

Obsidian Provenance Research in the Americas

MICHAEL D. GLASCOCK*

Research Reactor Center, University of Missouri,
Columbia, Missouri 65211

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ABSTRACT

The characterization of archaeological materials to support provenance research has grown rapidly over the past few decades. Volcanic obsidian has several unique properties that make it the ideal archaeological material for studying prehistoric trade and exchange. This Account describes our laboratory's development of a systematic methodology for the characterization of obsidian sources and artifacts from Mesoamerica and other regions of North and South America in support of archaeological research.

Introduction

In a world dominated by objects made of plastics, metals, and other synthetic materials, it is easy to overlook the fact that for more than two million years stone was the most important raw material used by humans for technological purposes.¹ Until a few thousand years ago, humans used stone tools for basic tasks such as hunting, cutting, scraping, and grinding. In Europe and the Far East the use of stone tools began to decline with the beginning of the Bronze Age, but in other regions of the world the Stone Age ended more recently. For instance, Native Americans were still using stone tools when Columbus discovered the Americas, and in New Guinea stone for making axes was being quarried until the 1950s.

During the process of producing a tool from raw stone, its usage, and the time between its eventual discard and recovery by an archaeologist, most of the basic physical and chemical properties of the stone remain unchanged. Knowledge of the physical properties and chemical composition of an artifact made from stone can frequently be used to identify the original source, in some instances with a high degree of certainty. Archaeologists commonly use the term *provenance* to describe the process of determining the source of an artifact.

Although many types of stone were used by different prehistoric cultures, the lithic material providing archaeologists with the clearest evidence of contact between different cultures is the volcanic glass obsidian. Obsidian is shiny and attractive, easily recognized, and relatively

easy to work, either through flaking or through cutting and polishing.

Prehistorically, the acquisition of obsidian for tool manufacture and usage developed in different ways, ranging from local collection to complex systems involving long-distance commerce over land and/or sea. As a result, the sourcing of archaeological obsidian can be of great assistance for investigating the cultural, social, and economic development of ancient societies.

In this Account, our achievements in developing a systematic methodology for characterizing thousands of obsidian artifacts and hundreds of sources throughout the continents of North and South America are presented. In the next section, we describe the properties of obsidian that make it ideal for archaeological research along with related developments in obsidian research preceding the work of our laboratory.

Background

Obsidian is a glass, a supercooled liquid, formed when a highly viscous volcanic lava of high silicon and aluminum content cools rapidly, usually at the margins of a lava flow, such that the process of mineral crystallization is precluded.^{2,3} The glass is generally black or gray in color (other colors are possible, depending upon the composition and the circumstances of formation), and it is sometimes banded or streaky in appearance. Due to an atomic structure that is entirely disordered, obsidian is physically amorphous and isotropic; this is one of the main reasons why it makes such effective tools, since flakes can be struck from a core in almost any direction. Some low-quality obsidians may contain a significant proportion of crystals (i.e., phenocrysts) made of minerals similar in composition to the obsidian, but due to their poor fracturing properties they produce low-quality tools.

Because glass is not a stable material at ambient temperatures and pressures, it gradually hydrates through the diffusion of water into the outer surface and along cracks to form perlite. As a consequence, its lifetime is relatively short by geological standards. The sources of obsidian exploited by prehistoric peoples are almost entirely restricted to young volcanic areas. Few obsidian sources are greater than 10 million years old, and many are less than 100 000 years of age.⁴ Geographically, obsidian sources are found in the Mediterranean, Central Europe, Turkey, Eastern Africa, the Andes Mountains of South America, the trans-Mexican volcanic belt, the western United States and Alaska, Japan, New Zealand, and the islands of the South Pacific.

The composition of most obsidians ranges from about 66–75% SiO₂, 10–15% Al₂O₃, 3–5% Na₂O, 2–5% K₂O, and 1–5% total Fe₂O₃ + FeO. Peralkaline varieties of obsidian are typically higher in Fe composition than are rhyolitic obsidians. In addition, the intrinsic water content of obsidian ranges from 0.1 to 0.5%. As hydration occurs, the

Michael D. Glascock, born in Hannibal, MO, on June 27, 1949, received his B.S. from the University of Missouri–Rolla in 1971 and his Ph.D. from Iowa State University in 1975. After three years of postdoctoral work at the University of Maryland, he joined the University of Missouri–Columbia in 1979, where he is currently a Senior Research Scientist and Group Leader of the Archaeometry Laboratory. His research interests are in developing analytical methods to characterize archaeological materials and interpreting the compositional information from the artifacts to better understand prehistoric trade and exchange in several regions of North and South America.

* E-mail: glascockm@missouri.edu.

water content of obsidian increases to about 5 wt %, resulting in concentric, “onionskin” cracking patterns that gradually destroy the glass. The remaining elements in obsidian are present at concentrations below 1% and are usually referred to as the trace elements.

Most obsidian sources are chemically homogeneous, with variations in composition on the order of a few percent or less. However, different sources have different compositions as a reflection of the compositions of parent rocks that were melted and changes that take place in the magma chamber prior to eruption. Although the major elements in obsidian are restricted to a relatively narrow range of composition, the abundances of trace elements may differ by orders of magnitude between sources. If the variations within sources are smaller than the differences between different sources, the provenance of obsidian artifacts can be successfully established (i.e., the “Provenance Postulate”).⁵

Obsidian lava is formed at high temperatures (>1000 °C) and in equilibrium with solid materials; the trace elements are distributed between the liquids and solids. During this process, many of the transition elements such as chromium, cobalt, and nickel are strongly absorbed by the solids. These elements are typically known as the *compatible* elements because they are compatible with the crystallization of solids and are readily removed from the liquid phase. Other elements *incompatible* with the solid phase tend to become more concentrated in the liquid. The incompatibility of elements with the solid phase is caused by two factors.⁶ First, ions such as rubidium, cesium, strontium, and barium are too large for the available ionic sites in the solid. A second cause of incompatibility may be that certain ions have too high a charge to fit within the crystal structure of the solid. Examples of the latter include the triply charged rare earths such as lanthanum and cerium, the quadruply charged ions of hafnium, zirconium, and thorium, or the quintuply charged ions such as tantalum and niobium. As a result, the incompatible element mixtures may be different from one source to the next and become a sensitive indicator of origin. It also is possible within a single magma chamber, as the magma evolves, that glass eruptions occurring at different times will have different trace element chemistries.

Prior to the 20th century, wet quantitative chemistry by gravimetry or volumetry was the main analytical method employed by the early chemists who studied objects from antiquity.⁷ Because the dissolution of lithic materials (i.e., pottery and rocks) is challenging, the early chemists limited their interests to ancient coins, glasses, pigments, and other remains. The French mineralogist, M. A. Damour, was the first to report on a study of polished stone axes from Celtic sites in Europe.⁸ By examining the mineralogical and chemical compositions of artifacts and comparing to raw materials, Damour suggested it might be possible to use the chemistry to track the migratory movements of prehistoric peoples.

The emergence of instrumental methods such as emission spectroscopy, neutron activation analysis (NAA), and

X-ray fluorescence (XRF) spectrometry in the mid-20th century rapidly changed the analytical scene.⁹ Eventually, the increased application of these methods to studies on stone tools, pottery, and other archaeological specimens contributed to the development of a new discipline known as *archaeometry*.¹⁰

Cann and Renfrew¹¹ led the way when they used emission spectroscopy on obsidian artifacts from the Mediterranean to show evidence of prehistoric cultural contact. In North America, Gordus et al.^{12,13} measured Mn and Na in obsidian by NAA to identify the geologic sources of obsidian projectile points found on prehistoric sites in the Great Plains and Ohio Hopewell regions. Later, Pires-Ferreira¹⁴ used Gordus's procedure to trace obsidian artifacts from the Valley of Oaxaca, Mexico, to sources in central Mexico. Heizer¹⁵ and his colleagues used semi-quantitative XRF to analyze five trace elements in obsidian artifacts from sites in Veracruz, Mexico. In 1969, Perlman and Asaro¹⁶ developed a standard-comparator approach to the NAA of archaeological samples currently in use by most archaeometry laboratories in existence today.

In particular, XRF and NAA have been employed most often for the analysis of obsidian artifacts in the Americas. The advantages of XRF¹⁷ are that little sample preparation is necessary (possible destruction of artifact is avoided); the analysis is rapid; and the cost is low. The disadvantages are that only 8–10 elements are measured well; the sample surface should be flat; and the sample must be of a minimum thickness and diameter. The advantages of NAA¹⁸ for obsidian analysis are that thermal neutrons are very penetrating such that the method provides a bulk analysis and about 30 elements can be measured with the majority of these at high precision (<2.5%). The disadvantages of NAA are that a portion of the sample must be destroyed when preparing it for irradiation, one must have access to a nuclear reactor, and NAA is more expensive.

Reviews of the compositional studies of obsidian from North and South America conducted prior to 1980 indicate that only a fraction of the chemical and geological information potentially available was being recovered and too many sourcing errors were occurring.¹⁹ In general, unsatisfactory characterization of sources was the most serious limitation of the early obsidian provenance studies. For example, some of the problems included collection and analysis of only two or three specimens from each source, poor descriptions of source areas, failing to consider secondary source areas, and analysis of only a few elements. In particular, the Valley of Oaxaca artifact study¹⁴ mentioned above was later shown²⁰ to contain many sourcing errors due to inadequate information about the sources. Another problem was the illogical assumption by some archaeologists that the elements successful in differentiating sources in one region would be successful in other regions also. By 1980, it was becoming clear to several archaeologists and chemists that a comprehensive compositional and descriptive database for obsidian sources in North and South America was needed.¹⁹

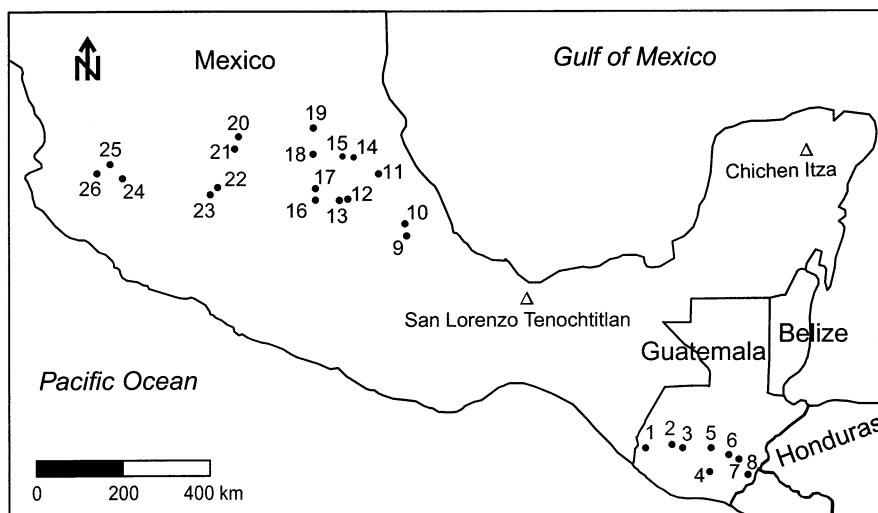


FIGURE 1. Map of the region known as Mesoamerica showing the locations of major obsidian sources. The sources in Guatemala are (1) San Lorenzo, (2) San Martín Jilotepeque, (3) San Bartolomé Milpas Altas, (4) Laguna de Ayarza, (5) El Chayal, (6) Sansare, (7) Jalapa, and (8) Ixtepeque. The sources in Mexico are (9) Pico de Orizaba, (10) Guadalupe Victoria, (11) Zaragoza, (12) Paredon, (13) Santa Elena, (14) Tulancingo, (15) Tepalzingo, (16) Otumba, (17) Malpais, (18) Pachuca, (19) Zacualtipan, (20) El Paraiso, (21) Fuentezuelas, (22) Ucareo, (23) Zinapécuaro, (24) Tequila, (25) Magdalena, and (26) Teuchitlán.

Obsidian Research at MURR

In 1979, an archaeologist, Robert Cobean, and two analytical chemists from MURR, James Vogt and the author of this Account, initiated a collaboration to characterize obsidian from sources in Mesoamerica. The primary goals of this project were to better define and differentiate between the major known obsidian sources located in east-central Mexico and the highlands of Guatemala. This region, commonly called Mesoamerica by archaeologists, was inhabited for several thousand years by numerous cultures including the Olmecs, Toltecs, Aztecs, and Maya, all of whom relied heavily upon obsidian to make sharp-edged tools and some objects of art. Evidence of the importance of obsidian is widespread throughout the region, where many sites are densely covered with obsidian artifacts. By using neutron activation analysis and collecting high-precision data for as many elements as possible, we hoped to improve significantly on the accuracy with which obsidian artifacts could be attributed to specific sources.

Cobean spent most of 1980 collecting obsidian source samples in Mexico, in part collaborating with archaeologists from Mexico's Instituto Nacional de Antropología e Historia (INAH). The fieldwork obtained 818 samples of obsidian (weighing a total of 710 kg) from 25 source areas in central Mexico.²¹ An earlier collection²² of 77 source samples from some of these sources and other sources located in the Guatemala highlands originally analyzed by XRF was also made available for NAA. Samples from flows and secondary deposits for all of the known obsidian sources in east-central Mexico were represented. The number of samples collected for each source was dependent on its size. As many as 150 source samples were collected for the largest source, and as few as 10 samples were collected for the smallest sources. The geographic coordinates for individual samples were recorded so that

we could map the chemical variability within each source. If differences were observed within sources, we would be able to trace artifacts to specific quarries.²³ Figure 1 shows the locations of obsidian sources in Mexico and Guatemala where source specimens were collected.

Detailed studies of sample preparation procedures²⁴ and evaluations of different analytical standards²⁵ were conducted at the beginning of the project. The sample preparation study concluded that grinding the obsidian into a fine powder introduced unacceptable and variable levels of contamination from sample to sample. As a result, a procedure involving the fracturing of source samples and careful selection of interior fragments free of dirt, cracks, stress fractures, and mineral inclusions was used to obtain representative samples for NAA. The evaluation of standards led to identification of a new standard reference material, SRM-278 Obsidian Rock, as the most suitable primary analytical standard because it was most similar to the unknowns. In addition, a second reference material, SRM-1633a Coal Flyash,²⁶ was selected for quality control purposes. Analyses of the two standards were conducted in a number of different laboratories, establishing both as highly reliable comparator standards for conducting multielement NAA on geological specimens.²⁷

Three NAA procedures were applied in order to comprehensively analyze the obsidian (i.e., approximately 30 elements were determined). A short-irradiation NAA procedure was optimized to measure the elements Al, Cl, Dy, K, Mn, and Na in a single count following neutron irradiation. A long-irradiation NAA procedure was employed to measure the elements Ba, La, Lu, Nd, Sm, U, and Yb in one measurement a few days after irradiation. A few weeks later, the long irradiation samples were counted a second time to measure the elements Ce, Co, Cs, Eu, Fe, Hf, Rb, Sb, Sc, Sr, Ta, Tb, Th, Zn, and Zr. A

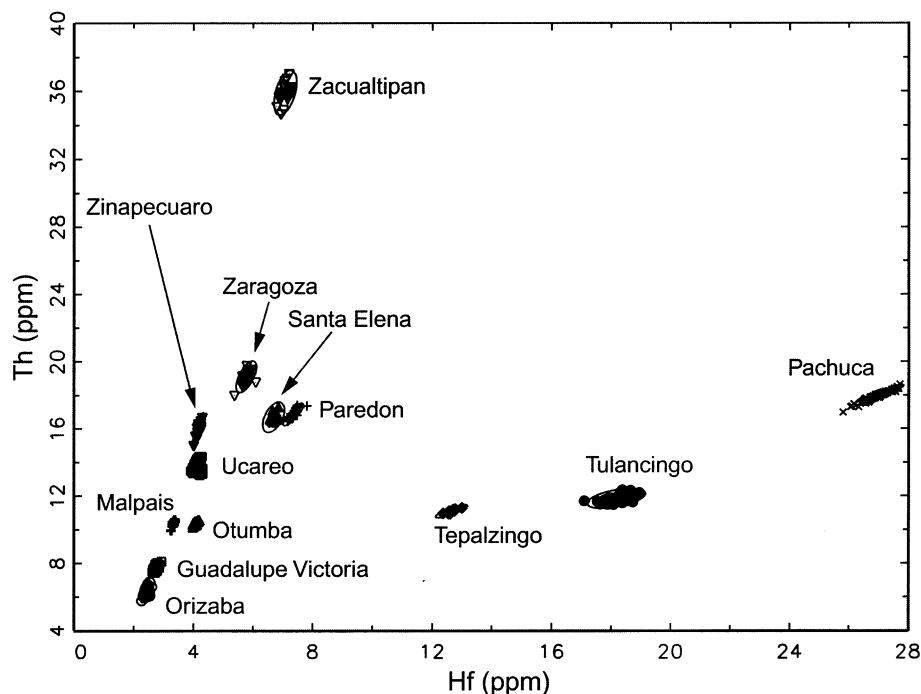


FIGURE 2. Bivariate plot of Hf versus Th for obsidian sources in central Mexico. Probability ellipses are shown at the 95% confidence level.

final procedure known as prompt gamma neutron activation analysis (PGNAA) was used to measure the elements B, Sm, and Gd. In addition, corrections from the interferences caused by certain uranium-fission products were required²⁸ to accurately determine the concentrations for Ba, La, Ce, Nd, and Zr.

Although the PGNAA procedure was later discontinued due to an extended interruption in its availability, all irradiations of obsidian source materials up to the present time have utilized the above-mentioned short- and long-irradiation procedures. Repeated usage of the Obsidian Rock and Coal Flyash standards with analyses of obsidian sources over the past 20 years have resulted in a well-documented monitoring of data quality control.²⁷ The last of the Mesoamerican source samples collected by Cobean were analyzed in 1990, but other archaeologists working in the same region continued to submit samples for newly discovered sources along with artifacts submitted for provenance determination.^{29–31}

A few bivariate element plots are often adequate to identify the sources of obsidian artifacts from Mesoamerica with a high degree of confidence. Figures 2 and 3 illustrate this by showing bivariate plots for the sources in Mexico and Guatemala, respectively. Although these are among the best pairs of incompatible elements for source differentiation in Mesoamerica, in other regions other elements may be more successful. The author recently showed³² that the probability of misclassifying an artifact from one of the central Mexican sources is less than 4 in 100 000 and from one of the Guatemalan sources is less than 1 in 100 000. Over the past decade of operation, only a small handful of the several thousand artifacts from Mesoamerica analyzed have not been securely linked to

a known source. In most cases, artifacts not linked to known sources have come from as-yet undiscovered sources.

The strength of our multielement approach is also illustrated by our capability of differentiation between individual flows and outcrops for some obsidian sources such as one located near the small town of San Martín Jilotepeque, Guatemala.³³ In this study, a total of 63 source specimens were collected from obsidian quarries within a radius of 10 km of the town and analyzed by NAA at MURR. As shown in Figure 4, our data indicate six different chemical subgroups are present, and these data provide solid evidence that the volcano responsible for the San Martín Jilotepeque obsidian erupted at least six different times. Thus, in some cases, our NAA data are capable of correlating artifacts to specific quarries within a large source.

In those instances where the archaeologist is satisfied with a less robust analytical approach (due to the need for quick results or reduced cost), we will conduct only the short-irradiation NAA procedure on batches of artifacts. Using the elements Ba, Mn, and Na and the discriminant analysis method mentioned earlier, we have shown that more than 95% of obsidian artifacts from Mesoamerican sites can be securely sourced with only these three elements.³⁴ Thus, only about 5% of Mesoamerican artifacts require the more expensive, long-irradiation NAA procedure to determine their sources.

The Chichén Itzá Example

The Maya, famous for their knowledge of astronomy and for developing a writing system, built numerous cities in southern Mesoamerica that were occupied and abandoned long before the arrival of Europeans. Maya civiliza-

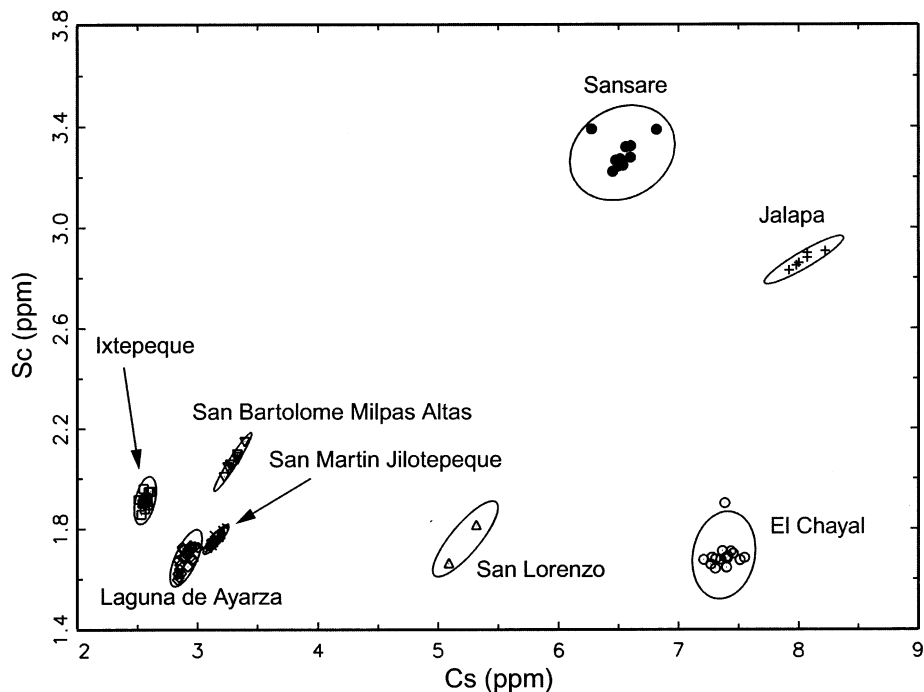


FIGURE 3. Bivariate plot of Cs versus Sc for obsidian sources in Guatemala.

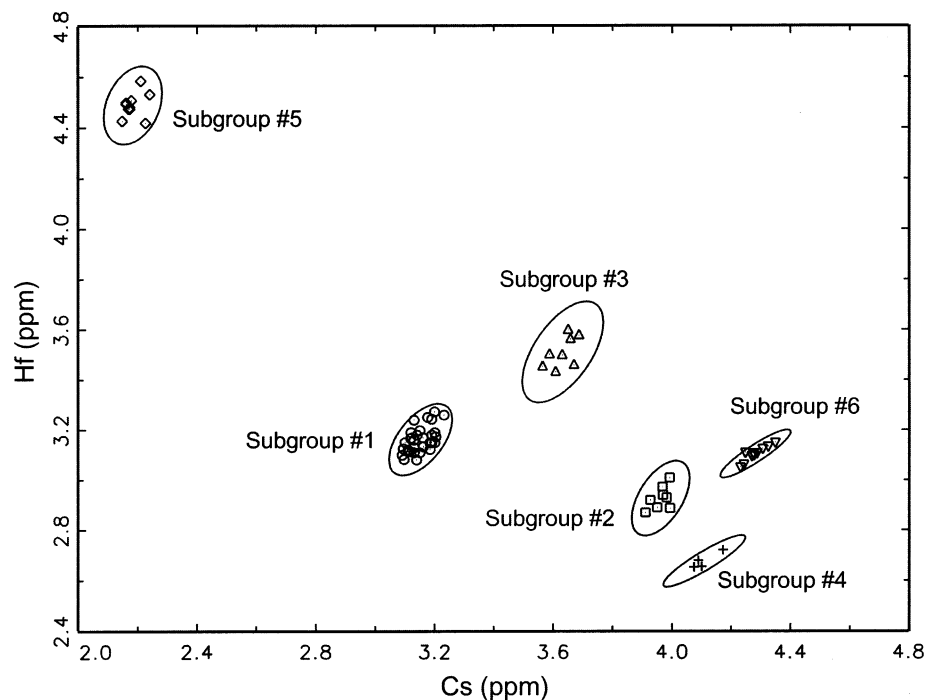


FIGURE 4. Bivariate plot of Cs versus Hf for obsidian subgroups identified within the source at San Martín Jilotepeque, Guatemala.

tion originated in the Yucatán Peninsula and spread throughout much of southern Mexico and Guatemala. A number of lowland Mayan sites such as Chichén Itzá, Tulum, Palenque, and Tikal show widespread evidence of obsidian usage. In particular, the religious and ceremonial site of Chichén Itzá is located more than 700 km from the Guatemalan obsidian sources and more than 1000 km from the nearest source in central Mexico.

A collection of 421 artifacts from Chichén Itzá and nearby sites was submitted for compositional analysis by Geoffrey Braswell (an archaeologist from the University

of Buffalo). The samples were subjected to our short-irradiation NAA procedure and the data were compared to the Mesoamerican obsidian source database. Figure 5 shows a bivariate plot of Mn vs Na for the artifacts projected against probability confidence ellipses for nine sources located in Guatemala and Mexico. The comparison was highly successful with sources for nearly all of the artifacts securely established. Nineteen (i.e., 4.5%) of the artifacts with the lowest probabilities of membership on the Mn vs Na plot were submitted to the long-irradiation NAA procedure. Examination of the additional

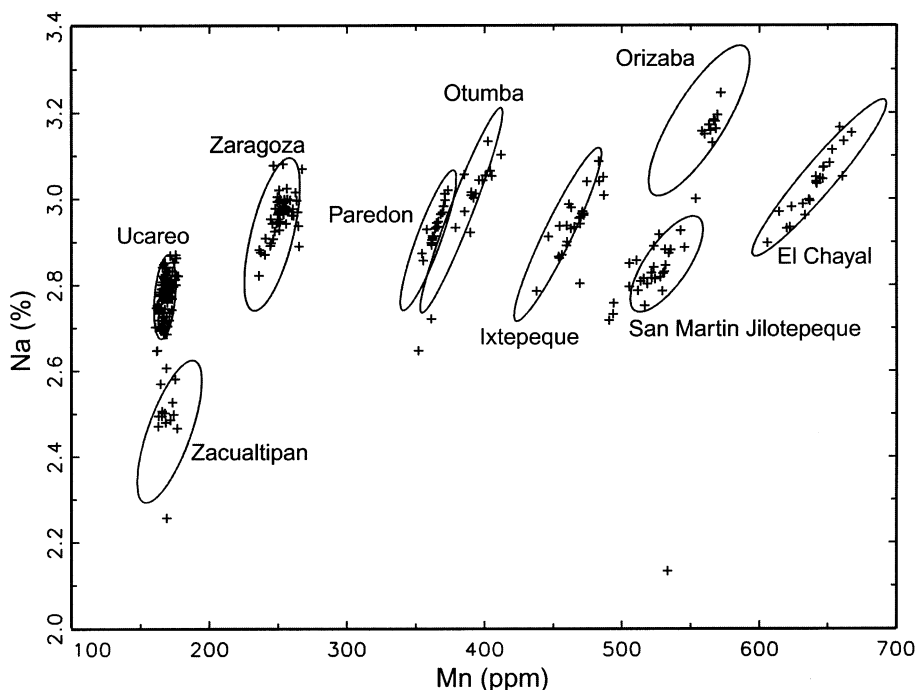


FIGURE 5. Bivariate plot of Mn versus Na for 421 obsidian artifacts from the site of Chichen Itzá projected against the 95% confidence ellipses for sources in Mexico and Guatemala.

data found that 17 of the 19 artifacts agreed with the most likely source as suggested by the short-irradiation procedure. Two samples, with the lowest overall membership probabilities, were found to be tektite (a meteoritic glass) instead of obsidian.

The Chichén Itzá example illustrates that obsidian provenance studies can be very beneficial to archaeologists interested in studying long-distance interactions between prehistoric humans in the form of trade and exchange. Many interesting questions about the inhabitants of Chichén Itzá and their contacts with the peoples living near the obsidian sources can be examined using the data from obsidian provenance studies. The objective of archaeological research on obsidian is to say something about the people who used obsidian and why exploitation or trade patterns changed in antiquity. It is clear that such answering these questions depends heavily upon a reliable obsidian source database such as the one we have created in the MURR laboratory.

The Future

Although the obsidian research described above is primarily limited to the sources we have studied in Mesoamerica, our collection and analysis of obsidian sources is not limited to that region. A few years ago, we established a goal of locating and characterizing all of the obsidian sources in North and South America that were potentially utilized by prehistoric peoples. Over the past decade, dozens of archaeologists and geologists working throughout the western hemisphere have contributed to this effort by submitting more than 5000 source specimens to MURR. The comprehensive analysis we are conducting by NAA have helped to identify approximately 300 unique chemical fingerprints for sources ranging from northern

Alaska to southern Chile.^{35,36} We actively encourage our colleagues to submit specimens from any new sources they encounter.

Finally, we also recognize that the analytical scene is changing.³⁷ Although NAA is a powerful multielemental analytical method, the number of available research reactors is in serious decline, such that in another 15–20 years it may no longer be possible to analyze obsidian by NAA. New methods such as ICP-MS with laser ablation are emerging as viable alternatives to NAA because of the large number of elements one can measure. To participate in this transition, we recently acquired a high-resolution ICP-MS with laser ablation system for MURR. With the information in our NAA-based obsidian database and our collection of source samples, we are in the process of intercalibrating NAA and LA-ICP-MS in order to preserve our obsidian database for future archaeological research.

Summary

Over the past 40 years, studies of obsidian artifacts and other archaeological materials have revolutionized archaeology. Archaeologists have come to rely on provenance studies to investigate human activities such as trade and exchange, population mobility, and settlement patterns. The successful collaboration between archaeology and chemistry in the study of obsidian is one of the greatest achievements.

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