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Abstract: The goal of this paper is to review some of the biomolecular archaeological discoveries of ancient wine that my laboratory and colleagues have made over the past fifteen twenty years. I will interweave this discussion with the re-examination of samples using more precise chemical techniques. I will also raise questions about the domestication of the Eurasian grapevine (*Vitis vinifera* L. subsp. *vinifera*), and present a scenario of how I think that the ancient Near Eastern "wine culture" spread across the Mediterranean from east to west, ultimately reaching Italy, France, and Spain

Resumen: El objetivo de este trabajo es revisar algunos de los descubrimientos de la Arqueología Biomolecular del Vino que mi laboratorio y mis colegas han hecho durante los últimos quince años. Voy a entretejer esta discusión con la revisión de las muestras mediante técnicas químicas más precisas. También se plantean cuestiones acerca de la domesticación de la vid (Vitis vinifera L. subsp. vinifera), y se presenta un panorama de cómo creo que la antigua «cultura del vino» del Próximo Oriente se difundió a través del Mediterráneo, de Este a Oeste, llegando finalmente a Italia. Francia y España.

Palabras clave: Medicina antigua, Arqueología Biomolecular, ADN, Arqueología Experimental, resinas de árboles, Vinicultura, Cultura del Vino.

Key words: Ancient Medicine, Biomolecular Archaeology, DNA, Experimental Archaeology, Tree Resins, Viniculture, Wine Cculture

PATRICK E. MCGOVERN

From East to West: The Ancient Near Eastern "Wine Culture" Travels Land and Sea

The vibrant illustration in scarlet and black of Dionysos as a seafarer from Phoenicia by the 6thc. BC Athenian potter, Exekias, inside a drinking-cup, captures what mean by the ancient Near Eastern "wine culture" traveling from east to west. It shows the wine-god, who is equated with Baal or El in ancient Canaan, lying back in his single-man sailboat. When he encounters pirates on his way to Egypt or Cyprus, he turns the tables on them by entrancing them with the miraculous growth of a grapevine up the mast. Huge clusters of grapes shower down fragrant, sweet wine on the pirates, and turn them into dolphins

Much of what I summarize here draws upon my books on ancient wine (McGovern et alii, 1995; McGovern, 2003/2006). The latter received the grand prize in history, literature and the fine arts from the OIV in Paris, one of the sponsors of this conference.

This past year, my book, *Uncorking the Past* (2009/2010), appeared. This book is more than just about wine it traces alcoholic beverages around the world as far back in time as possible. It tries to show how far we've come in understanding the origins and biocultural importance of alcoholic beverages world-wide since our species came out of Africa some 100,000 years ago.

Together, these books present recent evidence that we can draw upon to reconstruct really ancient

viniculture, based upon the archaeological evidence: pottery sherds, grape seeds, and other remains that survived the millennia. This evidence must then be followed up by scientific analyses, which is my speciality. These contemporaneous data are most important, but they should be coupled with ancient textual and artistic evidence where available, ethnographic accounts of traditional winemaking, and so-called "experimental archaeology" in which ancient methods are used to see whether it is even possible to make a wine in accord with the ancient evidence and perhaps to discover whether it tastes any good.

The Mas des Tourelles winery, along the southern Rhone River at Beaucaire, illustrates what can be achieved by experimental archaeology. Here, the well-known archaeologist of Roman viniculture, André Tchernia, the honorary president of this conference who is seen on the left, and his colleagues, including Jean-Pierre Brun, also at this conference, erected a replica of an ancient Roman winepress, based on excavations at the site. They then proceeded to make some ancient wines, including Turriculae. Their formulation followed the formulation of the ancient Roman writer Columella in which lilac and fenugreek were added to the wine and the wine was aged in subterranean jars. It has a distinct sherry-like nose, which is a compliment to this most famous wine of Spain.

Such sweet wines were often made by drying the grapes and then pressing and vinifying the juice, to produce an intense raisiny wine. In keeping with my east/west theme, it should be noted that our earliest description of such wines occurs in a viticultural manual of Carthaginian writer Mago of the 3rd and early 2nd century BC (who Columella later cites: XII, 39.1-2). This winemaking tradition thus likely owes much to the Phoenicians, who founded the colony of Carthage.

What Tchernia and Brun accomplished at Mas des Tourelles has set the pattern for what many innovative winemakers are now doing. It also represents the way I like to think of my own research. Archaeological Chemistry or Biomolecular Archaeology combines discoveries from the past and present. A revolution in scientific techniques over the past forty years has made it to possible to re-examine and, in many cases, re-write the history of wine. We are just at the beginning of this process, which promises many more discoveries.

My research into ancient wine took a quantum leap forward when I organized a conference on "The Origins and Ancient History of Wine" at the Robert Mondavi winery in the spring of 1991. I was brought me in touch with wine scholars from around the world, setting my research agenda for the next two decades, as our laboratory went in search of ever more ancient wine.

The star of the show at the Mondavi conference was a pottery jar from Godin Tepe, in central Zagros Mountains of Iran, dated to about 3500-3100 BC At the time, it represented the oldest chemically confirmed wine jar in the world, based on the presence of tartaric acid, the fingerprint compound or biomarker for the Eurasian grape in the Middle East (see below, for more analytical details). Our study also showed that the wine in the jar had been

laced with a tree resin, important in helping to preserve wine. In other words, we had discovered a resinated wine, similar to modern Greek retsina.

The Godin Tepe jar inspired me to look for even earlier evidence of wine. The Neolithic period from about 8500 BC down to 4000 B.C offered the best prospects, because pottery was invented around 6000 BC in the Middle East. Pottery is virtually indestructible, and is ideally suited to absorb and retain compounds in liquids including wine. Neolithic villages, like Hajji Firuz Tepe, farther north in Zagros Mountains than Godin, were among the first, permanent, year-round settlements. They were a direct result of humans taking control of their food resources by domesticating a variety of animals and plants, including quite possibly the Eurasian grape. After the Mondavi conference, I simply asked a Neolithic archaeologist at the Penn Museum, Mary Voigt, if she had ever noted intriguing residues on any of the Neolithic pottery she had excavated at Hajji Firuz. She remembered a yellowish residue on the bottom of a narrow-mouthed jar [Fig. 1], but the chemical techniques were not sensitive enough when she excavated the site (1968) and proved inconclusive. The sherds then sat in the Near Eastern storage room of the museum for twenty-five years. This jar and five others were set into the clay floor and lined up along the wall of the kitchen of a mudbrick house, with an oven and other cooking vessels. Each had a capacity of about nine liters when full. Once we dug the sherds out of storage, we went to work using more modern methods of biomolecular archaeology to solve the archaeological puzzle of what the jars originally contained. Without going into all the analytical details here (references provided in McGovern, 2003/2006 & 2009/2010), we targeted tartaric acid, which occurs in large amounts only in the Eurasian grape (Vitis vinife-

One of six resinated wine jars (no. 68-205, University of Pennsylvania Museum; 69-12-15; height 23.5 cm.), from a "kitchen" of a Neolithic residence at Haiii Firuz Tepe (Iran), dating to ca. 5400-5000 BC (Photograph courtesy of Hasanlu Project, University of Pennsylvania Museum)

ra) in the Middle East, and confirmed its presence by diffuse-reflectance infrared Fourier-transform spectrometry, high-performance liquid chromatography, and a Feigl spot test. This acid is absorbed into the pottery fabric, and its more insoluble potassium and calcium salts readily precipitate out from solution and make up much of the lees that one sees in an unfiltered wine and at the bottom of an ancient vessel holding a liquid. Its dominant presence in the Hajji Firuz sherd and residue pointed to a grape product, most likely wine since once the grapes have been expressed as a liquid (obvious from the narrow mouth on the vessel and the accumulation of residue on the bottom of the jar), it will quickly ferment to wine in the warm climate of the Middle East.

In the past five years, we have gone on to detect tartaric acid in our ancient samples by a more powerful technique: liquid chromatography tandem mass spectrometry (LC/MS/MS) (see Guasch-Jané et alii, 2004; McGovern et alii, 2009). In brief, tartaric acid ($M_r = 150.1$) is ionized when it comes off an LC column at a specific time in the first cell of the quadrupole mass spectrometer, where it is massfiltered. The deprotonated molecular ion is then fragmented in a collision cell, and the daughter ions are again filtered by a second quadrupole. Tartaric acid is identified based on discrete fragmentation products that are detected. We have developed special extraction procedures using ammonium hydroxide in the extraction process, which accentuates the contribution of the tartrate ion and salt.

Of course, if oxygen remains available, fermentation can continue and eventually the acetic acid bacteria will convert all the wine to vinegar, and any competent winemaker, even one living in the Neolithic period, wants to avoid wine disease. Although cork was not yet available, raw clay stoppers function



the same way, absorbing liquid and expanding to seal off the mouth of the jar. Such stoppers were found in the vicinity of the wine jars at Hajji Firuz. Another chemical constituent of the Hajji Firuz residue, which prevents the dreaded wine disease, made it virtually certain that the jar originally contained wine. We identified terebinth tree resin, derived from the sap of a tree that grows throughout the Middle East and produces large amounts of resin in Fall when the grapes are maturing. Pliny the Elder, the famous 1st century. A.D. Roman encyclopedist, devoted a good part of book 14 of his Natural History to the problem of preventing wine turning to vinegar. Tree resins - pine, cedar, frankincense, myrrh, and especially terebinth, known as the "queen of resins" - were added to Roman wines for just this purpose. The most famous and expensive tree resin additive to Roman wine was myrrh, which came from the Arabian Peninsula and the Horn of Africa. Pliny went so far as to say that "There is no department of man's life on which more labor is spent".

Our chemical analyses over the past fifteen years have shown that resinated wine, which was also one of the most widespread human medicines in antiquity, was the overwhelming favorite for thousands of years before Pliny. Strangely, the tradition was perpetuated up to the present only in Greece as retsina.

The six wine jars in the Hajji Firuz kitchen, amounting to about 55 liters of wine are the "smoking gun" that the winemaking industry took off in the Neolithic period. This house was an ordinary residence, and if the amount of wine attested there were multiplied many times over by houses throughout the settlement, then the total quantity of wine would be much more than could be produced by gathering wild grapes. If this were true, then it was likely that the Eurasian grapevine (V. vinifera L. subsp. sylvestris) had been taken into domestication, like so many other plants and animals during the same period.

What makes the domesticated Eurasian vine so desirable is that it is hermaphroditic, the male (stamen) and female (pistil) organs are located together on the flowers of the same plant so it produces much more fruit on a predictable basis than does the dioecious separate male and female plants of the wild subspecies, which depend on wind and insect pollination. The self-fertilizing domesticated plant could then be selected for desirable traits, such as juicier fruit, thinner skins, and fewer seeds. Since grape seeds are genetically too variable to perpetuate desirable traits, humans also had to discover how to "clone" the vine horticulturally by transplanting branches, buds, or rootings. How early did such developments occur, and did they happen in only one place, perhaps at Hajji Firuz Tepe or elsewhere in the upland region of the Near East where the wild grape thrives?

The on-going debate about the domestication of the Eurasian grapevine comes down to whether it was domesticated in only one place at one time and then transplanted from place to place, with possible crosses occurring with wild plants in those other places from time to time, or whether it was separately domesticated in different places and times, including Europe

(e.g., Spain has been proposed as a separate domestication site: Arroyo-Garcia et alii, 2006). During historical times, when viticulture was better understood, it would be expected that the wild grapevine could be re-domesticated, but was it?

The "Noah Hypothesis" is an apt phrase to describe a one-time origin of viniculture in one place, because the biblical patriarch's first goal, after his ark came to rest on Mount Ararat, was to plant a vineyard and then make wine (Genesis 8.4 and 9.20). Like the Eve Hypothesis, which claims to trace all of humanity to an original human mother or mothers in East Africa - whether 2 million or 200,000 years ago - on the basis of mitochrondrial DNA lineage trees, a similar investigation of the Eurasian grape would seek the ultimate progenitor of modern domesticated grape varietals. In collaboration with colleagues in Europe, the United States and the Middle East, our lab and others are currently developing techniques to extract ancient DNA and compare modern wild and domesticated microsatellite sequences, including nuclear and chloroplast DNA (e.g. Vouillamoz et alii, 2006, Arroyo-Garcia et alii, 2006). An important conclusion of the Vouillamoz paper was that a numbereight of Georgian varietals (Tsitska, Kundza, Tsolikouri, Chkovra/Krakhuna, Tetri, Dzvelshavi, Maglari Tvrina, and Otskhanuri) show a close relationship to the western varietals Pinot Noir, Nebbiolo, Syrah, and Chasselas. The implication of this result is that a Near Eastern Noah Hypothesis is borne out, and that as the domesticated grapevine was transplanted westwards, there was introgression with wild vines to yield come of the modern European cultivars.

A recent paper (Myles et alii, 2011), using a microarray of thousands of grape genetic markers, including thousands of single nucleotide polymorphisms (SNPs) and 60 base-pair long oligo probes, similarly argues for Georgian varietals as closest to all the western cultivars included in their database. In this paper, the hermaphroditic gene has been isolated on chromosome 17. The latter sequence needs to be better defined, but together with other parts of the genome related to desirable winemaking traits of the domesticated vine, it should eventually be possible to resolve whether a single domestication in the Near East best accounts for modern European cultivars. Extraction of ancient DNA remains is also urgently needed to provide a chronology of the transplantation process and emergence of new cultivars as the grapevine traveled from east to west.

We do not know whether the Hajji Firuz wine was made from the domesticated or the wild Eurasian grape. The site does lie within the modern distributional zones of the wild grapevine [see Fig. 2], so the grapevine could have already have been domesticated there. However, because of its wider distribution from Spain to the oases of central Asia, other areas cannot be ruled out.

To test the Noah Hypothesis, we and other researchers have been focusing on three areas, where the wild grapevine thrives the Caucasus Mountains, sometimes argued to be the so-called "world center" of the Eurasian grape where its greatest genetic diversity is found; the Taurus Mountains of eastern Turkey where stupendous Neolithic sites have been found recently and where einkorn wheat and probably chickpea and bitter vetch — so-called founder plants for the Neolithic period — were first domesticated; and, of course, the Zagros Mountains. This upland region of the Caucasus, Taurus, and Zagros Mountains are all possibilities for the earliest domestication and the beginning of winemaking. What especially makes me think that the origins of viniculture will be found here is that there is

a great deal of archaeological and historical evidence for what can be called a "wine culture" gradually radiating out in time and space, from small beginnings in the northern mountains of the Near East in the Neolithic by at least 7000 B.C., to become a dominant economic and social force throughout the region and later across Europe in the millennia to follow.

You might say that we are looking for the vinicultural Garden of Eden. There is a lot of territory to cover in these mountains, and the modern search has only begun a concerted investigation for about ten years in search of relevant archaeological and grapevine evidence there. What is becoming increasingly clear, however, from these combined archaeological and chemical investigations is how entrenched "wine cultures" were there as early as the Neolithic period. These are cultures in which everyday meals, social events and special celebrations from birth to death, including rites of passage and major festivals, are marked by the drinking or offering wine. With time, viniculture comes to dominate the economy, religion, and society as a whole.

It is not difficult for this audience to understand what a "wine culture" is, since you are well-acquainted with those in Spain and elsewhere in southern and central Europe. Since I am from Pennsylvania, I need more convincing evidence. My trip to the Caucasus in 1998 showed me the hold that a "wine culture" could have over a people. Wine is still made there in large jars (kwevri) underground, and on the lees, as also attested by the numerous buried pithoi of Bronze and Iron Age buildings in the region. Dinners are presided over by a toastmaster (tamada), who offers up numerous toasts throughout the meal to the motherland, family, and life itself impossible to imagine apart from wine.

The "wine culture" in the Caucasus appears to have begun in the Neolithic period, based on pot-

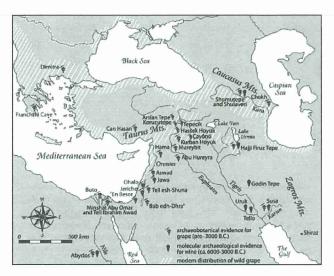


FIG. 2
Map of the ancient Near East and Egypt. The distribution of the modern wild grapevine (Vitis vinifera sylvestris) is shown by hatching; isolated occurrences of the wild grape also occur in Turkmenistan and Tajikistan, off the map to the east. The grape cluster symbol indicates wild and domesticated grape remains; primarily pips but occasionally skins and woodwhich were recovered from representative sites dating from the Neolithic down to the beginning of the Early Bronze Age (ca. 8000-3000 BC). The jar symbol marks wine jar types for the same time period, which have been chemically confirmed. (Map after version prepared by P. Zimmerman, MASCA, University of Pennsylvania Museum of Archaeology and Anthropology)

tery types and the finding of grape seeds of the "domesticated" type at Shuaveris-Gora. It reached a high-point in the Bronze Age when exquisite drinking goblets in gold and silver were produced, showing typical hunting and war scenes. The climax of such scenes is a festive banquet in which the ruler sits on a throne with cup in hand, attended by a cupbearer. This motif eventually became the standard symbol of Near Eastern and Egyptian civilization, dominated by wine, in which the king by raising his wine cup assured the fertility of the land, the prosperity of his people, and the continuation of his rule.

Although Georgia is perhaps the most enduring wine culture of this mountainous region, equally strong evidence for a Neolithic wine culture has begun to emerge elsewhere in the highlands. For example, the spectacular Neolithic sites of eastern Turkey have also yielded grape seeds in association with specialized cups and goblets, probably intended for wine-drinking and special ceremonies. They were made of an absorbent clay mineral, chlorite, which might well have paved the way for the invention of pottery.

Recently, what has been called the earliest winery, with a plastered treading floor and large underground jars to collect the juice and vinify and age it, was discovered in the Areni cave of Armenia (Bernard *et alii*, 2011). This site, dated to ca. 4100 B.C., is only about 300 km north of Hajji Firuz where we found evidence of the earliest wine (above). Such facilities begin to be recorded increasingly in other part of the Near East after ca. 3000 BC and throughout later periods, including Crete, Italy, and other Mediterranean countries.

Although not yet proven, a single domestication theory (Noah Hypothesis) would help to explain the gradual movement of a "wine culture" from the northern mountainous regions of the Near East to points south, east, and west reaching the Jordan Valley around 4000 B.C., then Egypt by ca. 3000 B.C., Shiraz in southwestern Iran about the same time, Greece by 2500 B.C., and Italy and the western Mediterranean at even later dates. The wine culture also spilled over into Egypt. Like

The wine culture also spilled over into Egypt. Like the southern Levant, the wild grape never grew in Egypt. Yet, a thriving royal winemaking industry had been established *de novo* in the Nile Delta by at least Dynasty 1, ca. 3000 B.C., probably under the tutelage of Canaanite winemakers from Lebanon and the southern Levant.

Numerous tomb reliefs and frescoes illustrate the vinicultural process, from picking the grapes overhead from well-trained pergolas, to stomping them out in small presses, to transferring the red-colored must to amphoras for fermentation, and stoppering the jars for storage.

Egypt first imported wine from the Jordan Valley and its environs, laying the foundation for the royal winemaking industry in the Delta, which began around 3000 BC One hundred and fifty years earlier, one of the first pharaohs of Egypt, Scorpion I of Dynasty 0, had 700 jars containing some 4500 liters imported from the eastern Mediterranean and piled high in his tomb at Abydos, to speed his way into the afterlife.

Our analyses of the wine residues in these jars illustrate what is now possible with much more sensitive chemical techniques. DNA analysis revealed what is thus far the earliest intact sequence of yeast DNA, 840 base-pairs long, and likely a precursor of the bread and beer Saccharomyces cerevisiae (Cava lieri et alii, 2003). We used LC/MS/MS to establish the presence of tartaric acid. Headspace Solid Phase Microextraction and Thermal Desorption gas chromatography coupled to a mass spectrometer (GC/MS) opened up a whole new world to us when, in addition to showing that the wine was resinated, it revealed that many different botanical herbs — probably including savory, wormwood, coriander, mint, sage and thyme, among others were mixed in (McGovern et alii, 2009).

We took this research a step further by testing compounds identified from the ancient wine and confirming that some of them had anti-cancer and other medicinal properties (McGovern et alii, 2010). In other words, among the many purposes of wine in an ancient culture (e.g., nutritional value, source of energy, social lubricant, mind-altering substance, economic product, etc., etc.), it also served as the principal medicine, long before the advent of modern synthetic drugs. Alcohol by itself relieved pain, stopped infection, and seemingly cured disease. Additionally, it is the perfect medium for dissolving and dispensing botanicals. We have dubbed this new project Archaeological Oncology: Digging for Drug Discovery.

Let me conclude by briefly tracking the Near Eastern wine culture from east to west across the Mediterranean.

What I propose is that the Canaanites and Phoenicians of Lebanon, with their renowned sailing vessels called Byblos ships, were instrumental in establishing new wine industries and spreading the wine culture, along with Royal Purple dyeing (McGovern & Michel, 1990), the alphabet and much more, one step at a time spread across the Mediterranean. The recent discovery of two Phoenician ships off of Ashkelon in Israel (Stager, 2005), loaded with thousands of wine amphoras being transported to Egypt or Crete from a Lebanese port, is just the tip of the archaeological iceberg. More explorations using remotely operated vehicles will likely discover much more evidence of the Canaanite and Phoenician role.

We have evidence that the wine culture probably first reached Greece by at least 2200 BC (McGovern et alii, 2007), so the Canaanites might have been plying these waters with their loads of wine long before the Phoenicians. We identified the earliest retsina at the site of Myrtos-Phournou Koryphe on the southern coast of Crete, and maritime contacts with Canaan / Phoenicia and/or

Egypt could well have played a major role in this development.

The Greek traders likely entered the picture later than the Levantine traders. Greece was still dominated by a mixed beverage, a kind of Greek grog, made from Pramnian wine, honey and barley, topped with cheese the so-called *kykeon* of the Homeric epics. It was prepared in large cauldrons and drunk from cups, like the marvelous Nestor cup, found at Mycenae and described in the Iliad. When the Greeks adopted the alphabet from the Phoenicians, the wine culture probably went with it. The earliest Greek and Etruscan inscriptions are wine inscriptions.

Eventually, the Etruscans came on the scene and were a major conduit of the wine culture to southern France. Their amphora is modeled after the Phoenician amphora, and where a similarity of form exists, one can make a good case that it is due to direct contact with the Phoenicians and was used for the same contents, namely grape wine (McGovern, 2007). In my opinion, the Etruscans thus took up the banner of the "wine culture" mainly from the Phoenicians, and to a lesser extent from the Greeks, who were more wedded to a mixed beverage, and eventually embarked upon the mass production of wine and its shipment by boat, especially to southern France by 600 BC.

The ultimate answer to the relative involvement of Phoenicians, Etruscans, and Greeks in transferring the wine culture to France and Spain partly lies at the bottom of the Mediterranean where so many shipwrecks full of wine-related pottery have been discovered and excavated (Parker, 1992). It should also be noted that a profusion of Iron Age shipwrecks have now been located and excavated along the Italian, French and Spanish coasts. They were so loaded up with wine-related vessels that, in a very real sense, one could say that the transfer of

first the Phoenician and Greek cultures and then that of the Etruscans in the western Mediterranean was mediated by the wine culture itself (Morel, 1983) On land, archaeological excavations can also contribute a great deal to solving the puzzle. We have begun a new project with Michel Py and colleagues at the Centre National de la Recherche Scien tifique, analyzing what are believed to be some of the earliest wine amphoras from sites in southern France. The heavily Etruscan influenced site of Lattes (Dietler, 2010), near Montpellier, is a major focus, but we are also looking at more Phoenician - and Iberian - influenced sites to the west, such as Pech Maho. Results are still preliminary, but very promising. Just like in Greece and on Crete, Etruscan and Celtic grogs might well have preceded the introduction of grape wine.

The history of civilization is, in many ways, the history of wine. The domesticated grapevine and winemaking was carried by the Etruscans and Phoenicians, later the Romans, to southern France and the Spanish coast. It traveled northwards gradually, eventually reaching the many fine winemaking regions of today, including Burgundy and Bordeaux and Jerez and Extremadura.

Relatively recently, viniculture has spread to the New World. It is my contention that almost every Old and New World cultivar, with their infinite varietal range of tastes and bouquets, ultimately derive from the Eurasian species of the mountainous Near East, which have been transplanted or crossed again and again. Each culture (whether Spanish, Canaanite, Phoenician, Egyptian, Californian) has its own story to tell about its relationship with wine and the vine. Together, they form a truly remarkable history of a truly remarkable plant and its product intertwining itself with human culture throughout the world.

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