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Lithic raw materials provenance in the Caribbean islands:

flint, jasper, obsidian and so on.

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Abstract

Lithic materials in archaeological records have always been the object of special attention by archaeologists, especially from prehistoric contexts. They draw such attention thanks to their often excellent preservation, and the information that the intrinsic characteristics of the rocks can yield about past behaviors including mobility of goods and people through chemical analysis and provenance studies. Although they do not represent the majority of the artifacts recovered from precolonial archaeological sites in the Lesser Antilles, their diversity necessitates varied knowledge, methods, and skills to be studied. Here, we present preliminary results from lithic specialists in characterization of volcanic rocks, diorite, turquoise used for bead production, and silicites.

Several volcanic glass artifacts have been identified for the first time from Saladoid sites from Guadeloupe and Dominica. However, many questions remain about the geological origin and the petrogenesis of these artifacts given their very specific chemical composition. On the other hand, detailed microtextural and micropaleontological studies of the different siliceous raw materials provide first results identifying and mapping their origin and distribution among the Caribbean islands. As a first glance of what will become a long-term project, results presented in this paper demonstrate the capacity of petrological studies carried out with appropriate technical methods to help us improve our knowledge of the Ceramic Age populations' movement and rock distribution networks.

Keywords

Lithic raw materials, Silicites, Obsidian, Ceramic Age, Lesser Antilles, Sourcing

Introduction

The mobility of Amerindians within and between islands during the Ceramic Age has been a particularly intriguing topic for Caribbean archaeologists. In this paper, we aim to contribute to the knowledge of intra-island mobility, inter-islands exchanges, and regional networks within the Caribbean archipelago. Our previous efforts to describe circulation of lapidary raw materials at a regional scale (Queffelec et al., 2018, 2020, 2021) inspired us to pursue a new direction and investigate the provenance and circulation of lithic raw materials more broadly. Our analysis of the current state of knowledge of lithic raw materials in the Caribbean reveals a lack of thorough studies and descriptions, as well as a shortage of specialists in this domain. To address this gap in knowledge, our project aims to gather specialists in a variety of stones used as utilitarian or lapidary raw material and with this in mind, create, describe, and valorize a reference collection (i.e., lithoteca). The aim is to make it available to the community to enable robust identification using objective criteria and ultimately to contribute to an understanding of the landscape exploitation by people who lived in the Caribbean.

The current state of knowledge regarding inter- and intra-island mobility based on lithic material distribution in the Caribbean is heavily reliant on the observation of few key stone sources. These include i) the identification of Antigua's flint in numerous archaeological sites (e.g., Knippenberg, 1999; Knippenberg & Zijlstra, 2008; van Gijn, 1993), ii) the studies of the distribution of the distinctive rocks from St. Martin found in several sites on that island and on others (Knippenberg, 2004, 2007; Stouvenot, 2001), and iii) the identification of radiolarite from La Désirade in several archaeological sites (de Waal & Knippenberg, 2020).

To achieve our goals, we have organized our project into three distinct work packages, each focused on the different highly specialized knowledge and methods required for the analysis of i) silicites¹, ii) volcanic rocks, and iii) greenstones. Our approach involves the combination of pre-existing lithotecas with new fieldwork to establish a comprehensive georeferenced collection. We undertake thorough characterization of the samples, using binocular observations for silicites and conducting geochemical and mineralogical analysis for volcanic rocks, in addition to documenting the criteria of identification with photography. Finally, all

¹ The term silicites designates all the rocks having undergone a diagenetic silicification of chemical or biochemical origin (e.g., flint, chert, silcrete, hydrothermal silica). It allows to avoid the term of "flint", the meaning of which is subject to controversy and misunderstandings depending on whether it is used by geologists, petrographers, sedimentologists or archaeologists. We therefore use the term flint only to designate rocks having undergone an initial diagenesis followed by a siliceous epigenesis and bearing a cortex.

data will be used for the study of complete archaeological collections without subsampling, allowing the identification of both common and rare materials. By producing new knowledge on these rocks and creating a reference collection, we hope to contribute to the wider archaeological community's understanding of the Caribbean's pre-Columbian past.

Method

The petrological characterization of geological samples of silicites follows the method developed by P. Fernandes and J.-P. Raynal (Fernandes & Raynal, 2006). This method involves filling out three observation and description grids, which were developed based on the concept of the evolutionary chain of silicites (Delvigne, 2016; Delvigne et al., 2020, 2021; Fernandes, 2012; Thiry et al., 2014). This concept was formalized following the observation of the evolution and transformation of this type of material during their non-anthropogenic path, particularly after their genesis. Indeed, after being brought to the surface, they undergo mechanical and/or chemical alterations that are characteristic of the different environments they crossed. The goal of petrological characterization is therefore to identify the different processes at the origin of such alteration. It should be noted that petrology (i.e., the study of rock evolution) is coupled with petrographic analysis, which aims to describe the composition and structure of silicites, allowing for the distinction of genetic types. All observations carried out using exoscopy at low magnification (50x to 200x), with the sample under a slice of water, were recorded in a data table, which was then synthesized into a summary table. This table lists the featured elements (authigenic, non-organic, and organic), as well as inherited structures from the protolith (thus pre-genetic), during siliceous diagenesis, and during different moments of alteration (thus post-genetic). This very detailed description is necessary for identification of archaeological artifacts and possibility of replication by other archaeologists. Applied to the archaeological record, this method also makes it possible to differentiate the alterations undergone before the collection by the prehistoric collectives (predepositional), from those undergone during the burial phase in the archaeological site (postdepositional). In this way, it is possible to determine the location of the outcrop collected by the prehistoric groups, also specifying the type of deposit exploited (primary or secondary).

Non-destructive elemental analysis by PIXE (Particle Induced X-Ray Emission) was performed on the AGLAE facility (Calligaro et al., 2002) at the Centre de Recherche et de Restauration des Musées de France (Paris) using the procedures presented in Bellot-Gurlet *et al.* (1999). This analysis uses a proton beam to stimulate the X-ray fluorescence of most major elements and

several trace elements in the sample. Raman spectroscopy was performed on minerals visible on some artifacts' surfaces with a Bruker SENTERRA Raman microspectrometer equipped with a 532 nm laser, and a 1200-line diffraction grating resulting in a resolution of 3 to 5 cm^{-1} . Portable XRF analyses were performed with a Bruker Tracer 5i, equipped with a SDD detector and a 4W rhodium X-ray tube. Measurements are carried out in two modes of tube operation: a mode at 15 kV and 100 μA to analyze light elements (aluminum to manganese) and a mode at 45 kV and 64 μA to analyze heavier elements (iron to zirconium). Analyses were carried out on the standards and on the artifacts using a collimator to reduce the beam diameter to 3 mm.

Results

Reference collection and preliminary archaeological study for silicites.

The analysis of the reference collection created by one of us (CS) and the samples we collected during our field mission allowed for the identification of 24 types of raw materials. They can be grouped in several categories: orthoquartzite (Figure 1a-c), hypersiliceous volcanic rocks (Figure 1d-f), silicified cinerite, hydrothermal siliceous rocks (Figure 1g-i), radiolarite, agate and silicified coral. These types have been described thoroughly and photographed, so that future research can be based on these documents, either by our team or other researchers.

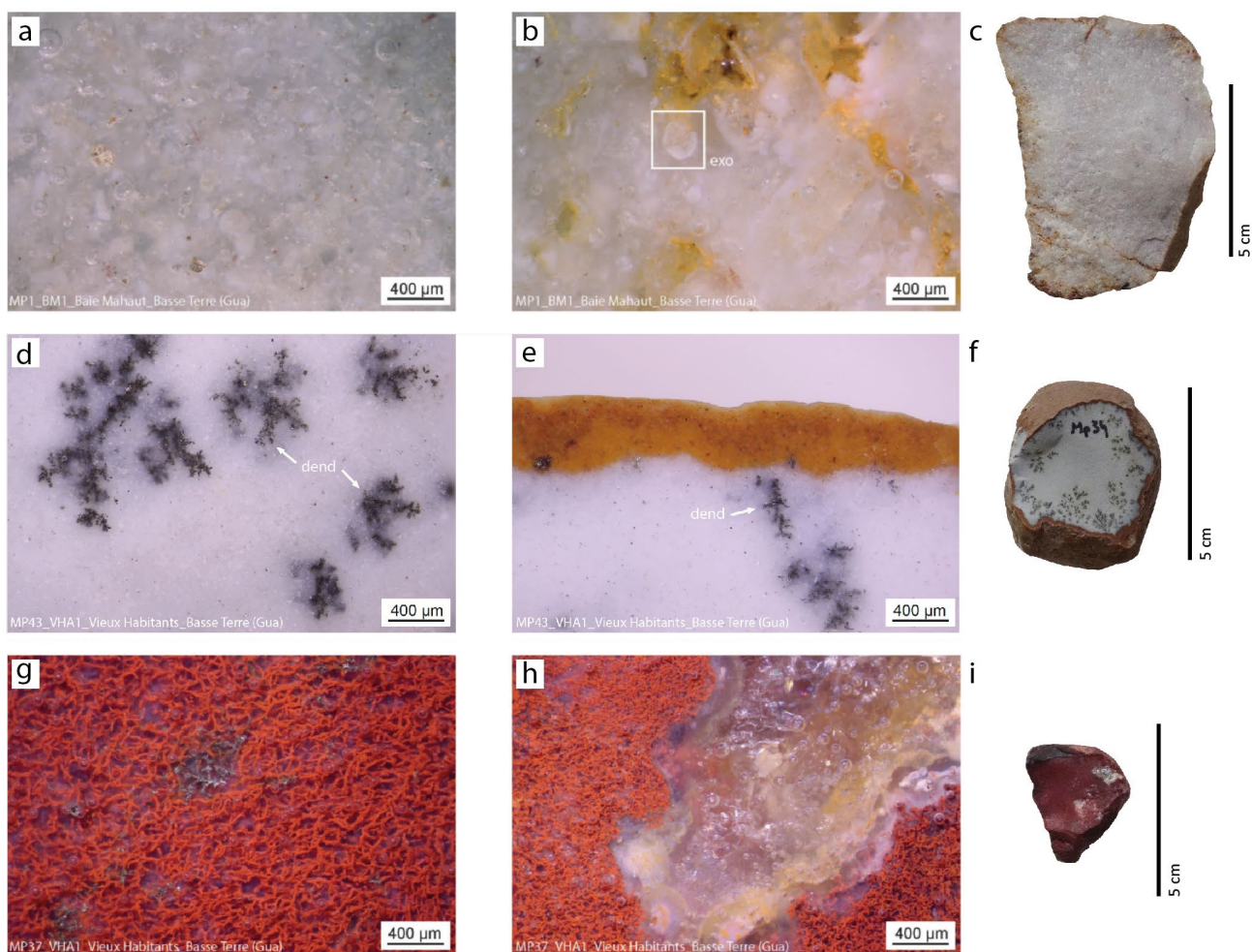


Figure 1. Examples of microphotographs from several types of silicites from Guadeloupe, showing internal texture, presence of featured elements like manages dendrites (dend) or exoclasts (exo).

Alongside our efforts to complete and describe the reference samples, we conducted a preliminary study of the archaeological site of Stade José Bade located in Marie-Galante (Serrand, 2016). Due to time constraints during our first mission, this first study is a first approach to a re-examination of the collection. This collection had already been studied, without extensive description and binocular observations. This initial work identified eight different types of siliceous raw materials. Upon the application of the methodology by trained specialists, and with time dedicated to binocular observations, the same collection of artifacts was robustly described as composed by raw materials belonging to four types, one of which consists of two sub-types. These four types are notably different from the eight categories previously identified, demonstrating the importance of using such a methodology (Figure 2). The detailed description of the artifacts from this site will also allow later identification when we update the reference collection with new raw materials.

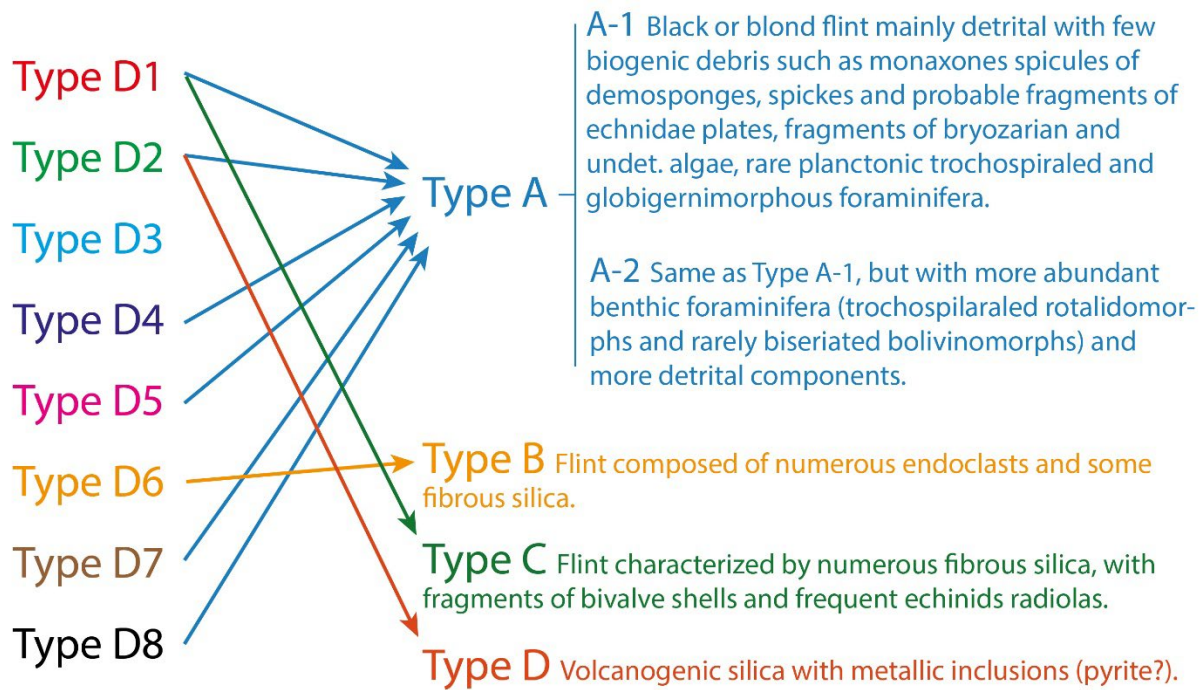


Figure 2. Comparison of silicite identification on the same archaeological collection by two separate specialists with different methodologies and experience.

In the end, the largely dominant Type A was attributed to Antigua’s flint, more precisely to the “distal part of the lagoon” (Type A1) and the “entrance of lagoon” (Type A2) facies of Flinty Bay. While two other types of flint were identified, their sources are still unknown and will hopefully be recognized later in the project. Additionally, we described one type of lithic material that is likely of volcanogenic origin. This preliminary study demonstrates the significant contributions that trained specialists can make to the field.

Several islands in the Caribbean (Figure 3), if not most of them, display chert, flint, jaspers or other siliceous rocks described in the archaeological or geological literature (Bérard, 2004; Donovan & Harper, 2005; Knippenberg, 2007; Pantel, 1988; Ray, 2015; Stouvenot, 2003). Our project aims to study these materials as well as archaeological collections. Another future goal for this long-term project is to expand into the Greater Antilles, a region with a complex geological history with great potential to yield a variety of unique materials.

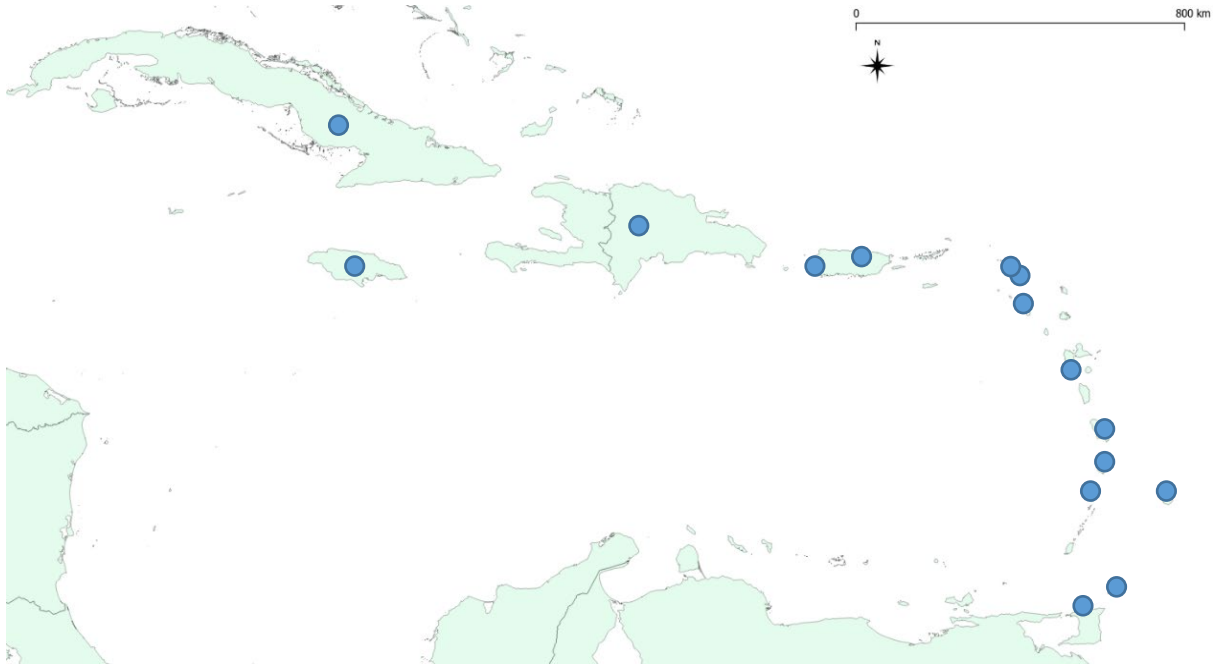


Figure 3. Natural occurrence of silicites mentioned in the literature. Only one point has been located on each island of the Greater Antilles even if more occurrences exist on each of these islands.

Volcanic glass identification in the Caribbean archaeological record

Despite the widely accepted belief that obsidian does not occur naturally in the Caribbean nor is found in the archaeological record, we present here evidence of the contrary. We identified twenty artifacts in the literature, recovered from seven archaeological sites spanning Early to Late Ceramic age contexts and located on four islands (Table 1). We were able to analyze sixteen of them (Figure 4). In the Lesser Antilles, only one potential sources of glassy material (i.e., obsidian), is known: a limited outcrop integrated in the “Volcan du Tuf” (Boudon et al., 1987; Gadalia et al., 1988; Gadalia & Westercamp, 1984). It is located a few kilometers from the archaeological site of Ruelle Roches Caraïbes.

Site	Island	Longitude	Latitude	Nb artifacts	Period	Reference
Bisdary	Basse-Terre	-61.709	15.988	5*	Early Ceramic	(Romon et al., 2006)
Ruelle Roches Caraïbes	Basse-Terre	-61.736	16.038	2	Early Ceramic	(Bonnissent & Chancerel, 2001)
Tourlourous	Marie-Galante	-61.224	15.889	7*	Early Ceramic	(Colas, 2002)
Stade José Bade	Marie-Galante	-61.224	15.889	1	Early Ceramic	(Serrand, 2016)
Place St. François	Basse-Terre	-61.73	15.995	1*	Early Ceramic	(Sellier-Ségard, 2001)
Caille à Bélasse	Ilets de Petite Terre	-61.11	16.17	2	Late Ceramic	(Gagnepain, 2007)
Del-3	Dominica	-61.256	15.275	1	Early Ceramic	(Shearn, 2014)
Mill Reef+	Antigua	-61.679	17.057	1	Early Ceramic	(Murphy, 1999)

Table 1. List of archaeological sites with volcanic glass artifacts (+: yet to be confirmed) and their location. *:

one of the samples analyzed in 2007 was missing in 2021.

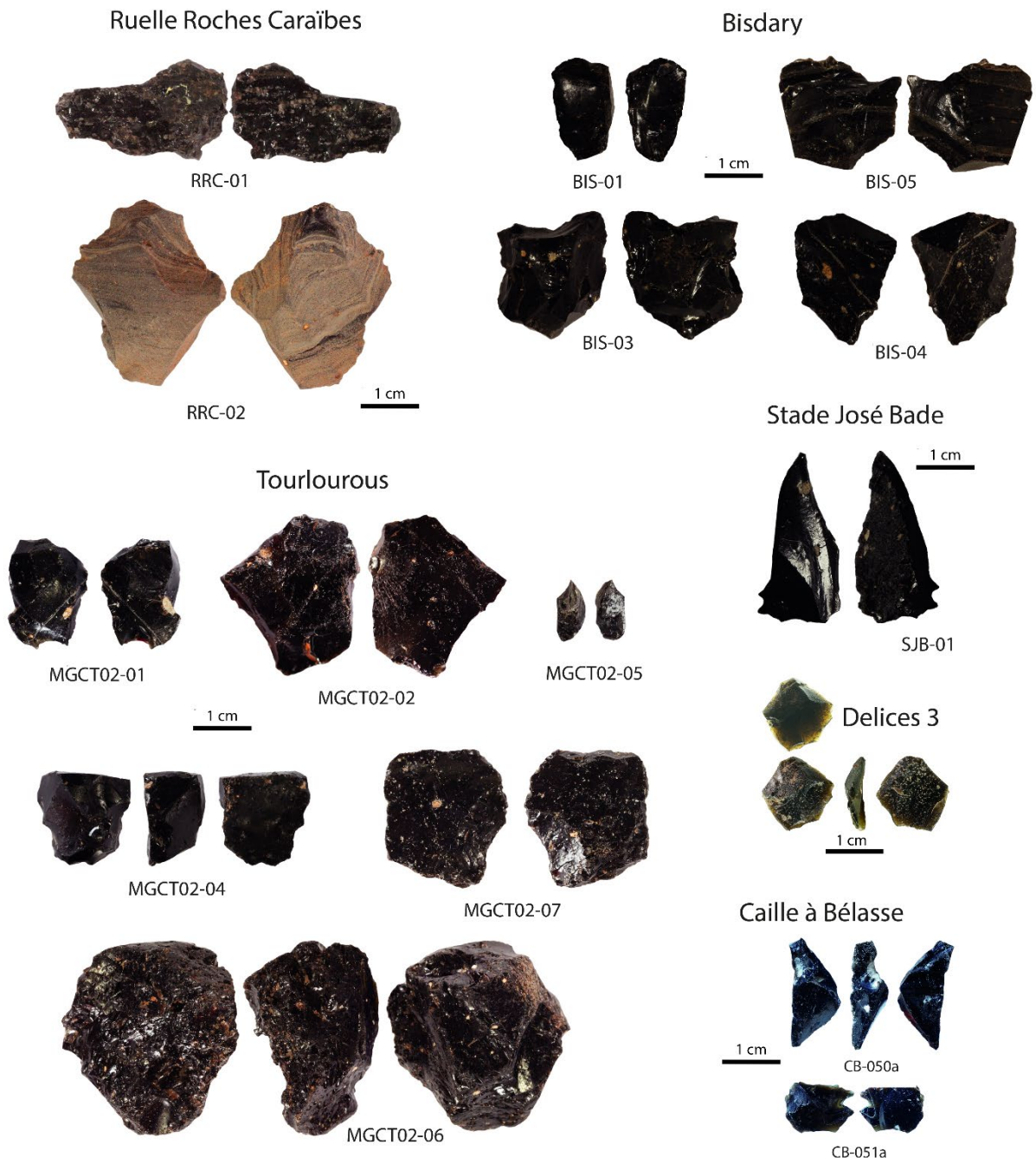


Figure 4. Macrophotographs of the sixteen glassy artifacts recovered from six archaeological sites.

In 2007, a preliminary non-invasive PIXE analysis was conducted on 16 artifacts and one sample from the “Volcan du Tuf” (Bressy & Bellot-Gurlet, 2007). Subsequently, additional analytical techniques including photographs, microphotographs, pXRF analysis, and Raman spectroscopy were employed in 2019 and 2020 to investigate further most of these artifacts and some discovered after the initial study. The results indicated that nearly all the samples had a comparable composition, which was not consistent with the typical rhyolitic composition of obsidian. Rather, their low silica content classified them as andesitic volcanic glass (Figure 5), and their highly similar composition for minor and trace element suggests that the samples likely share a common origin. The two most recent discoveries – Stade José Bade and Delices (Del-3) – showed the same “non-rhyolitic” chemical composition based on pXRF analysis. The unique artifact made of obsidian *sensu stricto* (RRC-01) originated directly from the “Volcan du Tuf”, as underlined by their common chemical composition. It was excavated at the Ruelle Roches Caraïbes site, only a few kilometers away from the source.

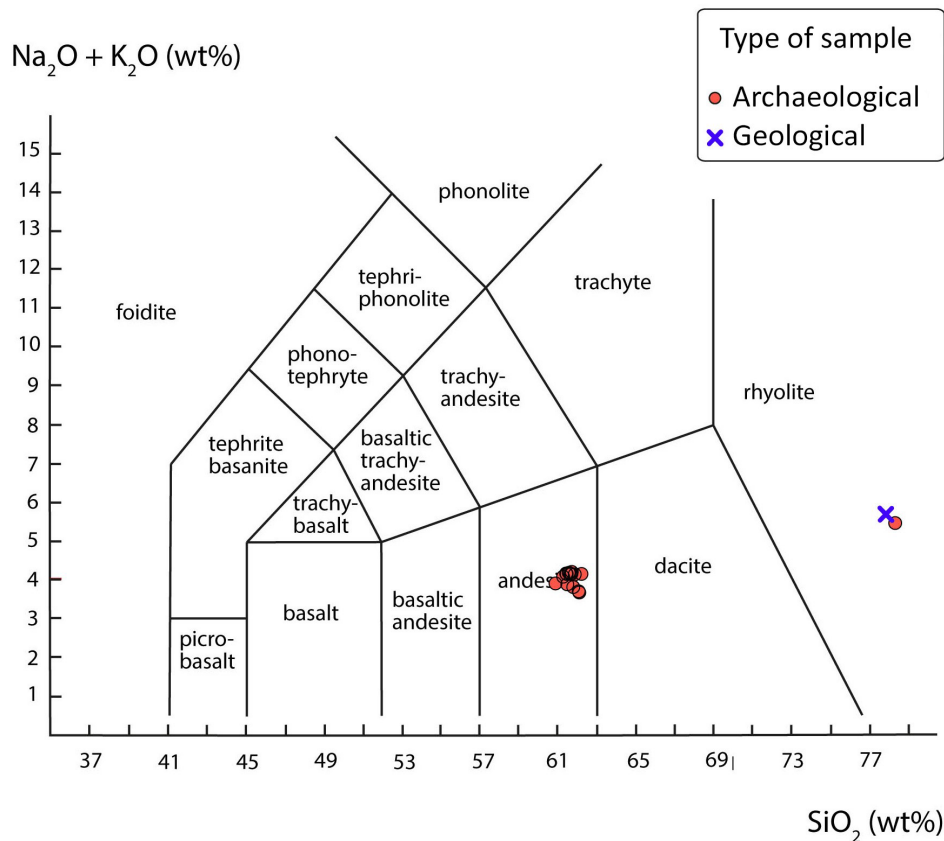


Figure 5. Total Alkali Silica (TAS) diagram (mod. after Le Maitre et al., 2005) from the PIXE analysis, showing that most samples are andesitic glasses, while one of the artifacts (RRC-01) and the geological sample of glass from “Volcan du Tuf” are of rhyolitic composition.

The origin of the remaining 17 samples (15 analyzed by PIXE and 2 by pXRF) still remains unknown. The andesitic chemical composition of the samples is consistent with a Lesser Antilles origin given that most of the recent volcanic arc consists of andesitic rocks. However, our global scale literature review did not yield a single reference to natural andesitic glass suitable for knapping, either in the geological or archaeological domains, except for rocks originating from asteroid impacts (Rochette et al., 2021). Raman spectroscopy analysis of minerals inclusions identified some plagioclase feldspars and augite (Figure 6), both standard minerals of andesites but that can be also present in impactites (French, 1998). Additional analyses are currently underway to further investigate the cosmic hypothesis.

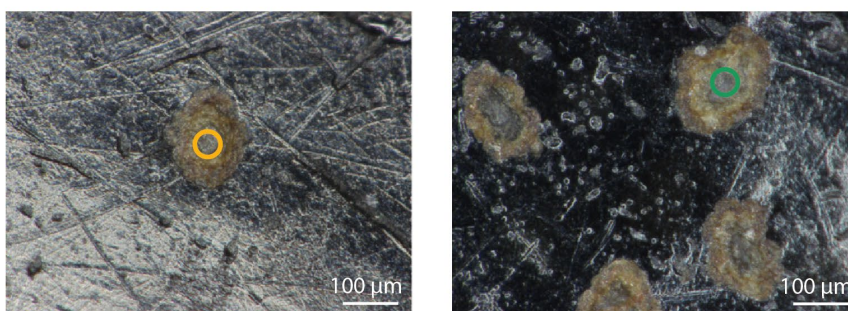
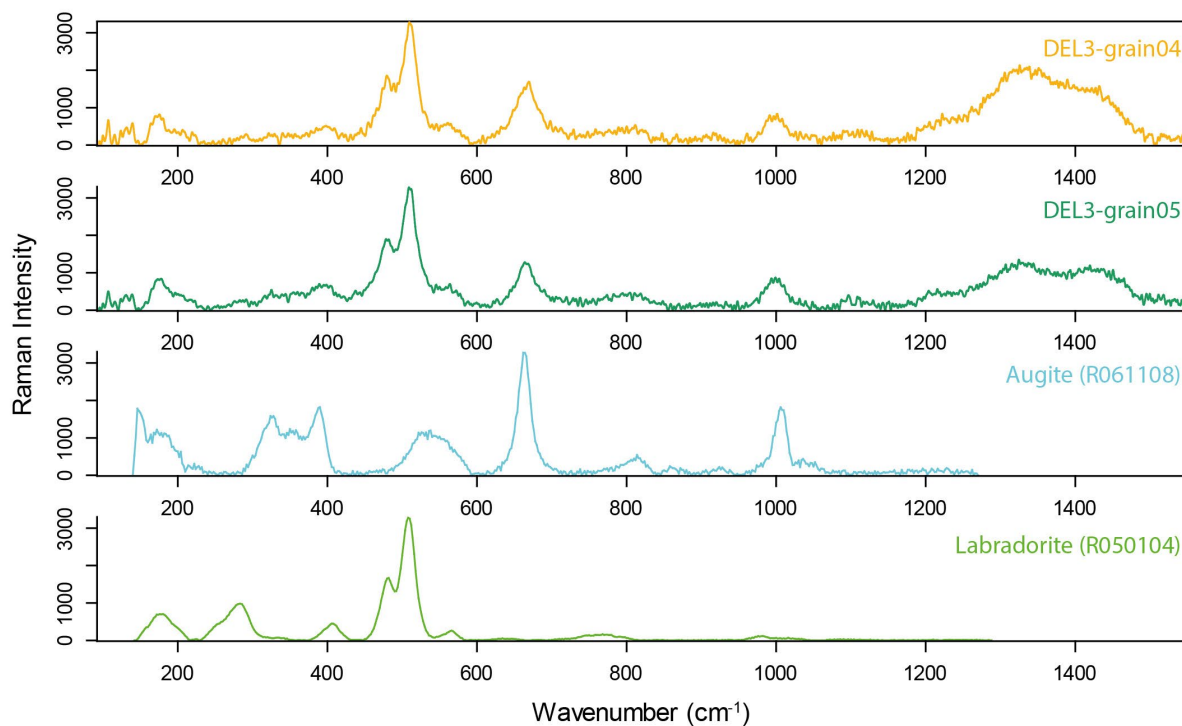


Figure 6. Mineral identification by Raman spectroscopy of the crystals included in the Del-3 sample (top two spectra). The minerals are identified as labradorite (a plagioclase feldspar) and augite (a clinopyroxene) by comparison with the RRUFF database (bottom two spectra).

Although the provenance of the glassy material in question remains unknown, we conducted a comprehensive survey of its geographic distribution throughout the Caribbean islands. This inquiry led us to investigate relevant literature, notwithstanding the widely held notion that obsidian is not present in this region. The samples listed in the literature from Maruca and Maisabel, in Puerto Rico, are probably not obsidian (Pers. Comm Rodriguez-Ramos 2021, Siegel 2021). One sample is listed in R. Murphy's dissertation (Murphy, 1999), and we aspire to examine it in the future. Some samples are listed in the site of Fond Brûlé in Martinique (Mattioni, 1982), but these were mistakenly identified². Despite the scrutiny of existing

² After careful examination of the complete collection from this site, we can conclude that these flakes and nucleus are not made of volcanic glass.

literature, discussions with numerous Caribbean archaeologists, and exchanges held during the IACA congress in Cuba, the only volcanic glass artifact that we could identify outside French islands was the flake from Delices (Del-3) in Dominica (Shearn, 2014).

The other magmatic rock that we aim to study for provenance studies is the diorite. This distinctive black and white rock was widely used for lapidary production during Saladoid and Post-Saladoid periods, all over the archipelago (Figure 7). Few sources are mentioned in the literature, mainly at Tobago (Mones, 2007) and Puerto Rico (Turner et al., 2003), but other samples from La Désirade, Saint-Martin, British Virgin Islands, and French Guyana will be also integrated as references. Despite its granular texture, which induces a certain analytical challenges, petrographic observations coupled with geochemical analyses using X-ray fluorescence and X-ray spectroscopy in a SEM have been successfully employed in recent studies (e.g., Burg et al., 2021; Turbay et al., 2019).

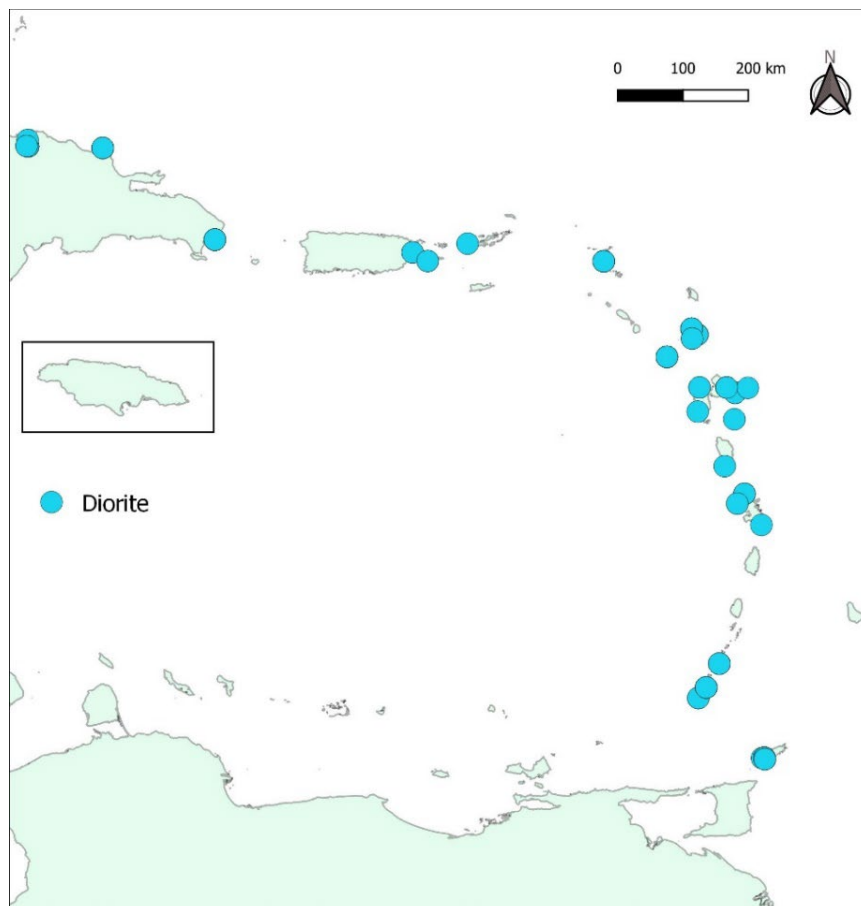


Figure 7. Map of the archaeological distribution of diorite beads and pendants during the Ceramic age in the Caribbean islands (mod. after Queffelec, 2022).

Sourcing of turquoise by non-destructive isotopic analysis

Turquoise, another rock used for lapidary production during the Early and Middle Ceramic periods, is also studied in this project. This gemstone is well distributed among the Lesser Antilles, even if always as small quantity in the sites (Figure 8). The source of this material, on the other hand, is unknown. The well-known sources of turquoise are located far in the American Cordillera, both in North and South America (Queffelec, 2021), while a source is mentioned in coastal Brazil (Costa et al., 2004), and a single source is known so far in the Caribbean, in the US Virgin Islands (Alminas et al., 1994; Tucker et al., 1985).

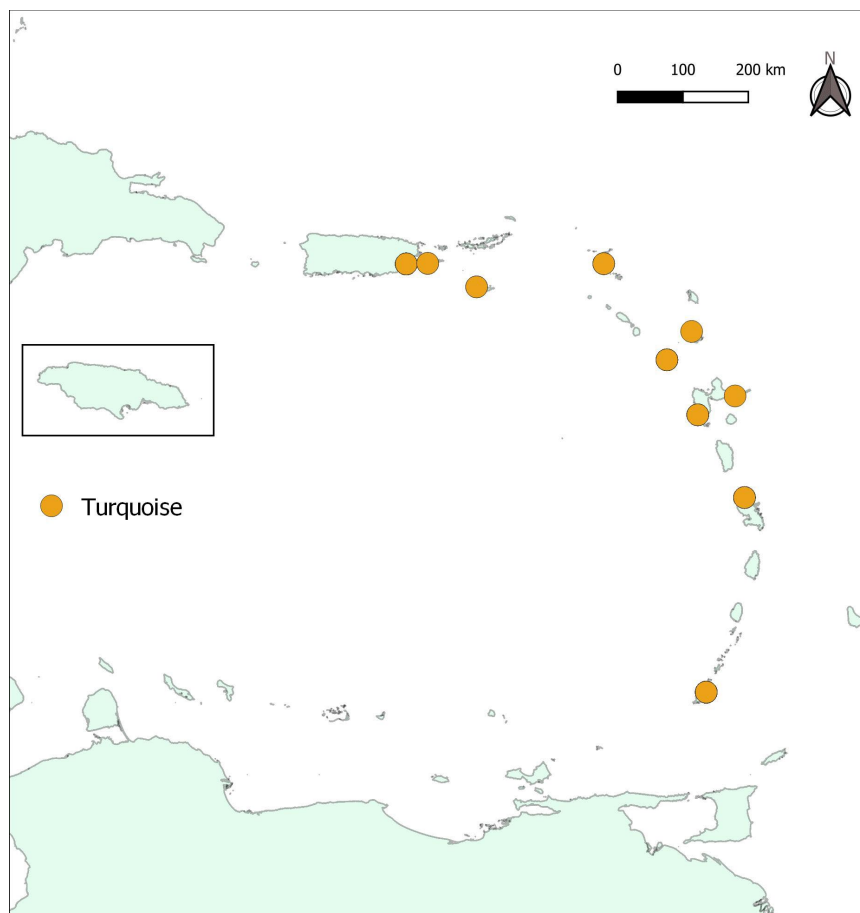


Figure 8. Distribution of the turquoise artifacts in the Caribbean archipelago (mod. after Queffelec, 2022).

The application of non-destructive isotopic analysis of copper, as proposed by Hull *et al.* (2008), and of lead, as measured destructively by Thibodeau *et al.* (2012), is not straightforward to implement without risk for the analyzed artifacts and with good reproducibility. We are presently in the process of creating internal standards for isotopic analysis by laser ablation mass spectrometry, to avoid the rather invasive sample preparation of SIMS analysis (Secondary Ion Mass Spectrometry).

Discussion and conclusion

The creation of a reference collection of silicites, and its characterization through microscopic observation, started recently in Guadeloupe and will continue in Martinique, hopefully gathering raw materials from other islands as well. Microscopic observation will be used to characterize the archaeological collections too, building upon a methodology established in continental France over the past decade that has successfully identified and described dozens of different types of lithic raw materials. This approach will prove invaluable in describing the range of raw materials encountered in archaeological contexts. A very preliminary study of an archaeological collection from the Saladoid period (Stade José Bade) already demonstrated the applicability and utility of this method. Future applications to archaeological collections from different islands and periods will rely on these detailed descriptions, enabling the identification of raw materials beyond the sole source of flint on Antigua.

This study also sheds light on the presence of volcanic glass artifacts in the region, which was previously unknown. The study reveals two significant observations: i) Caribbean people from the Saladoid period, living in present-day Basse-Terre, used a local volcanic glass, found today in the “Volcan du Tuf”, and ii) Caribbean people from Basse-Terre, Marie-Galante, and Dominica, during this same period, used a unique and hitherto unknown andesitic glass to produce small flakes through bipolar debitage. Two of these flakes have also been found in Troumassoid context on the Ilets de Petite Terre, in an unfortunately unpublished excavation.

The creation of a reference collection of diorites – a rock used for lapidary production on many islands spanning all Ceramic age – will be also pursued in the coming years. Development of the analytical methodology for sourcing such coarse-grained rocks will be needed.

The project is also working towards sourcing turquoise, with a focus on developing a non-destructive method for analyzing the isotopic composition of this mineral. The goal is to create a methodology that can be used on archaeological artifacts such as small beads and pendants from the Early and Middle Ceramic periods in the Lesser Antilles.

The creation of lithic raw material reference collection for all the Caribbean archipelago is a long-term project, and so will be its use for accurate provenance studies of lithic artifacts. The development of such a database for silicites, and its comprehensive and detailed description, will also be helpful for the community of Caribbean archaeologists, who can hopefully apply this method to their own studies. By combining the different lithic raw materials, we aim at

determining the litho-space of Caribbean communities during the Ceramic age. This will guide a more general reflection on the circulation of materials in the Caribbean islands.

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