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Provenancing turquoise: a difficult task

De la difficulté d'établir la provenance des turquoises

Alain QUEFFELEC^a

Abstract: Turquoise is a blue-green phosphate gem used for millennia in the lapidary artwork of numerous past societies. It was one of the first gemstones, and has proved to be a valuable material that was part of long-distance exchange networks on every continent where it is available. Nonetheless, provenance studies for this material is absent from gemological literature, and scarce in the archaeological literature. This work reviews the mineralogy, geology, geographical and archaeological distribution of this gemstone, and presents different mineralogical, geochemical and spectroscopic approaches used in provenance studies. Sophisticated methods are needed to reconstruct the origin of turquoise and the commercial networks in the ancient times for this lapidary raw material, and we show that, even with these methods, a lot of work still remains to be done.

Résumé: La turquoise est un minéral phosphaté bleu-vert, utilisé pour l'art lapidaire depuis des millénaires, dans de nombreuses sociétés du passé. C'est une des premières gemmes utilisées, faisant partie de réseaux d'échange à longue distance sur tous les continents où elle est disponible. Cependant, les études de provenance de ce matériau sont curieusement absentes de la littérature gemmologique, et rares dans la littérature archéologique. Cet article résume ainsi la minéralogie, la géologie, la distribution géographique et archéologique de cette gemme, et présente les différentes approches minéralogiques, géochimiques et spectroscopiques utilisées pour en établir la provenance. L'utilisation de techniques de pointe est nécessaire pour identifier la provenance de ce matériau et comprendre les réseaux commerciaux des temps anciens. Nous montrons également que malgré l'application de ce type de méthodes, beaucoup de travail reste encore à faire.

Keywords: gemology, gitology, provenance studies, turquoise.

Mots clés : études de provenance, gemmologie, gitologie, turquoise.

INTRODUCTION

Turquoise, a hydrated phosphate of aluminum and copper with the ideal chemical formula $\text{CuAl}_6(\text{PO}_4)_4(\text{OH})_8 \cdot 4\text{H}_2\text{O}$, has been used as a gemstone for thousands of years and is still used today to create modern or traditional jewelry. It is indeed a very ancient material used for the production of jewelry, which has been used extensively in ancient times in Egypt, Persia, China, Mesoamerica, North America, South America and Europe (Beale, 1973; De'an, 2002; Domínguez-Bella & Herrero, 2012; Lowry & Lowry, 2010; Mathien, 2001; Pogue, 1915; Qin *et al.*, 2015; Ruppert, 1983; Ruppert, 1982). Today, it is also a fine stone still in use in artistic productions that maintain traditional styles (mainly from North America) or in contemporary creations.

There is a wealth of literature on the identification of the various simulants and treatments applied to this gem. Many simulants exist today but also in historical periods (Elwell, 1979; Lowry & Lowry, 2010). These include odontolith (Reiche *et al.*, 2000; Reiche *et al.*, 2001), Egyptian green pigment (Pagès-Camagna & Colinart, 2003), or the very ancient fake turquoise made in Syria about 5000 years ago, probably by heating apatite in the presence of iron and manganese (Taniguchi *et al.*, 2002). The identification of these different materials continues to be studied (Bernardino *et al.*, 2016; Reddy *et al.*, 2006; Sabbaghi, 2018). The enhancement of the mechanical and visual properties of this gem by various methods is also widespread and widely studied in gemology (Choudhary, 2010; Fritsch *et al.*, 1999; McClure & Smith, 2000). While the identification

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of simulating materials and treatments is topical in gemology, this is not the case for studies of origin, which, on the contrary, are non-existent to our knowledge with regard to turquoise extracted and sold today. The absence of any such study is probably due to the fact that the question does not arise: the price of this gem is not high enough to stimulate such studies, and provenance is not a fundamental criterion in the attribution of the value of this gem. For older periods, on the other hand, many studies exist on the origin of turquoise, which is of great interest to archaeologists. Indeed, this material is involved in more or less long-distance exchange networks in many cultures of the past, and is therefore one of the tools for understanding the diffusion of cultural goods, communication between groups, the installation or disappearance of exchange networks, etc. Many studies concern Mesoamerica, North and South America, where the use of turquoise by pre-Columbian populations is very important (Ball, 1941; Pogue, 1915; Ruppert, 1983; Ruppert, 1982).

This work first summarizes the mineralogical and geological knowledge about this mineral, and then presents methods attested in the scientific literature for establishing the origin of this gem material. Ball (1941) wrote about turquoise: “Since many rocks and certain minerals have characteristics that indicate their source, petrographic studies of these artifacts could largely illuminate the supply circuits of the former Americans”. It is much easier to write it, than to realize it.

1. MINERALOGY

Turquoise is a hydrated phosphate of aluminum and copper, whose ideal composition is represented by the formula $\text{CuAl}_6(\text{PO}_4)_4(\text{OH})_8 \cdot 4\text{H}_2\text{O}$. This mineral crystallizes in the triclinic system, most often in the form of massive aggregates, forming botryoid-shaped nodules. The term turquoise is also the name of the mineral group with the general formula $\text{A}_{0-1}\text{B}_6(\text{PO}_4)_{4-x}(\text{PO}_3\text{OH})_x(\text{OH})_8 \cdot 4\text{H}_2\text{O}$, where the octahedral site A may be occupied by bivalent ions such as Cu^{2+} , Fe^{2+} or Zn^{2+} , but which can also be empty or partly empty (x may thus vary between 0 and 2). The tetrahedral site B is occupied by trivalent ions such as Al^{3+} or Fe^{3+} (Foord & Taggart, 1998) (Table 1). The crystal structure of turquoise is described by Kolitsch & Giester (2000).

It is noteworthy that, since the redefinition of the entire mineral group by Foord & Taggart (1998), the turquoise name is now restricted to the copper endmember of the Turquoise-Planerite solid solution only, in which octahedral sites may not be entirely filled with copper. Thus, the majority

Site A	Site B	$(\text{PO}_4)_{4-x}$	$(\text{PO}_3\text{OH})_x$	Mineral
Empty	Al^{3+}	2	2	Planerite
Cu^{2+}	Al^{3+}	4	0	Turquoise
Zn^{2+}	Al^{3+}	4	0	Faustite
Fe^{2+}	Al^{3+}	4	0	Aheylite
$\text{Ca}^{2+}?$	Al^{3+}	4	0	Coeruleolactite
Empty	Fe^{3+}	2	2	Unknown
Cu^{2+}	Fe^{3+}	4	0	Chalcosiderite
Zn^{2+}	Fe^{3+}	4	0	Unknown
Fe^{2+}	Fe^{3+}	4	0	Not named
$\text{Ca}^{2+}?$	Fe^{3+}	4	0	Unknown

Table 1: Minerals (endmembers) of the turquoise group, modified after Foord & Taggart (1998).

Tableau 1 : Minéraux du groupe de la turquoise, modifié d'après Foord & Taggart (1998).

of gems sold under the name of turquoise, like archaeological objects, are found to be mineralogically related to planerite, since most of them contains less copper than they should to be called turquoise in a very strict definition. This study will continue to use the term turquoise, however, as recommended by Foord & Taggart (1998) and Fritsch *et al.* (2019).

2. GITOLGY

Beyond the mineralogy of the gem, the question of the origin of the turquoise requires to study the gitology of this mineral. It is most often found in the form of veins or lenses/nodules, in fracture fillings or in faults (Chen *et al.*, 2012; Kievlenko, 2003; Lijian *et al.*, 1998; Moro Benito *et al.*, 1995). A hydrothermal or supergenic origin of turquoise has long been debated, the consensus largely tends towards the supergenic formation (Bergen *et al.*, 2007; Hull & Fayek, 2012; John *et al.*, 2010; Kievlenko, 2003; Thibodeau *et al.*, 2015). In general, the interaction between Al-silicate and phosphate-rich rocks containing sulphidic copper minerals with meteoric water is a precondition for the crystallization of turquoise.

This mineral is mainly found in environments connected to porphyry copper ore deposits (Ruppert, 1983; Singer *et al.*, 2002; Thibodeau *et al.*, 2015), allowing meteoric water to be enriched with copper. These porphyry copper deposits are often located on the top of plutons, formerly altered by hydrothermal solutions enriched in heavy metals and sulphide ions during the crystallization of the pluton (Salze *et al.*; Sillitoe,

2010) (fig. 1). In veins and fractures of this altered zone and the surrounding rocks, metallic elements such as copper, iron, zinc, cadmium, lead, as well as arsenic form sulphides (John *et al.*, 2010). During uplifting close to the surface, meteoric water may oxidize the sulphidic minerals, producing dissolved metal ions and sulphuric acid. The acid may dissolve hosting rock's Al-silicate and phosphates, mainly apatite or phosphorite (Kievlenko, 2003; Thibodeau *et al.*, 2015). If enough copper, phosphate and aluminium are present in solution, they may coprecipitate as turquoise in areas where the pH of the solution increases. During these weathering reactions, other elements released from the sulphides as mentioned above can be integrated turquoise during precipitation.

Porphyry copper deposits represent in most cases the parental material for turquoise, as is the case for the mines in the southwestern United States and Mexico, in the Andes, in Central Asia, in Armenia, and Mongolia (Kievlenko, 2003; Ruppert, 1983). In France, another source of turquoise was identified in the Montebrias area, where it was found in a mine dug in a granite dome overlooking a rare element microgranite rich in tin, tungsten, lithium and a little copper (Patureau *et al.*, 2011). In Bulgaria, turquoise was found in a Pb-Zn ore field (Kostov, 2010).

Rarer occurrences of turquoise exist in other geological contexts. The turquoise from Sinai in Egypt (Wadi Maghara & Sarabit al-Khadim) (Khazeni, 2014; Megahed, 2019; Shalaby, 2015) originated from copper leaching during

the soil alteration of Carboniferous sedimentary rocks and subsequent crystallization of copper minerals in soil horizons and the underlying karst (El Sharkawi *et al.*, 1990). However, this is the only occurrence attributed to this mode of crystallization to our knowledge. O'Donoghue (2006), however, indicates without citing a source that the origin of copper in Egypt is connected to porphyry ore deposits, but no publication describes such deposits for Egypt and Sinai (John *et al.*, 2010; Sillitoe, 2010; Singer *et al.*, 2005). In the western Hubei province and southeastern Shaanxi province in China, meteoric waters are believed to have dissolved copper sulphides and phosphate minerals from siliceous sedimentary shales, and allowed turquoise to crystallize in fractures (Lijian *et al.*, 1998; Qin *et al.*, 2015). In eastern Anhui province, the Maanshan turquoise is found in a altered area of Cretaceous andesitic volcanic tuff (Wei & Guan, 2003, cited by Qin *et al.* 2015).

3. DISTRIBUTION OF TURQUOISE

Geographical distribution

As mentioned, the mines of turquoise are almost all related to the presence of porphyry copper deposits, whose distribution is very well studied because of major economic interests in the production of copper (fig. 2). However,

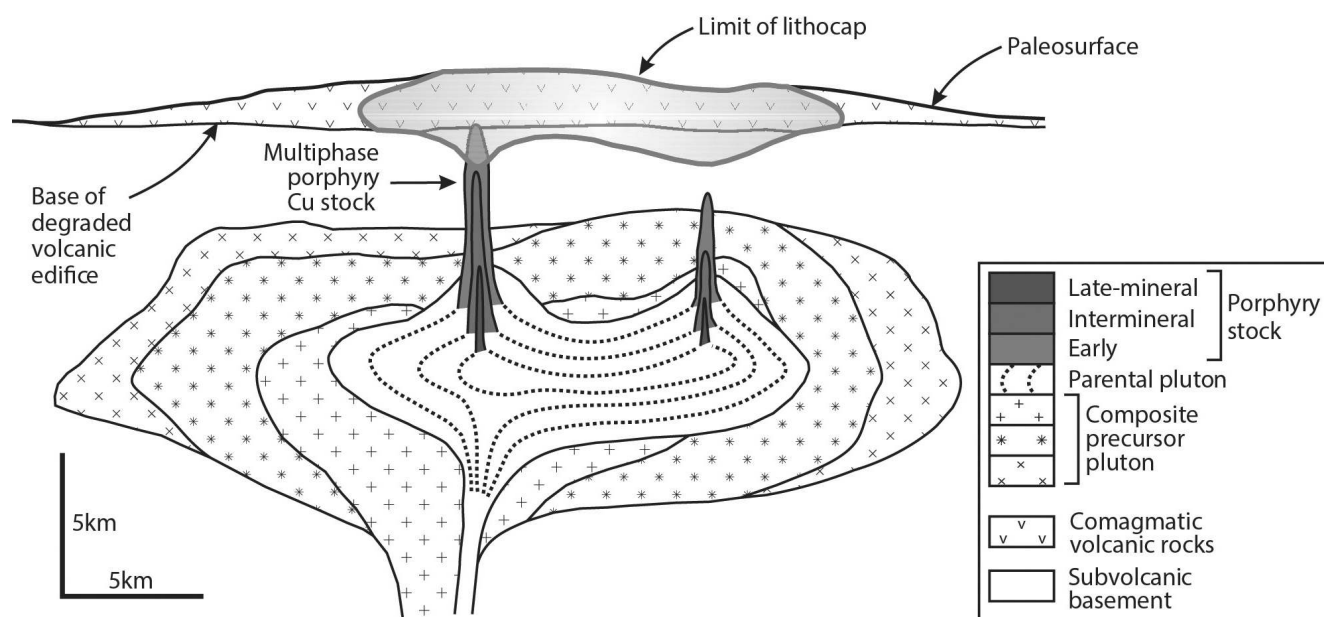


Figure 1: (Voir planche couleur III) General diagram of the formation of a porphyry copper system (modified after Sillitoe, 2010).

Figure 1 : (See colour plate III) Schéma général de la formation d'un porphyre cuprifère (modifié d'après Sillitoe, 2010).

only a few deposits contain turquoise. Two main reasons can be invoked to explain this rarity. First, phosphate-rich hosting rocks are necessary. Second, turquoise is only preserved in arid and semi-arid areas, but weathers in humid areas over the course of time (Ruppert, 1983). The presence of turquoise is also not systematically recognized or published. In that respect, turquoise survey and searching for potential sources must not only rely on published data but also on geological information. Thus, in its list of copper deposits, Singer (2008) lists only 24 localities with turquoise, and these localities are not necessarily exploitable for turquoise, because they may contain only traces of this mineral. Among these 24 localities, a large number (16) are located in the Rocky Mountains and Andes of America (John *et al.*, 2010; Sillitoe, 2010; Singer *et al.*, 2005). These turquoise deposits include those of the USA, Mexico, Peru, Chile and Argentina. There are far more turquoise mines than listed in copper porphyry studies (e.g. Hull & Fayek, 2012; Kievlenko, 2003; Lowry & Lowry, 2010; Pogue, 1915; Shigley *et al.*, 2000; Thibodeau *et al.*, 2015). Some sources of American turquoise originate outside of this area, such as in an area of Brazil (Costa *et al.*, 2004; Shigley *et al.*, 2010), where copper porphyry deposits are also recorded (fig. 2). There are also turquoise sources in the eastern United States (Barwood, 1997). In Asia, there are exploitable turquoise deposits in China (Keller & Fuquan,

1986), Mongolia (Kievlenko, 2003), Tibet (Laufer, 1913; Pogue, 1915; Watson, 1983), Malaysia (Murthy, 1989), as well as in several Central Asian countries such as Iran, but also Turkmenistan, Uzbekistan, Armenia or Tajikistan (e.g. Kievlenko, 2003; Momenzadeh, 2004; Singer *et al.*, 2005). The turquoise of the Sinai peninsula is well known archaeologically, but is no longer exploitable today. There are also turquoise deposits in Bulgaria (Kostov, 2010), France (Patureau *et al.*, 2011), Germany (Pogue, 1915) and Spain (Moro Benito *et al.*, 1995). Several sources are known in Australia (Kievlenko, 2003; Pogue, 1915). The presence of turquoise in Tanzania (Zwaan & Zwaan, 1974), Guatemala and Poland should also be mentioned.

Archaeological distribution

The interest to establish the source of turquoise in archaeological context is very strong, and useful in light of the wide use of this material in the past, both spatially and temporally. Turquoise beads are found in many cultures of the past, on almost every continent, going back up to 12,000 years in the past. The earliest occurrence of turquoise beads has been found in Israel, in the site of Gilgal II, and is dated to the final Natufian, between 10,800 and 9,500 B.C. (Bar-Yosef Mayer & Porat, 2008). The aim here is not to list all the cultures of the past that have used turquoise

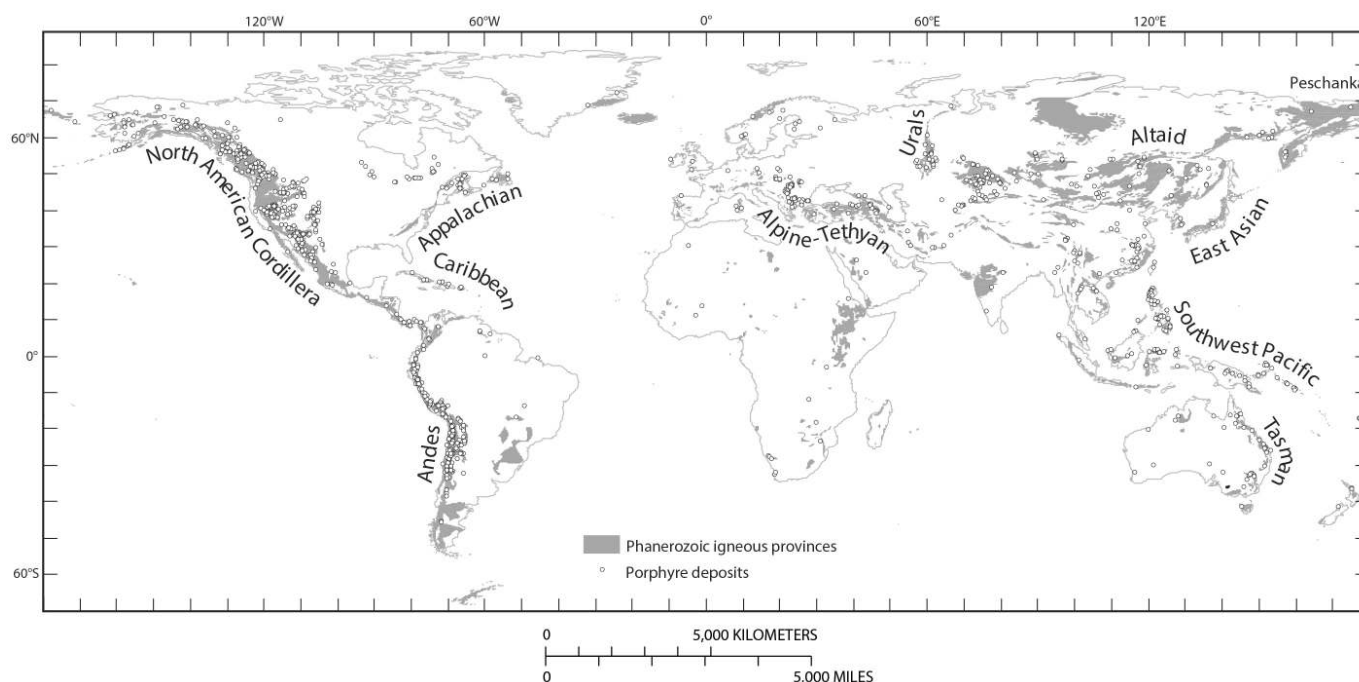


Figure 2: Global occurrence of the copper porphyry deposits (modified after John *et al.*, 2010).

Figure 2 : Répartition mondiale des dépôts de porphyres cuprifères (modifié d'après John *et al.* 2010).

as a material for making ornamental objects, but to show the diversity of this use. Its utilization can then be noted in the whole New World: North America (e.g. Barwood, 1997; Mathien, 2001; Pogue, 1915; Ruppert, 1982), Central America (Harbottle & Weigand, 1992; Ruppert, 1982; Thibodeau *et al.*, 2018; Weigand *et al.*, 1977), the Caribbean (e.g. Narganes Storde, 1995; Queffelec *et al.*, 2018, 2020a), and South America (e.g. Dominguez-Bella & Sampietro Vattuone, 2005; López *et al.*, 2018; Ruppert, 1982; Sampietro Vattuone *et al.*, 2017). It is also widespread in the Old World, as it is well known from archaeological contexts and mines in Egypt (Kobusiewicz *et al.*, 2004; Megahed, 2019; Shalaby, 2015), the Near East (Alarashi, 2016), the Middle East (e.g. Beale, 1973; Momenzadeh, 2004), China (e.g. De'an, 2002; Qin *et al.*, 2015; So, 2018; Wiesheu, 2013), and India (Biswas, 1994). In Europe, turquoise is known as archaeological artifacts (e.g. Errera, 1999; Herbaut & Querré, 2004; Kostov, 2010) as well as in several ancient mines (Odriozola & Villalobos-García, 2015; Villalobos García & Odriozola, 2016).

4. PROVENANCE IDENTIFICATION

As we have seen previously, turquoise sources are generally few in number and restricted to special geographical areas, but nonetheless this gem has been very important in many cultures of the past. A better knowledge of the precise origins of this rare material would help to elucidate the trading networks of these societies and their interconnections. However, despite numerous archaeological or geological studies on turquoise in areas where it can be found naturally, in many areas of the globe provenance studies are lacking. The studies mentioned above in this manuscript describe the presence of turquoise mines and turquoise objects, but do not include any archaeometric data, whether in Egypt, Iran or Bulgaria for example (Beale, 1973; Kostov, 2010; Megahed, 2019). Only the geographical proximity of the source or the fact of a known single source serves as an argument for the origin. It is also conceivable that the lack of the scientific literature on this issue for these regions can be caused by a publication bias for publications with negative results.

Colour

Historically, the identification of turquoise mostly has been based on the color of the finished object. The finished objects, often polished or at least shaped to make an ornamental object, are sometimes difficult to identify because the absence of characteristic petrographic criteria can make

a mineralogical identification difficult. Mathien (2000) explains that since color can change within a vein itself, with the exposure and the use of stone, this characteristic is of little help in the study of archaeological turquoise. Nevertheless, he mentions a provenance study based solely on color, in a study of the early 20th century by Pepper (1909). However, when reading this article more carefully, it becomes clear that Pepper does not speak at all about color to identify the source of the thousands of pearls and pendants from the archaeological site of Pueblo Bonito (New Mexico, USA), but about the petrographic association, the size of the turquoise nodules and the fact that this mine was intensively exploited during prehistoric times. It therefore seems that the color criterion has never actually been used for a serious attempt to determine the origin of a turquoise, but has been used in the past to identify, sometimes wrongly, the material.

Mineralogy and petrography

The mineralogical/petrographical investigations on turquoise objects can be an interesting way to identify the origin of turquoise. First of all, there can be a high variability of different minerals included into turquoise. For example, the silicon content in a turquoise analysis may be related to different minerals, different silicates, including phyllosilicates as well as amorphous silica, opal, or chalcedony (Foord & Taggart, 1998; Kievlenko, 2003). The water content measured in the analyses, which also varies, may come from other minerals, which are more or less hydrated (Foord & Taggart, 1998; Kievlenko, 2003). These minerals integrated into the turquoise masses can be syngenetic during the gem formation, or may come from its alteration. One provenance study in particular identified a very likely provenance of an archaeological turquoise object from a site in Arizona due to the presence of the very rare mineral metatornite, which can be related to turquoise from a nearby mine in the Canyon Creek area (Welch & Triadan, 1991). This study shows the potential of detailed systematic mineralogical investigations to determine the provenance of turquoise objects. However, this requires destructive analysis or non-destructive analysis quite difficult to carry out on large quantities of objects (X-ray microdiffraction for example). Turquoise being opaque, actually, it does not allow for an easy observation of inclusions as it can be done for translucent gems.

Beyond inclusions in turquoise, it is important to note that many archaeological turquoise objects, particularly the most imposing, are not entirely made of this mineral (Pepper, 1909; Qin *et al.*, 2015). Turquoise may be veined with other well expressed minerals (often sulphides or chrysocolla), or attached to its surrounding rock, allowing to

refine its origin, if the potential sources have distinct petrographies as is the case in China (Qin *et al.*, 2015). The size of turquoise in a deposit is another criterion that can provide evidence for its origin. Indeed, millimeter-sized pearls can come from any deposit, but a several centimeters long and several tens of millimeters thick pendant cannot come from a deposit with only small veins of turquoise.

Geochemistry

The chemical composition of turquoise is complex. This mineral may include elements available during its crystallization in the fractures, faults, cavities of the host rock. This incorporation is found for both major elements (the turquoise group presents several solid solutions [Foord & Taggart, 1998]) and trace elements fitting into the structure. These elements were coprecipitated together with copper, aluminum and phosphate during the crystallization from fluids. The composition of the turquoise depends therefore on the composition of the ore and host rock minerals, pH conditions, the availability of water etc. and can be specific for different mines (e.g. Ruppert, 1983).

Major, trace and rare earth elements (REE)

Most geochemical studies integrate the quantification of major and trace elements. This type of analysis, based on various methods applied over decades depend on laboratory equipment of the researchers (electron microprobe, arc emission spectrometry, neutron activation, PIXE, X-ray fluorescence, mass spectrometers), and the application of univariate or multivariate statistical methods for data evaluation. This kind of studies are currently often considered non-functional in general. However, most authors are only interested in North America and it is not always clear in their writings that this assertion only concerns this continent (Hull *et al.*, 2008; Thibodeau *et al.*, 2018; Thibodeau *et al.*, 2015). For North America in particular, there is only one study that seems to have found positive results in the attribution of a source of turquoise for archaeological artifacts (Sigleo, 1975; Sigleo, 1970). Sigleo (1975) establish the provenance of 13 turquoise beads from the Snaketown site to the mines of Himalaya (California). This result was supported by very low concentration of trace elements in both localities, and, moreover, on similarities coefficients comparing the archaeological artifacts and 24 mines, from which only Himalaya was recognized similar enough. The publications of Weigand and Harbottle about turquoise exchange between the pre-Columbian cultures of Southwestern US and Mexico are widely cited, but, despite a large number of analyses over many years of work, almost no chemical data are published

(Harbottle & Weigand, 1992; Weigand *et al.*, 1977). More recent studies concede that conclusions concerning the turquoise sources are difficult to draw and that mainly inter-site comparisons can be made (Mathien, 2000; Thibodeau *et al.*, 2015). Finally, Kim *et al.* (2003) used PIXE to identify turquoise within green minerals, but their study compares only two archaeological sites with each other, without analyzing sources. To summarize, all the studies on turquoise sourcing in North America, except from Sigleo (1975), came to the conclusion that it is very difficult to determine the origin of North American turquoise archaeological objects (Hull *et al.*, 2008; Mathien, 2000; Ruppert, 1983).

Unfortunately, there are only a few studies on other continents. Beyond the very recent and very localized work in Argentina (López *et al.*, 2018), there is only Ruppert's enormous work (Ruppert, 1982; Ruppert, 1983) concerning Meso- and South America, and some work in China (Qin *et al.*, 2015). In the work of Lopez *et al.* (2018) in Northwestern Argentina, unfortunately, the composition of the turquoise objects found in the archaeological excavation are hardly to compare with the chemical analysis of the turquoise from the wall of the same excavated pre-Columbian mining cave. Only two turquoise samples for the walls and five archaeological samples from the excavation were analyzed by EDS analysis with an electron microscope. The two wall samples contain titanium, unlike the archaeological samples. Three archaeological samples have chlorine, but there is none in the wall samples. The archaeological samples are all richer in phosphorus compared to the wall samples, and all lower in potassium. In short, there is almost no correspondence between the two sample categories. In the work of Qin *et al.* (2015), which is strongly focused on REE, some results from previous turquoise studies in China (and published in Chinese) can be read (He *et al.*, 2011; Mao *et al.*, 2005; Wang *et al.*, 2007). These studies indicate compositional differences between different sources based on trace element analysis. Qin *et al.* (2015) show very clear differences of the REE patterns in turquoise from the Hubei-Shaanxi sources and the Ma'anshan source (fig. 3). The first group is characterized by relatively low REE concentrations, a fractionation between light and heavy REE (LREE and HREE respectively), and two clearly visible anomalies: a negative one in Ce, and a positive one in Eu. On the contrary, the more easterly source of Ma'anshan contains more REE and shows a significant fractionation between LREE and HREE, no Ce anomaly and a negative Eu anomaly. The analysis of REE in turquoise from two archaeological sites shows a clear similarity with local sources for one of them (fig. 3h): the patterns of samples Djl1 and Hqg2 are strongly overlapping. Other archaeological sites provided turquoise objects with a spectrum of REE showing very few ano-

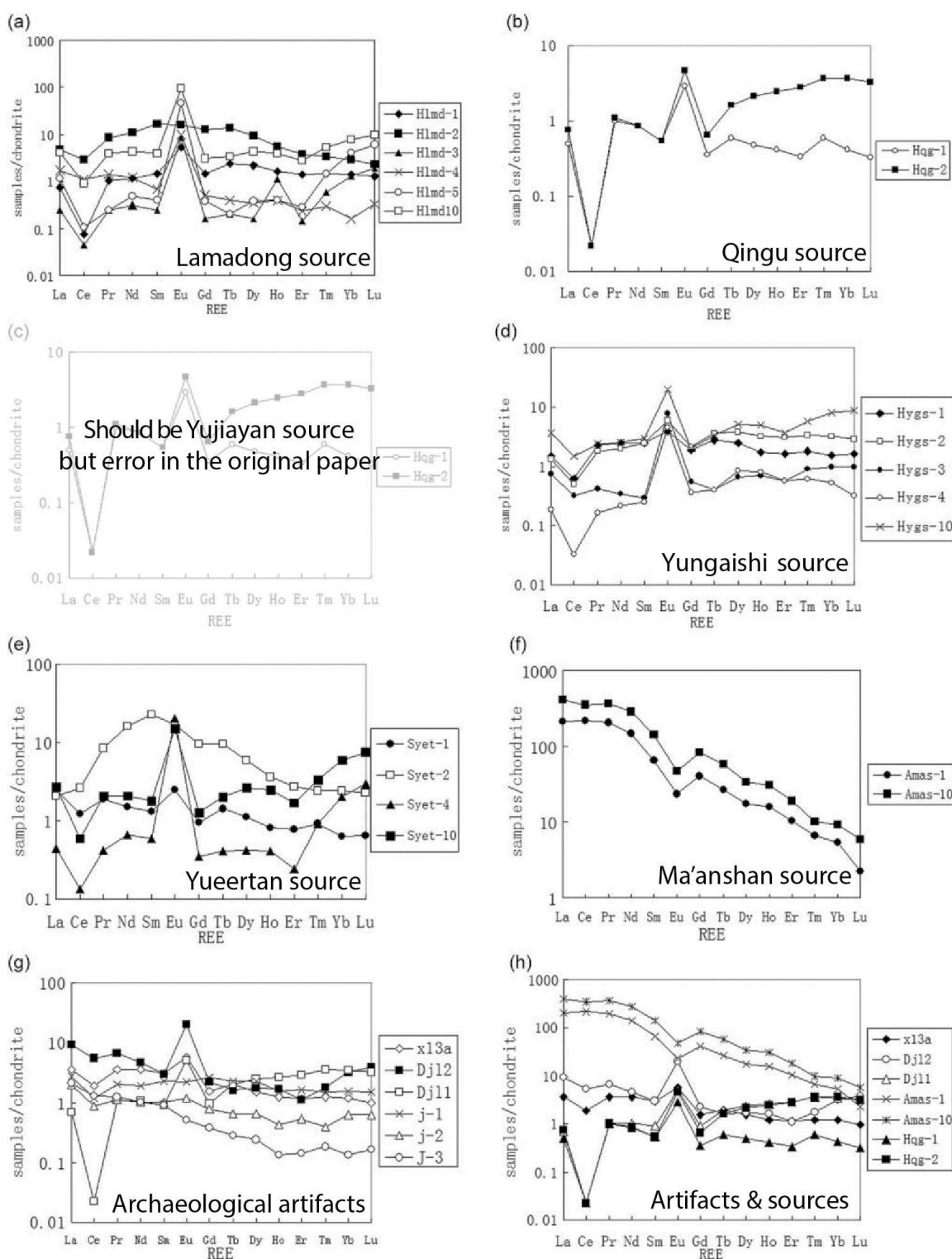


Figure 3: REE analysis results for several sources and some archaeological artifacts. Figure (c) in the original paper is unfortunately an error in the original article, since it is the same as sub-figure (b). Modified after Qin *et al.* (2015).

Figure 3 : Résultats d'analyses des éléments de terres rares pour quelques sources potentielles et quelques objets archéologiques. La sous-figure (c) dans l'article original est une copie malencontreuse de la sous-figure (b). Modifié d'après Qin *et al.* (2015).

malies (d-1, d-2 and d-3), which were not identified in any of the sources analyzed. The difference between the two sources can also be asserted for other trace elements, in particular uranium, which is much more concentrated in the Hubei-Shaanxi samples than in Ma'anshan. However, it is clearly stated in the study of Qin *et al.* (2015) that establishing precise sources is difficult with this method. Nearby sources of turquoise of similar geological origin have a similar signature.

Let us finish with the gigantic study of Ruppert (1982, 1983), who analyzed by electron microprobe about 1,500 samples from deposits and archaeological sites in the Americas. The statistical analysis of the results, based on clustering methods, revealed some results. First, significant differences are shown between North American and South American turquoise, based in particular on chromium and arsenic content. In South America, unlike North America, many of its results can be interpreted. Some samples of some cultures of the past, in limited chronological periods, are very similar, and are different from the sources currently known. Some samples from different archaeological periods are also geochemically distinct from each other, thus leading to the interpretation of a change in supply over time. Other archaeological turquoises, on the other hand, can be linked to known sources or regions, again allowing a very interesting discussion on the exchange networks of these pre-colonial populations. Although we were able to obtain both of Ruppert's articles from the author himself, it was unfortunately not possible to extract the numbers from them in order to attempt new statistical analyses as part of this paper. It is likely that these exceptional data, however, would deserve further consideration, as would the very large number of samples if they still exist.

Isotopes

Isotope geochemistry is a method applied to turquoise in North and Mesoamerica to overcome the limits of conventional geochemistry (Hull *et al.*, 2014; Hull *et al.*, 2008; Othmane *et al.*, 2015; Thibodeau *et al.*, 2018; Thibodeau *et al.*, 2015). Pioneering studies were performed by Young (Young, 1995; Young *et al.*, 1997, 1994 in Mathien, 2000). These studies were presented as student report and on two conference presentations. They are summarized by Mathien (2000) who is one of the co-authors at the conferences: "In a preliminary evaluation using stable lead isotope ratios derived from 26 samples from seven mining districts in the southwestern United States and northern Mexico (most from Cerrillos, New Mexico), Suzanne Young was able to separate the Cerrillos mines from all others using a ratio of $^{208}\text{Pb}/^{207}\text{Pb}$ (Young *et al.*, 1994). However, when additional samples from more sources were included, the indi-

vidual mines no longer clustered tightly (Young 1995:7). Further analysis allowed broad geographical separation, but only areas as large as states could be distinguished (Young *et al.*, 1997)." Subsequently, the first studies using the SIMS method (Secondary Ion Mass Spectrometry) for almost non-invasive isotopic measurements of copper and hydrogen are performed by Hull *et al.* (2008). The incident ion beam makes craters of only 10 to 15 microns in diameter. SIMS requires careful calibration of the measuring devices to get the necessary accuracy (Othmane *et al.*, 2015). A careful comparison of the studies of Hull *et al.* from 2008 and 2014 demonstrates the importance of the calibration process realized in between both articles to get reliable results: the same deposits did not show the same isotopic ratios in both papers. Thibodeau *et al.* (2018; 2015) focused on lead and strontium isotopes in order to support the isotopic results by geological assumptions and in an attempt to date the minerals. As these elements are in much lower concentrations than copper and hydrogen in turquoise, they could not be measured by SIMS at that time. The measurements were performed by multi-collector thermal ionization mass spectrometer (MC-TIMS) and multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS), after grinding and dissolving a few hundred milligrams of turquoise. While this destructive handling is not a major problem for natural deposits, it can represent a major drawback for archaeological or gem objects. However, Thibodeau *et al.* (2018) were able to relate the source of most of the 43 investigated pre-Columbian turquoise artifacts from Mexico to a local source but not to mines in the Southwest of US, a contradiction to previous studies. Because of a much better sensitivity (detection limits) of today SIMS devices, also lead isotope ratios can be quantified, but strontium concentrations are still too low (Queffelec *et al.*, 2020b).

Unfortunately, all the isotope ratio analysis hitherto are focused entirely on turquoise from the southwestern United States and Mexico. The potential of this method is not known at the global level. Distinguishing turquoises from different continents could also be helpful for providing provenance information of more recent archaeological of contemporaneous turquoise gems.

Optical spectroscopies

Diffuse reflectance spectroradiometry

One study of the provenance of archaeological materials using diffuse reflectance spectrometry (more commonly known as UV-Vis-NIR spectroscopy) on turquoise and its simulants was realized by Errera (1999). He demonstrated

the full potential of this non-destructive and inexpensive method to distinguish turquoise from other minerals. He also described that spectra from the same deposit are “practically superimposed” and that it is possible to distinguish them from one source to another. This pilot study, unfortunately, does not integrate numerous turquoise sources. It does not either integrate, as is the case in the author’s later studies on jade, multivariate statistical analysis on the spectra to relate the spectra of unknown artifacts to their source.

Vibrational spectroscopies

Some vibrational spectroscopy studies have been conducted on gem phosphates, and turquoise in particular (Čejka *et al.*, 2015; Dumańska-Słowik *et al.*, 2019; Fritsch *et al.*, 2019; Reddy *et al.*, 2006; Rossi *et al.*, 2017). The detailed study by Čejka *et al.* (2015) is based on only three samples, two from the United States and one from Senegal. The authors conclude that it is impossible to use Raman or infrared spectroscopy to identify any source. A recent publication of Díez-Pastor *et al.* (2018) reports an intra-source distinction of variscite, a mineral close to turquoise mineralogy, by using Raman spectroscopy coupled with multivariate analysis by machine learning. Another study, comparing turquoise of different color from the same source, demonstrate that turquoise Raman spectra are effectively affected by the composition of the major elements responsible for its hue (Dumańska-Słowik *et al.*, 2019). Work in this domain seems promising and could perhaps be applied to turquoise in the future especially for intra-site variations.

CONCLUSION

Turquoise, a hydrated phosphate of aluminum and copper, has been used from ancient to present times as a gemstone for the production of beads, pendants, and inlays on every continent. Despite this wide use both in time and space, it is a quite rare mineral on earth, mainly restricted to copper porphyry deposits and a few other mine contexts. The wide distribution and its varying composition predestines turquoise to be a very interesting indicator for the reconstruction of networks and systems of exchange of goods in ancient cultures. For these reasons, the provenance of turquoise has been widely investigated by archaeologists.

The different characterization methods applied to turquoise provenance studies show that the identification of turquoise sources is very complex. Different mineralogical, geochemical and isotopic tools have been tested in different parts of the world, with different results. What is clear is that it seems necessary to systematically use sophisticated

methods to obtain usable results. The so-called classical gemology criteria do not provide any information. There seems to be some potential in the mineralogical study of inclusions and casings, but this is non-existent in the literature except for the work of Welch & Triadan (1991). As shown for other gems, mineral associations with the gem are sometimes used almost exclusively as origin markers as demonstrated for the provenance of jadeite, where provenance studies are mainly based on petrographic and mineralogical criteria because chemical analyses allowed no clear distinctions between sources (Harlow *et al.*, 2011; Harlow *et al.*, 2006). Concerning spectroscopy, the only UV-Vis-NIR spectroscopy study on turquoise (Errera, 1999) contradicts the only study in vibrational spectroscopy (Čejka *et al.* 2015). The last authors concludes that vibrational spectroscopy is not suitable for the provenance studies of the three samples investigated. It is conceivable that larger studies, including more detailed analyses (chemometrics, machine learning) could perhaps improve the results of these methods, as is the case for other phosphate minerals (Díez-Pastor *et al.*, 2018). Geochemistry usually provides the most powerful methods for provenance studies, but it does not present an ideal solution to determine turquoise origin or exchange. Geochemical tools work well in particular cases or for particular region (Qin *et al.*, 2015; Ruppert, 1983). Old geochemical should be re-examined with new statistical methods. To supplement conventional geochemistry, isotopic measurements should be carried out, because they are promising for the areas where they have been applied (Thibodeau *et al.*, 2018).

At present, the state of the art is far away from an analysis on a global scale to determine the origin of a gem of unknown provenance such as contemporary gemstones or archaeological museum object completely without context. The application of isotopic methods to chemical elements within the turquoise structure for artifacts and mines in other regions of the world seems at the moment the most promising tool for turquoise provenancing at local, regional, and global scale.

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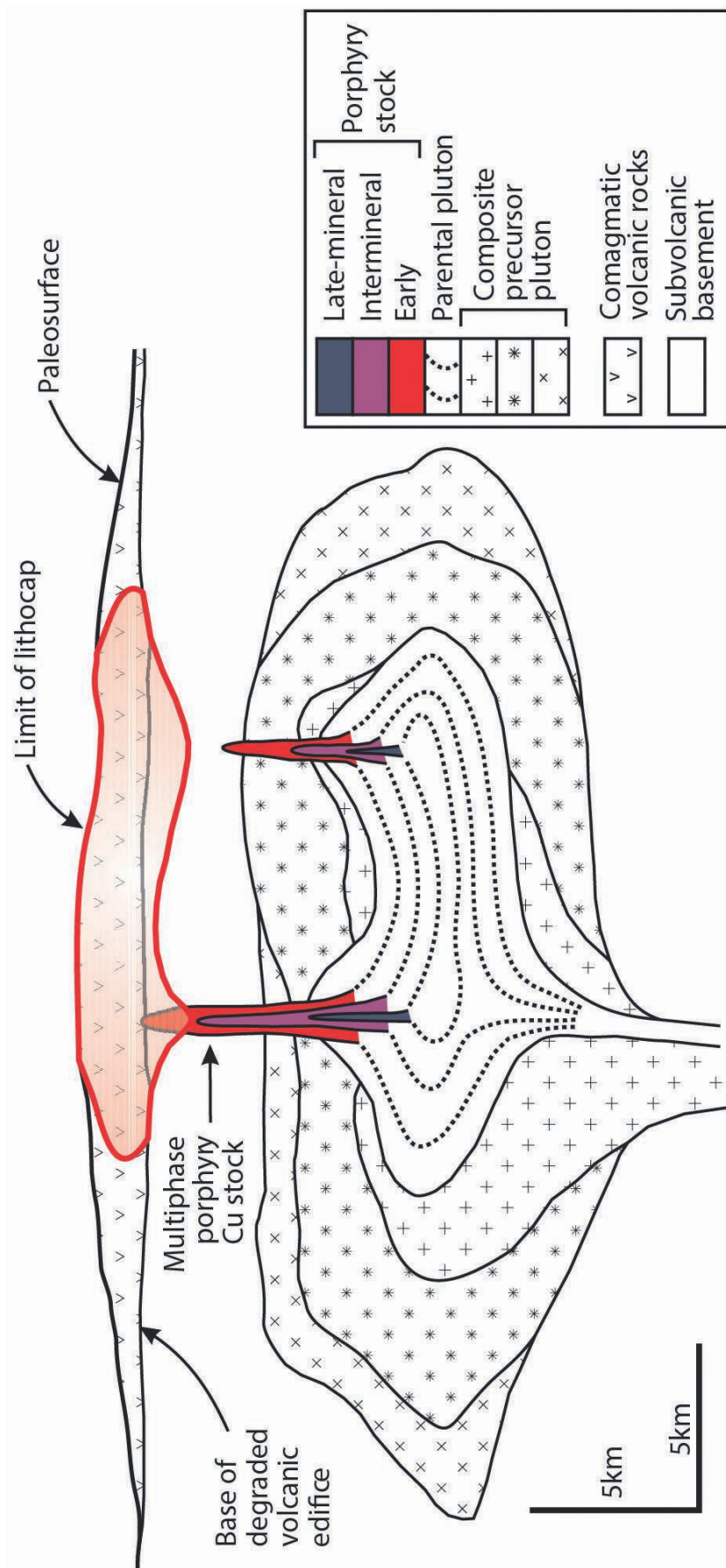


Figure 1: Alain QUEFFELEC, Provenancing turquoise: a difficult task (p. 21).