, liquid  $\text{CO}_2$  and liquid  $\text{H}_2\text{O}$ . The carbonic phase is nearly pure  $\text{CO}_2$  as the melting point of the  $\text{CO}_2$  phase is always very close to  $-56.6^{\circ}\text{C}$ . At low temperature a  $\text{CO}_2+\text{NaCl}$  hydrate is formed. It melts between  $+8.1^{\circ}\text{C}$  and  $+10^{\circ}\text{C}$  implying that the carbonaceous component in the fluid inclusions is nearly pure  $\text{CO}_2$  (pure  $\text{CO}_2$ +pure water hydrate has a melting temperature of  $+10.5^{\circ}\text{C}$ ; mT lower than  $+10^{\circ}\text{C}$  implies  $\text{CO}_2+\text{NaCl}+\text{H}_2\text{O}$  mixtures). The homogenisation of  $\text{CO}_2$  is done in liquid phase around  $+31^{\circ}\text{C}$  ( $\text{CO}_2$  critical temperature =  $+31.1^{\circ}\text{C}$ ).
- (ii) Some uniphased fluid inclusions do not freeze at a temperature as low as  $-80^{\circ}\text{C}$ . It could indicate the presence of  $\text{CH}_4$  or  $\text{N}_2$  but due to apparatus restrictions, we have not been able to determine their actual composition:  $\text{CH}_4$  melting occurs at  $-183^{\circ}\text{C}$ , and its freezing still lower down, due to surfusion phenomena, while the device we used can hardly reach less than  $-150^{\circ}\text{C}$ .

Such  $\text{CO}_2$  fluid inclusions or  $\text{CH}_4$  fluid inclusions have only been encountered in the Southern Alps, in the sites of Terres-Blanches and Alpe Veglia (in Italian Alps) for the first one, and in the sites of Terres-Blanches, Le Clapier and La Baume-du-Rif for the second one.

#### Temperatures of the homogenisation

Heating runs have been done only on the  $\text{H}_2\text{O}-\text{NaCl}$  fluid inclusions. Other fluid inclusions have not been used as (i) the  $\text{CO}_2+\text{H}_2\text{O}$  bulk homogenisation usually occurs higher than the maximum temperature allowed by the used device ( $300^{\circ}\text{C}$ ) and even at lower temperature, heating often leads to decrepitation of the inclusion (brittle failure of the inclusion walls due to heating related overpressure of the fluid), and (ii)  $\text{CH}_4$  or  $\text{N}_2$  fluid inclusions are already one phase inclusion at room temperature.

The homogenisation temperatures for the  $\text{H}_2\text{O}-\text{NaCl}$  bearing fluid inclusions in a site are represented as a "composite" histogram, corresponding to the summing up of several histograms, each obtained on a single quartz of the site. Considering these histograms, we can separate two cases:

- ♦ one characterised by a unimodal distribution of homogenisation temperatures (sites of Balme-de-Thuy, Aulp-du-Seuil, Le Clapier and La Baume-du-Rif),
- ♦ another one characterised by a plurimodal distribution of homogenisation temperatures (two or three peaks histograms: two peaks in sites of Terres-Blanches and three peaks in sites of Gerbaix-La Fru and Varcès).

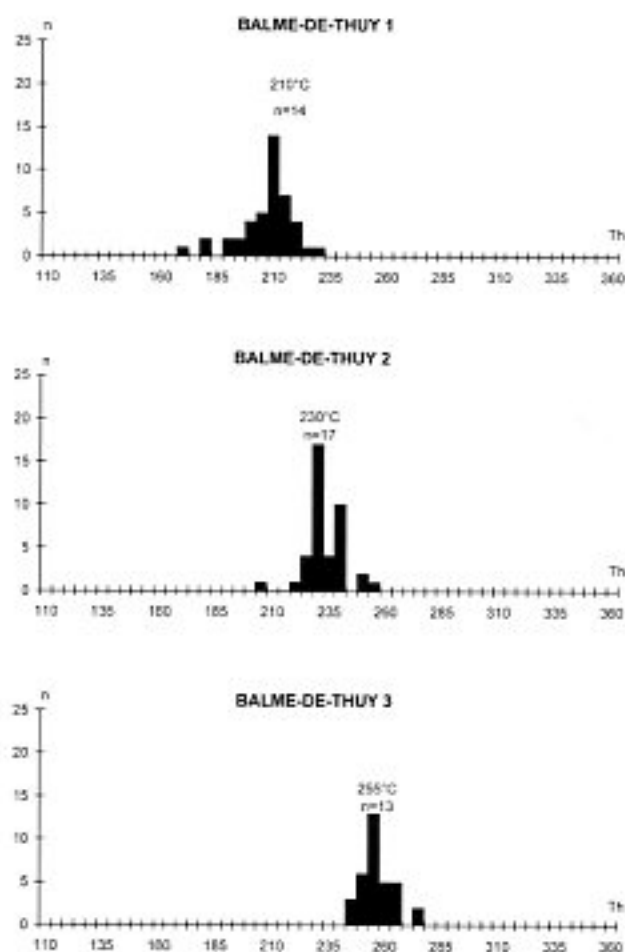


Fig. 2: Histogram of Th measured in the la Balme-de-Thuy quartz. Top: 66 fluid inclusions from two different quartz. Below: 100 fluid inclusions from four different quartz. Bottom: 36 fluid inclusions from two different quartz.

#### Site by site data

This two-sided approach (cryometry + thermometry) allows us to distinguish various families of quartz in several prehistoric sites in the Western Alps. From our data we can infer the following information for each site:

#### La Balme-de-Thuy (Haute-Savoie, France):

This site was excavated by J.-P. Ginestet (Ginestet 1984). Studied quartz artefacts were selected in layer 7A (Epipalaeolithic).

For the eight studied quartz artefacts, fluid inclusions have melting points spread out from  $-7.2^{\circ}\text{C}$  to  $-12.6^{\circ}\text{C}$ . These temperatures correspond to a salinity ranging from 13.5 to 16.2 wt% NaCl.

The Th histograms are unimodal but the temperature corresponding to the peak of the histogram can vary from one quartz to another. Studied quartz can be divided into three groups, each group being characterised by a distinct temperature, respectively  $210^{\circ}\text{C}$ ,  $230^{\circ}\text{C}$  and  $255^{\circ}\text{C}$ , with little recovery of the peaks of the histogram from one group to another (Fig. 2).

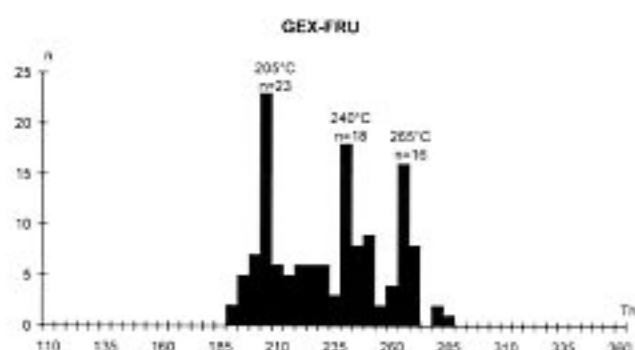


Fig. 3: Histogram of Th measured in the Gerbaix-La Fru quartz. 245 fluid inclusions to 8 different quartz. In each quartz, we can find unless 2 of 3 maxima seen on the histogram.

It can be inferred that the site of La Balme-de-Thuy has a lithic industry knapped of at least three families of different quartz. It can be three distinct veins may close from each other but which are probably not crosscutting (no fluid mixture).

#### Gerbaix and Fru (Savoie, France):

These sites were excavated by G. Pion (Pion 1990). Studied quartz artefacts were selected in the Mesolithic layer of the site of La Fru and in the Epipalaeolithic layer of the site of Gerbaix. These two sites of only few meters distance show the similarities of the measured ranges of Th which would imply a same geographical quartz source.

For the eight studied quartz objects, fluid inclusions have melting points spread out from  $-7^{\circ}\text{C}$  to  $-13.4^{\circ}\text{C}$ . These temperatures correspond to a salinity ranging from 13.5 to 16.7 wt% NaCl.

The single quartz histograms are more often bimodal, with a peak at low temperature (fluid with relatively high density), approximately  $20^{\circ}\text{C}$  lower than the peak at high temperature (fluid with relatively lower density). If we draw out the composite histogram for all the measurements of the site (Fig. 3), three modes arise clearly at  $205^{\circ}\text{C}$ ,  $240^{\circ}\text{C}$  and  $265^{\circ}\text{C}$ . But no Th hiatus is observed between the different samples: if we sort the quartz by increasing temperature peaks, each histogram for an individual thin-section has at least one common mode with the previous or next sample.

Thus, although the composite histogram suggests three different types of quartz we more probably deal with only one quartz type: the quartz of the site would come either from a same vein in which several generations have circulated, or from several veins crosscutting each other (*i.e.* possibility of fluid mixture) and having been excavated successively.

#### L'Aulp-du-Seuil (Isère, France):

This site was excavated by P. Bintz (1993). Studied quartz objects were selected in the layers C1 (Mesolithic-Neolithic transition) and C2 (Middle Mesolithic).

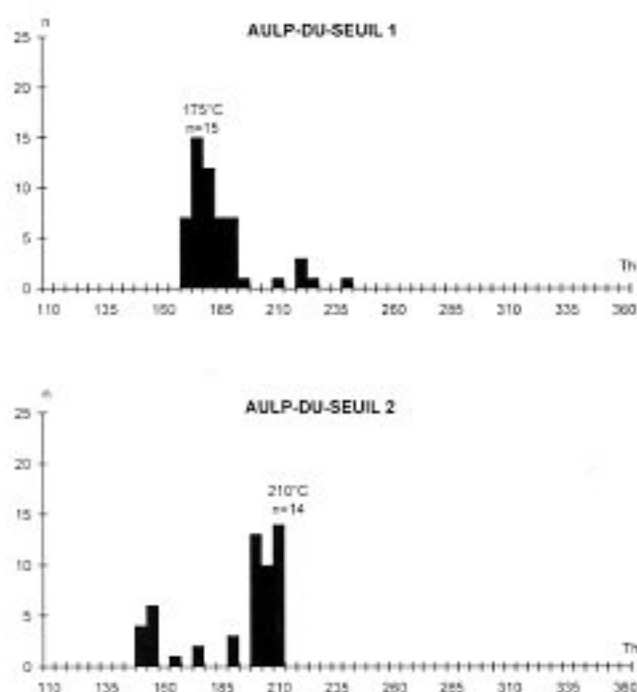


Fig. 4: Histogram of Th measured in the L'Aulp-du-Seuil quartz. Top: 58 fluid inclusions from four different quartz. Bottom: 52 fluid inclusions from four different quartz.

For the eight objects studied, fluid inclusions have melting points spread out from  $-5^{\circ}\text{C}$  to  $-10^{\circ}\text{C}$ . These temperatures correspond to a salinity ranging from 8 to 14.2 wt% NaCl. The Th histograms are unimodal but the temperature corresponding to the peak of the histogram can vary from one quartz to another. Studied quartz can be divided into two groups, each of them being characterised by a distinct peak temperature,  $175^{\circ}\text{C}$ , and  $210^{\circ}\text{C}$  respectively, with little recovery of the histogram from one group to another (Fig. 4).

We can infer, that at the L'Aulp-du-Seuil, the lithic industry is made with at least two families of different quartz.

#### Varces (Isère, France):

Located on the layout of the motorway A51, this site was excavated during rescue operations by the AFAN (Association des Fouilles Archéologiques Nationales) in 1994. For the twelve studied quartz objects, fluid inclusions have melting points spread out from  $-7.5^{\circ}\text{C}$  to  $-16.5^{\circ}\text{C}$ . These

Fig. 5: Histogram of Th measured in the Varces quartz. 339 fluid inclusions to 12 different quartz. In each quartz, we can find unless 2 of 3 maxima seen on the histogram.

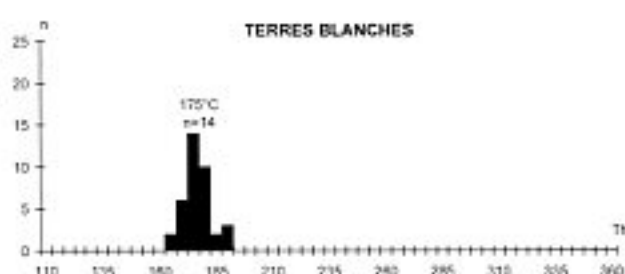
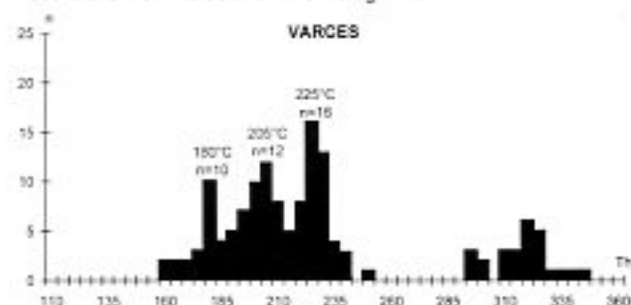


Fig. 6: Histogram of Th measured in the Terres-Blanches quartz. 49 fluid inclusions to two different quartz. In each quartz, we can find 1 maxima seen on the histogram.

temperatures correspond to a salinity ranging from 13.7 to 18 wt% NaCl.

Individual histograms (Fig. 5) are bimodal. From the composite histogram we can infer, as in the Gerbaix-La Fru sites, only one family of quartz and three main fluid circulation events, corresponding to Th around  $180^{\circ}\text{C}$ ,  $205^{\circ}\text{C}$  and  $225^{\circ}\text{C}$ .

#### Terres-Blanches (Drôme, France):

This site was excavated by H. Müller (Müller 1930; Henon 1990). Studied quartz artefacts were selected in the layers of the recent Mesolithic and the old and final Neolithic layers.

In eight studied quartz objects, we have observed three types of fluid inclusions:

- ◆ three samples in which all fluid inclusions have very low freezing temperatures (probable  $\text{CH}_4$  or  $\text{N}_2$  fluid inclusions). For these samples no cryometric neither thermometric data have been obtained.
- ◆ three samples have  $\text{CO}_2+\text{H}_2\text{O}$  fluid inclusions (melting points of  $\text{CO}_2$  at  $-56.6^{\circ}\text{C}$  and  $\text{CO}_2$  homogenisation temperatures are  $+31^{\circ}\text{C}$ ).
- ◆ two samples have  $\text{NaCl}+\text{H}_2\text{O}$  fluid inclusions (measurements of melting points spread out from  $-9.1^{\circ}\text{C}$  to  $-10.4^{\circ}\text{C}$ , corresponding to a salinity from 13.2 to 14.3 wt% NaCl).

The individual histograms are bimodal. When we draw the composite histogram (Fig. 6), we observe only one family of quartz, with two main fluid circulation events, corresponding to Th around  $175^{\circ}\text{C}$  and  $180^{\circ}\text{C}$ .

#### Le Clavier (Drôme, France):

This site was excavated by A. Jourdan and D. Orand (Beeching & Brochier 1994). Studied quartz samples were selected in the layers of the Mesolithic and the Neolithic. In the three studied quartz samples, we have observed two types of fluid inclusions:

- ◆ in one sample, all fluid inclusions have very low freezing temperatures (they probably are  $\text{CH}_4$  or  $\text{N}_2$  fluid inclusions). No cryometric neither thermometric data could be obtained from this sample.
- ◆ two samples have  $\text{NaCl}+\text{H}_2\text{O}$  fluid inclusions (melting points from  $-4^{\circ}\text{C}$  to  $-5.2^{\circ}\text{C}$ , corresponding to a salinity from 6.4 to 8 wt% NaCl).



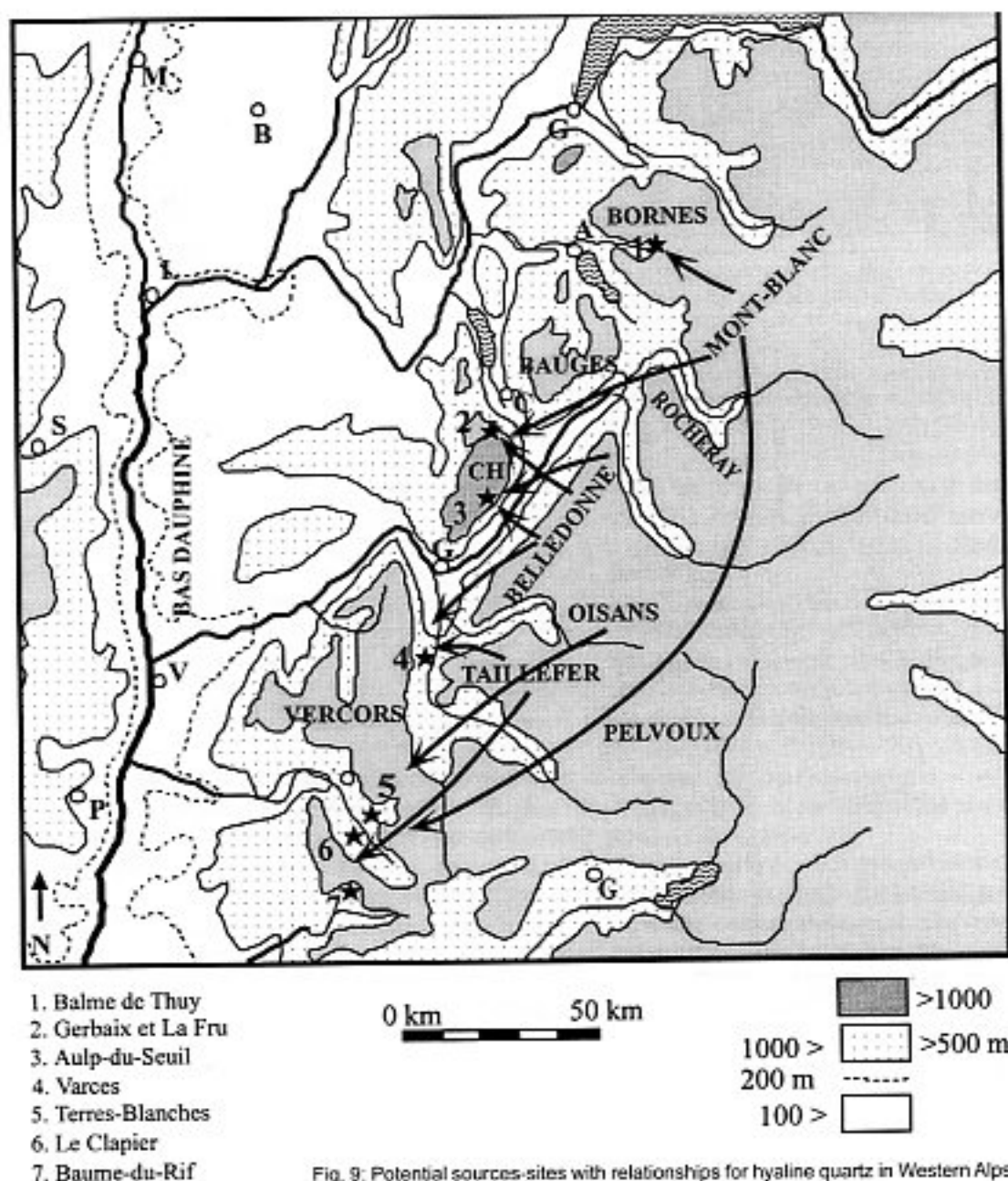


Fig. 9. Potential sources-sites with relationships for hyaline quartz in Western Alps.

line massifs data show a rather regular trend in average values of Th, which decrease from North to South, and/or fluid compositions more salty and  $\text{CO}_2$ -rich in the Northern massifs:

- in the Mont-Blanc massif, Poty (1969) measures in various veins values of Th gathered in the ranges 179–207°C, 183–230°C and 225–310°C. We can remark that the majority of quartz studied in the Mont-Blanc massif are smoky quartz and amethyst, and that the highest temperatures correspond to fluid inclusions filled by  $\text{CO}_2$  and  $\text{H}_2\text{O} + \text{NaCl}$ .
- in the Belledonne massif Ochoa (1979) describes in the quartz of the Rocheray massif (Maurienne valley) several families of inclusions with ranges of Th respectively at 159–175°C, 188–232°C and 297–358°C, for aqueous fluids with a salinity usually less than 10 eq. wt% NaCl.

- in the Oisans massif, near the Romanche river, there are several data from the North of Pelvoux massif (Bernard 1978; Nziengui 1993) to the Bourg d'Oisans basin (Poty 1969; Bernard 1978; Jenatton 1981). The results are very similar: the fluids included are NaCl poor (in general, less than 5 eq. wt% NaCl), the homogenisation temperatures are low in the North of Pelvoux and in the Oisans basin (110 to 150°C), higher close to the internal border of the Grandes-Rousses massif (until 185°C).

If we compare the values already known in the veins of the Alpine External Crystalline massifs and the values measured in the quartz from archaeological sites, we can deduce the most probable source areas and draw the potential sources-sites relationships for hyaline quartz in the French Western Alps (Fig. 9):

- ♦ for the site of La Balme-de-Thuy there is a good compatibility between the measured values and those observed in the Mont-Blanc massif. Complementary measurements are currently done on not smoky quartz of the Mont-Blanc veins to better validate our interpretation.
- ♦ for the sites of Gerbaix-La Fru and of L'Aulp-du-Seuil, although we cannot exclude the Mont-Blanc massif, proximity and compatibility of measurements from Belledonne suggest this last origin.
- ♦ for the site of Varcès we still notice temperatures compatible with those measured in the Mont-Blanc and Belledonne massifs. They are nevertheless lower than in the previous sites. Considering the decrease of temperatures noted in the crystalline massifs from North to South we could suggest a southernmost origin (quartz from the Oisans massif or Eastern border of the Taillefer massif). The values obtained in Oisans veins (Th < 185°C) are too low, therefore this area can be excluded. New data obtained on quartz from the Taillefer massif (Th from 160°C to 270°C) as well as the proximity of the massif allow us to consider this area as the most probable source for the site of Varcès.
- ♦ for the site of Terres-Blanches: the temperatures all are much lower than 185°C, fitting best with the Oisans massif data.
- ♦ for the site of Clapier quartz seems to have two distinct and relatively distant origins, both located in the External massifs: temperatures measured in Clapier 1 are similar to those found in the Taillefer massif while those measured in Clapier 2 are very similar to North Belledonne and the Mont-Blanc massif zone.
- ♦ for the site of Baume-du-Rif, again best accordance is with the Taillefer massif data.

## Conclusion

The fluid inclusions method is not yet a tool commonly used in the field of prehistory but has overcome the best phase (Cousseran *et al.* 1999) because:

- ♦ fluid inclusions of hyaline quartz from prehistoric sites in the Western Alps may be used in provenience studies. Using fluid inclusions, we have highlighted significant differences between the fluids of studied samples. We can (i) distinguish different quartz families from a site to another and (ii) characterise several quartz families inside the same site,
- ♦ it is now possible, using fluid inclusions to make reasonable assumptions which External Crystalline massifs could have provided hyaline quartz for artefacts found in Epipalaeolithic to Neolithic sites.

Even if fluid inclusions provide a good typology for the quartz artefacts the determination of their exact sources remains imprecise. The future study of fluid inclusions as typical markers should extend in two directions:

- ♦ the use of other methods to refine or supplement the analysis by FI. The analysis of the ions dissolved in the aqueous phase (for example by LA-ICP-MS) and the

quantitative and non destructive analysis of fluids (Raman spectrometry) will help to characterise the source areas in a more significant and discriminant way,

- ♦ an increase of the reference database for *in situ* hyaline quartz would allow a more accurate interpretation of the fluid inclusions data in archaeological quartz. Up to now we have used data restricted to the Northern French Alps. We must extend our database to all the Alpine arc (Suisse Valais, Northern and Southern French Alps and Lombardian Alps), either from published data or by new measurements in several more localities.

We will also apply these methods (i) to prehistoric sites whose sources are more difficult to determine as far away from the quartz formation areas, and (ii) to prehistoric sites where quartz has various fluid inclusions signatures. It will not only allow to better approach the role of quartz in the provisioning strategies of siliceous raw materials but also give an idea of quartz global circulation in all the Alpine arc.

Unfortunately our attempts failed to contact the authors for any proofs.

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