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### Characterization and geological context of the silex used as raw material for archaeological artifacts in the Taió region - Santa Catarina, southern Brazil

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#### Abstract

The town of Taió is home to archaeological sites, including the INDUMA SC-TA-19 site studied by the Anchieta Research Institute, which cataloged various lithic artifacts made from silex (flint). According to the different authors, silex is a rocky material with controversial formation processes in the literature. In this context, this work aimed to characterize these silex artifacts, define their source area and geological origin, based on field studies, optical petrography, scanning electron microscopy, and semi-quantitative chemical analysis. The methodology employed allowed characterizing its source area, defining that silex consists of bioclasts, wrapped in silica polymorphs, combined with fibrous chalcedony, in different proportions and establishing its formation in a marine environment.

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#### 1. Introduction

With the advance of petrographic studies, the analysis of origin of lithic raw materials in archaeological studies has become an important tool for understanding certain characteristics of prehistoric societies (Fernandes and Raynal 2006). These studies are part of Geoarchaeology, which according to Waters (1996), is based on archaeological research and focuses on the interaction between paleoecology, geomorphology, geology and pedology (Rubin and Silva 2008).

Due to its characteristics, silex is one of the most commonly used raw materials in the manufacture of artifacts in prehistoric societies, and for this reason it is widely studied. However, Fernandes et al. (2007) highlight that there are problems to be investigated further since the study of silex has produced different results in view of the difficulties of macroscopic examination, optical microscopy, scanning electron microscopy and the determination of its chemical composition.

Prospective studies and excavations conducted by the Anchieta Research Institute (IAP) in the Taió region of Santa Catarina, determined that the main lithic matter in the making

of archaeological artifacts was silex. However, such studies do not provide detailed analysis of this material or its source area (Schmitz et al. 2009). In order to analyze further this topic, this study aims to describe mineralogically and petrographically the silex, raw material of lithic artifacts, and relate it to its geological context. This work builds on pre-existing studies of the geological context in the Taió region, especially in the field of petrography, expanding knowledge of the silex used by the prehistoric society that occupied this region.

The municipality of Taió, where this research was conducted (Figure 1), is located at the coordinates UTM 22J 599296 mE and 7000213 mN (WGS-84), in the region of Vale do Itajaí in the state of Santa Catarina, southern Brazil. Access to the area from the capital Florianópolis is via the BR-101 towards the north of the state, at approximate distance of 95 km from the municipality of Itajaí, then 200 km on the BR-470 towards the southwest until the junction with the SC-422 highway and further 4 km to the center of Taió municipality.

The municipality is located on the eastern slope of the Southern Plateau, at altitudes between 300 and 700 m, and the sites studied by the IAP are both in the valley, between 300



and 400 m, and above the 600 m level, where underground houses characteristic to the Southern Jê people are found (Schmitz et al. 2009).

In the studied area, the geological units of the Paraná Basin are found, according to the Santa Catarina State Geological Map scale 1: 500,000 (Silva and Bortoluzzi 1987) (Figure 1). The municipality of Taió is in the domain of the Paraná Basin, including glaciogenic sedimentary rocks of the Rio do Sul Formation (Itararé Group), shale and sandstones of the Rio do Rasto Formation (Passa Dois Group) as well as basic intrusive rocks of the Serra Geral Formation, in addition to quaternary continental sediments.

## 2. Materials and methods

Initially, a survey and compilation of pre-existing data was performed, as well as a macroscopic analysis of the archaeological artifacts archived in the IAP, from the archaeological site INDUMA SC-TA-19, chosen as the object of this research. The study area polygon was defined using the ArcMap 10 and AutoCAD Map 3D softwares.

Two field stages were performed in the study area, when samples were collected for macroscopic and petrographic analysis, aiming to analyze local geology and to define the source area of the artifact's material.

The macroscopic analysis, performed with the naked eye and with binocular magnifying glass, of both artifacts and outcrop samples, 10% HCl solution was used. Subsequently, the petrographic analysis was performed, one of the field-collected material (rock sample taken from the outcrop layer called AM-01 with similar characteristics to the artifact samples provided by the IAP) and three of the samples provided by the IAP (2250, 2291 and 2574). These analyzes were performed using ZEISS AX10LAB petrographic microscopes at the Microscopy Laboratory of the University of the Vale do Rio dos Sinos (UNISINOS), in São Leopoldo, RS, Brazil. At this stage it was possible to describe physical, mineralogical and optical properties of silex.

After petrography, the selected samples were submitted to the Scanning Electron Microscope (SEM - ZEISS brand, model EVO/MA15) of the Technological Institute of Micropaleontology (ITT / Fossil) of UNISINOS. The analyzes were performed on

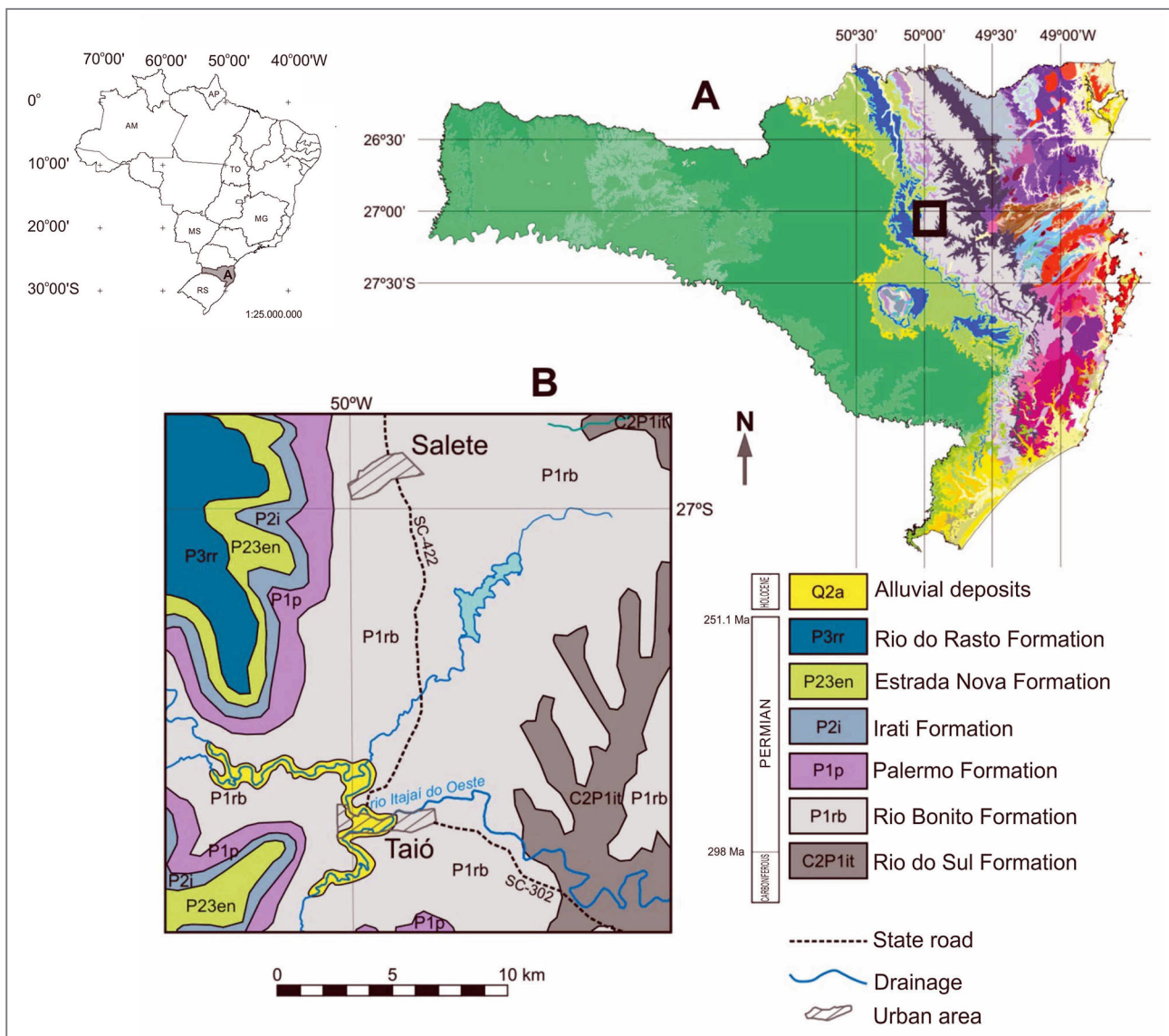


FIGURE 1 – Location of the study area, A) Geological Map of Santa Catarina (modified by Silva and Bortoluzzi 1987); B) Geology detail of the area.

a sample of artifact 2325, on the AM-01 outcrop sample and on thin sections (IAP 2291 and 2250 samples). The samples were plated with gold and palladium on aluminum support prior to analysis. The chemical characterization of the artifacts and rock samples was performed by means of chemical microanalysis with energy dispersion X-ray spectrometer (EDS) and chemical microanalysis with wavelength dispersion spectrometer (WDS).

The data obtained in the field stage were compared with laboratory data, which made it possible to formulate hypotheses about the geological context of the silex source in the Taió region.

### 3. Results

#### 3.1 Field Geology

During the two field trips to and around Taió, outcrops of all lithostratigraphic units mapped in the Santa Catarina State Geological Map scale 1: 500,000 (Silva and Bortoluzzi 1987) were recorded in Figure 1.

The search for the source area of the silex used in the lithic artifacts began with the first field trip in March 2013, which covered the northeast of Taió, at low altimetric elevations, near the INDUMA SC-TA-19 archaeological site. However, this lithology was not found in the outcrops visited. The INDUMA SC-TA-19 site is approximately 2 km from the INDUMA SC-TA-04 archaeological site: 22J 602441 mE / 7009043 mN, datum WGS-84, at the headquarters of the INDUMA farm (Schmitz et al. 2009).

During the second field trip, which took place in May 2013, we visited the localities in the northwest of Taió, in the highest altitudes of the municipality, near other archaeological sites studied by the IAP, where a single outcrop of lithostratigraphic units of the Paraná Basin was observed. It was quite altered, with the presence of a rock with macroscopic characteristics similar to those of the raw material used to make the archaeological artifacts found by the IAP in INDUMA SC-TA-19.

This outcrop had an approximate length of 70 m and is located on the access road of Ribeirão Cipriano, in the municipality of Saleté, north of Taió (UTM coordinates: 22J 594862 mE / 7013119 mN, datum WGS84. Figure 2A schematically summarizes the outcrop lithologies, including the altered silex layer.

In this outcrop a schematic vertical profile was performed, represented in Figure 2B, where the thickness of the lithological layers was measured, and the silex layer did not exceed 35 cm in cross-section. It was observed that in the contact of the silex layer and the gray colored carbonate silt, the layer attitude exhibits a horizontal behavior.

#### 3.2. Macroscopic Analysis

Samples from the IAP collection (Figure 3 B), from the Paleo-archaeological Museum of Taió and those collected in field were analyzed macroscopically.

Macroscopic analysis of both rock and artifacts found at the site showed that the material consists of dark gray to black opaque silex in the fresh fracture, turning yellowish to whitish in the natural fracture when weathered (Figure 3A). In this case, they show a layer of millimeter to submillimeter

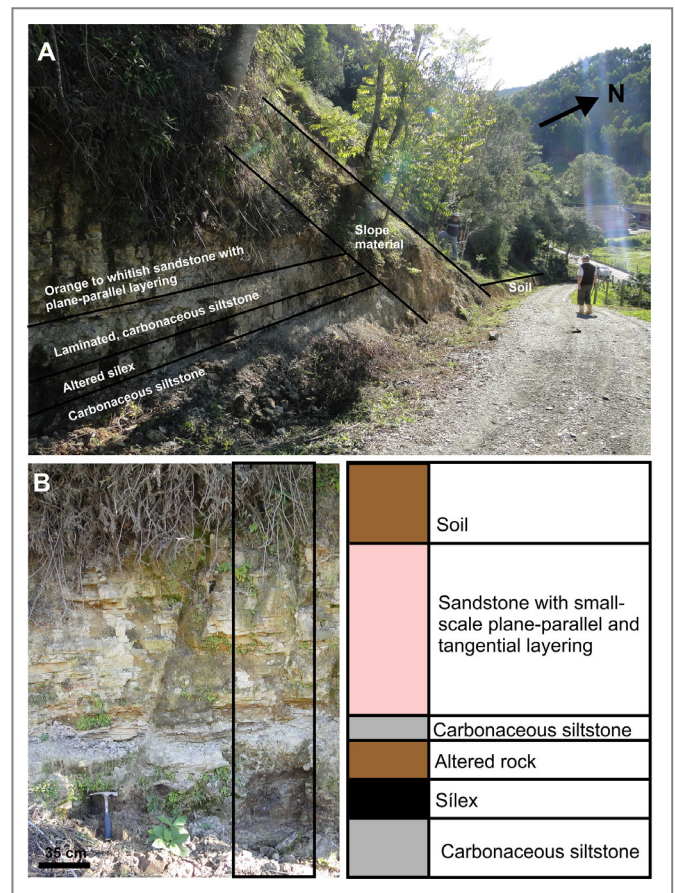


FIGURE 2 – A) Outcrop AM-01, in Ribeirão Cipriano, Saleté Municipality; B) Detailed profile of point AM-01.

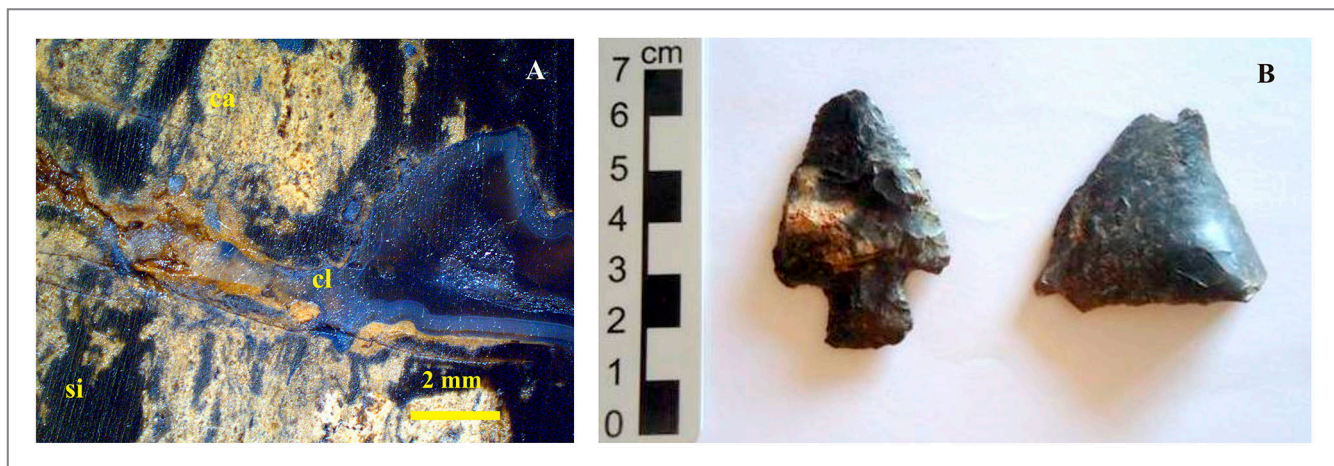
thickness with a powdery appearance, in the literature named *patina*, since it originated after the manufacture of the artifact (Almeida et al. 2003). The texture is aphanitic, and the structure is heterogeneous, particularly in the field sample, which has silex domains interspersed with irregular chalcedony and carbonate domains, recognized based on the reaction with 10% HCl.

The samples have good suitability for cutting with conchoidal fracture resulting in smooth and sharp surfaces. The glow is semi-glassy similar to chalcedony (cryptocrystalline quartz variety), the degree of hardness is high, greater than 6.5, as the material cannot be scratched with a stainless-steel tip and easily scratches the glass. Artifacts and chips reacted negatively to 10% HCl, indicating the absence of calcium carbonate. However, the field-collected rock sample had clear, more altered, powder-like domains that react weakly with 10% HCl indicating the presence of calcite, possibly characterizing what the literature calls the cortex. This change, in fact, may be a residue of the carbonate composition protolith only that later underwent diagenesis, with carbonate replaced by silica.

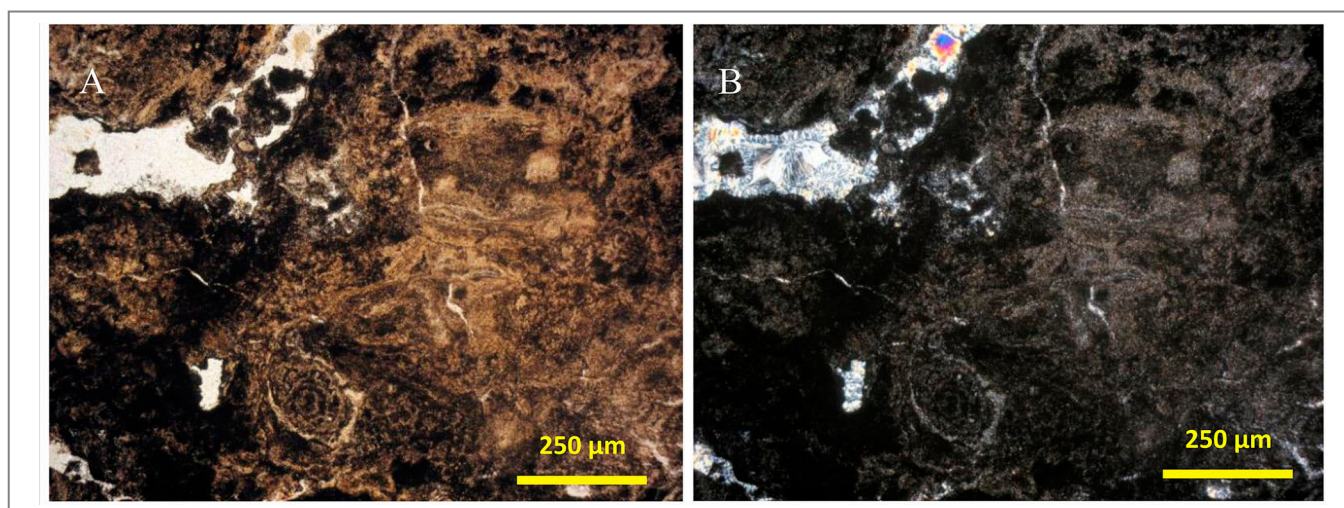
#### 3.3. Petrographic Analysis

The thin sections of the AM-01 field sample and the IAP samples (2250, 2291 and 2574) presented what the literature calls heterogeneous petrofabrics, that is, they consist of domains formed by abundant bioclasts surrounded by polymorphs of microcrystalline silica, which are amalgamated to interstitial, amoeboid domains, consisting of fibrous





**FIGURE 3** – A) Photograph of AM-01 showing the chalcedony (cl), silex (si) and carbonate (ca) domains (2 mm scale); B) Part of the archaeological artifacts belonging to the IAP collection that were analyzed.



**FIGURE 4** – Photomicrographs of sample 2291. A) Natural light photomicrograph showing domains of amorphous silica and fibro-radiated chalcedony. B) Photomicrograph identical to the previous one in polarized light.

chalcedony, sometimes radiated (Figures 4A and 4B).

Sample AM-01 occasionally contains some micritic carbonate, with rare calcite crystals smaller than 0.2 mm (200 μm), which stand out for their high relief and characteristic optical properties, as indicated by the slight reaction to 10% HCl obtained in the rock sample, amalgamated by subordinate chalcedony.

The domains of silica polymorphs are the most abundant in the studied samples composed of 60% to 95% of silica polymorphs and the remaining portion of the sample of chalcedony.

In general, these domains are irregular, yellow to black in natural light, with opaque concentrates, and possibly made up of opal (amorphous) and cryptocrystalline and microcrystalline silica, as they are weakly anisotropic in polarized light. Opaque mineral concentrates are possibly oxides or hydroxides of iron and/or organic matter.

In sample AM-01 the opaque dyes tint reddish part of the silica, having extremely fine grain, below the optical resolution limit. In sample 2250 the opaque portions are possibly goethite minerals. However, the presence of organic matter is not ruled out.

In the silica polymorph domains, there are concentrations of bioclasts of varying shapes and sizes, with samples AM-01 and 2250 showing the highest concentrations. The most abundant bioclasts are those of circular shape, of submillimeter dimensions, generally in the order of 0.05 mm (50 μm), marked by opaque edges, usually filled with chalcedony, occurring alone or grouped in pairs in AM-01 (Figures 5 A and B) or in the form of “grape clusters” in sample 2291 (Figures 6), normally non nucleated. These bioclasts must be carapaces and shells.

In sample AM-01, half-moon shaped bioclasts, marked by opaque edges and filled with chalcedony, are commonly recognized as bivalve fragments. They reach up to 0.5 mm (500 μm) in length and may occur alone or in pairs, joined at one end. In sample 2250 there are cylindrical shapes from elongated to curves, possibly resulting from the replacement of bioclasts with opaque material. These structures are distributed throughout the thin section, but may occur in higher concentration domains (Figures 7A, 7B and 7C).

The literature on the subject is not abundant, but Morgenstein (2006) describes lithic artifacts with pseudomorph goethite and maghemite on framboidal pyrite, which in turn



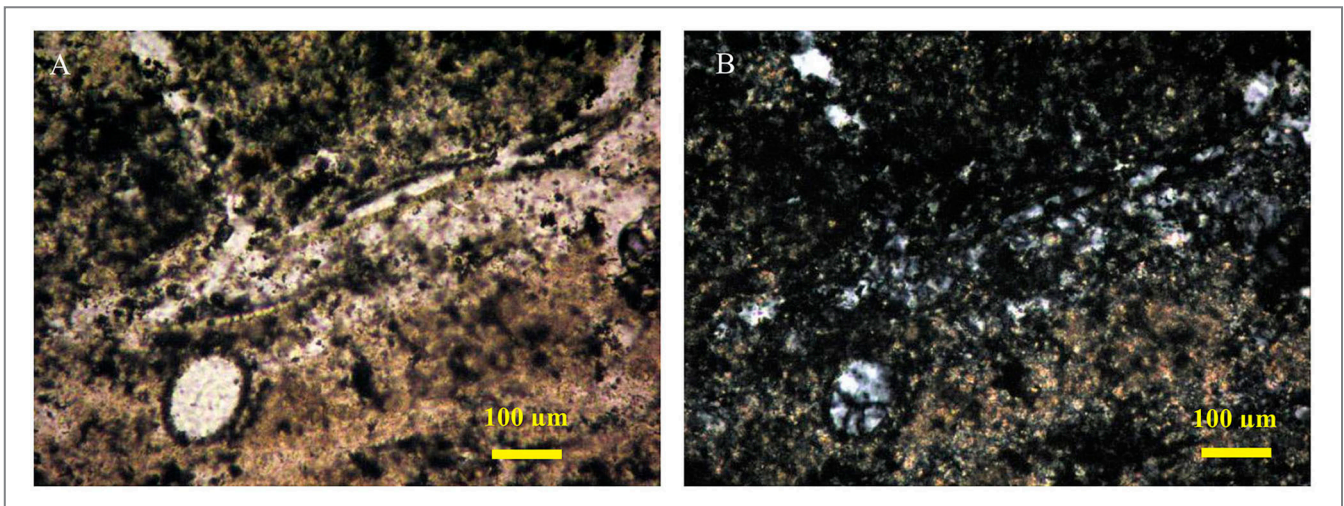


FIGURE 5 – Photomicrographs of sample AM-01. A) Photomicrograph showing circular and half-moon shaped bioclasts in natural light. B) Photomicrograph identical to the previous one, in polarized light showing the chalcedony filling of bioclasts.

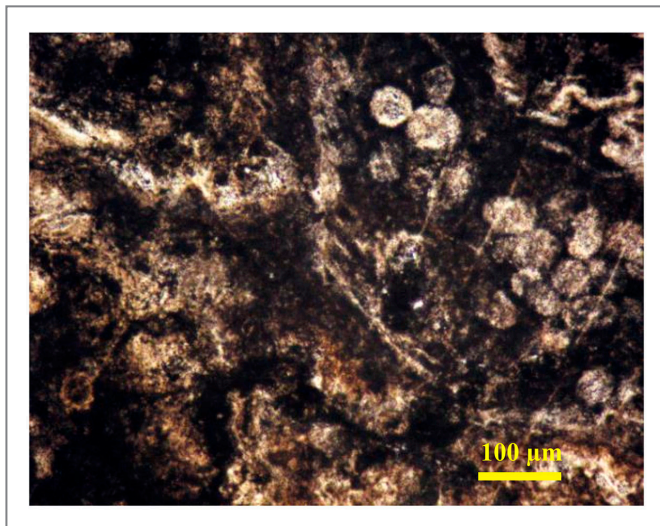


FIGURE 6 – Photomicrograph of sample 2291, detail of photomicrograph sampling circular bioclasts (Morgenstein 2006).

replace silex bioclasts, very similar to those found in sample 2250. The author attributes these bioclasts to foraminiferous carapaces and bivalve shells (Figure 7D). However, they also resemble cyanobacterial filaments.

Sample 2574 is the one with the most heterogeneous petrofabric of the analyzed group. Locally, it has gradual contact with a sedimentary host rock where silex fragments and/or portions partially mix with the protolith.

The sedimentary rock is composed of angular and low sphericity mineral fragments (quartz, white mica and zircon) with very fine grain size (less than 0.1 mm or 100 µm), and the material surrounding these grains is optically isotropic to weakly anisotropic, where sometimes some polymorph can be distinguished from silica (chalcedony?). Gradually, silex fragments increase in size and volume until they predominate. In this case, the silex is similar to the observed in other samples, with domains composed of silica polymorphs surrounded by fibrous chalcedony interstitial domains (Figures 8A and 8B). Chalcedony is perfectly colorless in natural light, featuring

low birefringence with first-order gray interference color in polarized light.

In the darker and isotropic domains again occur bioclasts of different shapes and sizes, with the current sample including the largest amount and more fragmented bioclasts. In all the samples some calcite crystals were observed in percentages below 1%.

The petrographic analysis allowed us to identify a biogenic sedimentary origin for the silex with which the artifacts were made, originating from the concentration of carapaces in the marine environment, as suggested by the presence of possible foraminifera, radiolaria and bivalves. It also allowed to link the artifacts to the silex outcrop found in the region, but the extremely fine grain of the samples did not allow the precise definition of the composition of amorphous to microcrystalline material as silica polymorphs. For this, different materials were examined with scanning electron microscopy, as discussed below.

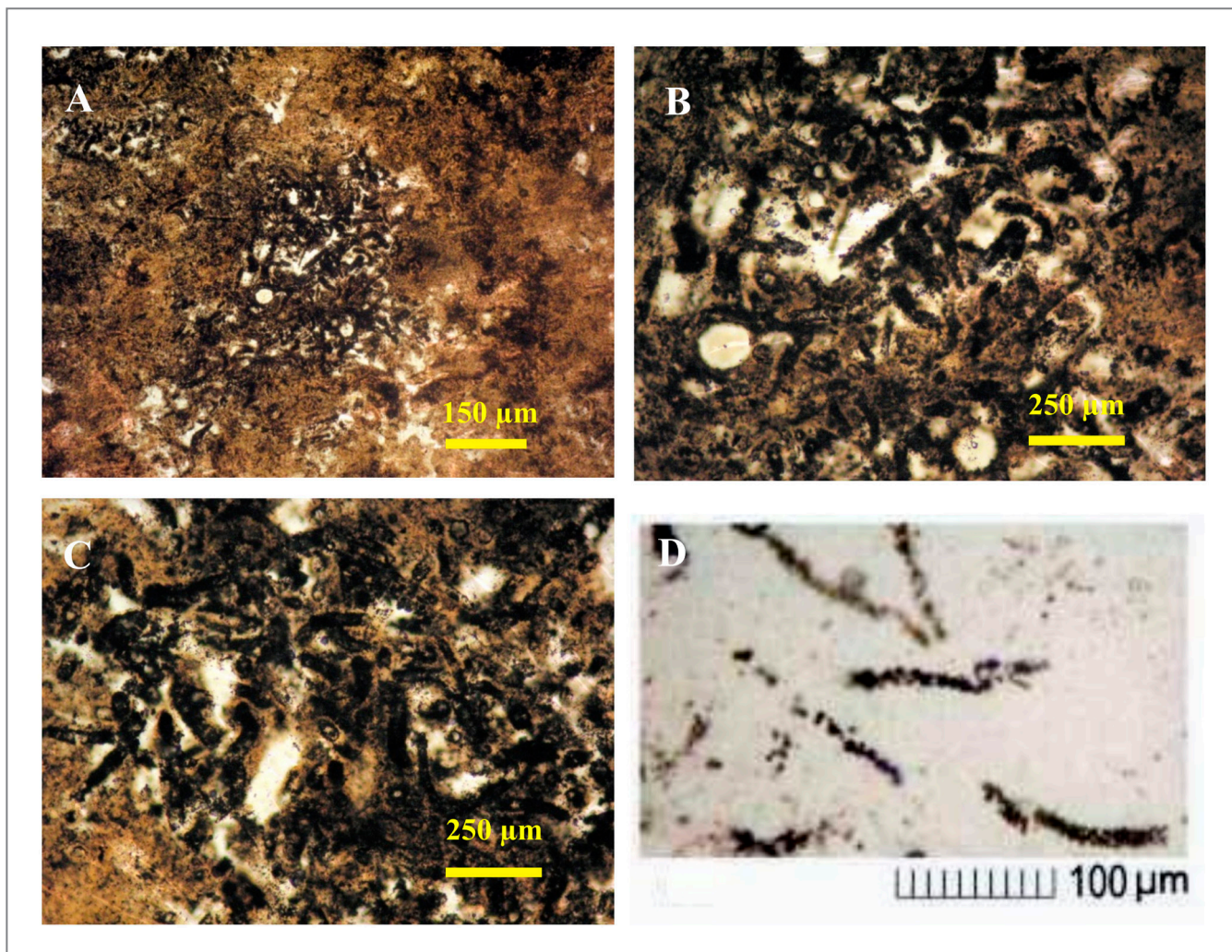
### 3.4. Scanning Electron Microscopy Analysis

Initially images were obtained from samples 2250 and 2291, where it was possible to observe that the material that constitutes the artifacts does not have a defined crystalline structure, because in the six observed points the structure remained botryoidal, typical of amorphous material (Figure 9), corroborating the observations made during the petrographic analysis.

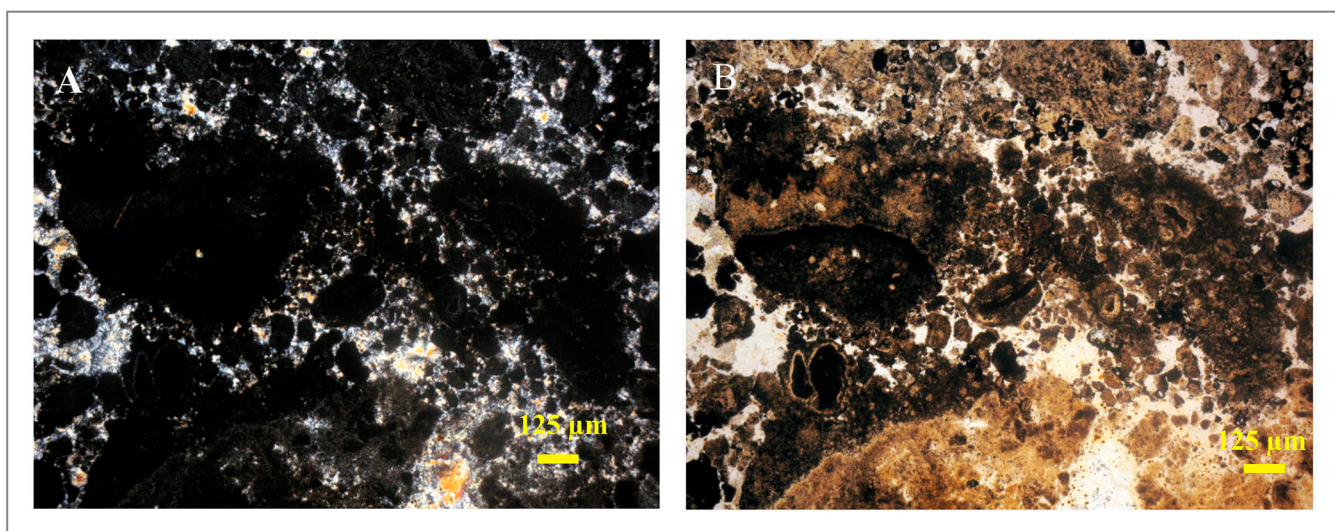
Once the structure was defined, the next step was to verify the chemical composition of the studied samples. In this respect, it was observed that all samples are formed almost exclusively of silica: in sample 2250 six points of chemical microanalysis with energy dispersion X-ray spectrometer (EDS) were made; in sample 2291, five points were performed; in sample AM-01 two points followed; and in the artifact fragment, also two points, all of which revealed to consist almost exclusively of silicon and oxygen, peaks of carbon and calcium occurred occasionally, indicating the presence of some carbonate and/or organic matter.

At some of these points, images of the element concentrations present were taken and the obtained result corroborated the EDS (Figure 10).





**FIGURE 7** – Photomicrographs of sample 2250. A) Photomicrograph in natural light showing opaque and chalcedony-filled bioclast domains in brownish opal. B) Detail of the previous photomicrograph showing circular bioclasts filled by chalcedony; C) Detail of photomicrograph showing elongated opaque-marked bioclasts; D) Photomicrograph of lithic artifact with goethite and maghemite-substituted bioclasts taken from Morgenstein (2006).



**FIGURE 8** – Photomicrographs of sample 2574. A) Natural light photomicrograph showing domains of amorphous silica and fibro-radiated chalcedony. B) Photomicrograph identical to the previous one, in polarized light.



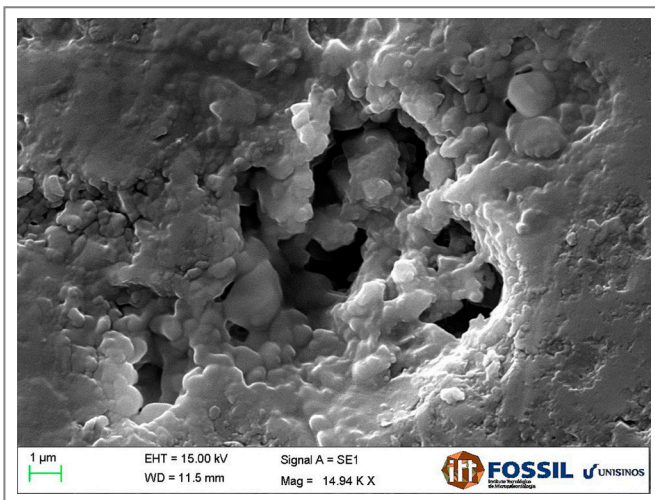


FIGURE 9 – WDS image of sample 2291 highlighting botryoidal structure.

#### 4. Discussion

Two hypotheses were raised regarding the origin and occurrence of silex in archaeological artifacts found at the INDUMA SC-TA-19 site, Taió municipality, as suggested in previous works, which with the results obtained in this research were discarded or improved.

First, we investigated among the various silex origins whether the material of the archaeological artifacts of Taió is the result of hydrothermal activity associated with the Serra Geral magmatism, interacting or not with the underlying sandstones of the Botucatu Formation; or if it is associated with the sedimentation context of the Paraná Basin in the Taió region. The latter proved to be more accurate, since the material analyzed is composed entirely of bioclasts, discarding a hydrothermal origin for the silex.

The origin of the silex is linked to the diagenesis that the Paraná Basin sediments underwent, which is suggested by the presence of bioclasts that are exclusively composed of

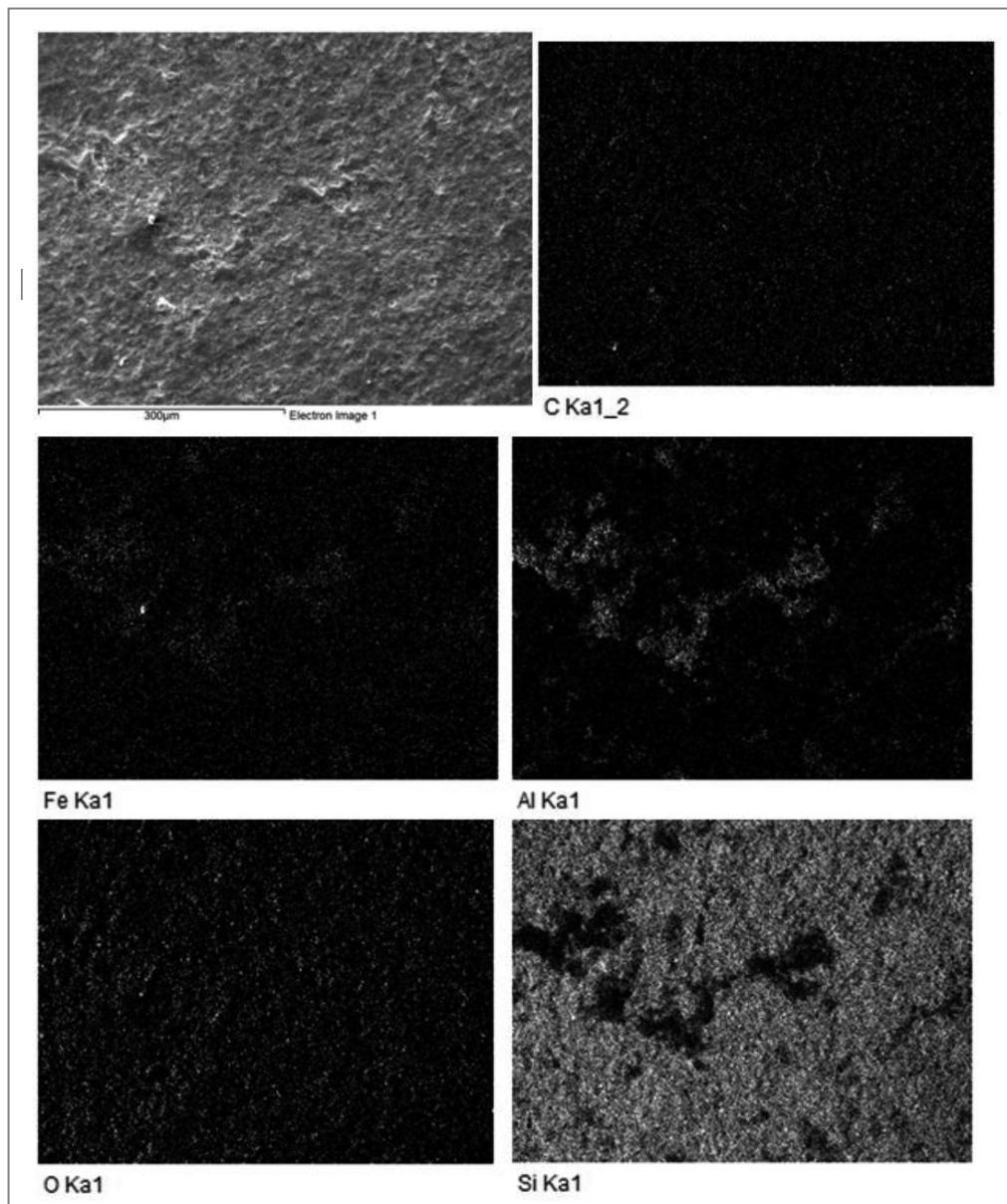


FIGURE 10 – EDS image of sample AM-01 highlighting composition and concentration of Si and O, with subordinate amounts of C, Fe and Al.

carbonates, but in this case are replaced by silica (amorphous and chalcedony) that are surrounded by opaque material, probably organic matter. As observed in the AM-01 outcrop the silex layer studied belongs to the Palermo Formation very close to the contact with the Rio Bonito Formation, however further studies are needed.

Considering the characteristics of the archaeological material, we hypothesized that the archaeological source area of the silex used at the INDUMA SC-TA-19 site could be far away, requiring the inhabitants to travel long distances for collection. However, this possibility was discarded, since fieldwork revealed an outcrop near the site, with material identical to that found in the lithic artifacts. Thus it is suggested that the presence of relatively abundant raw material is one of the reasons why the inhabitants of the INDUMA SC-TA-19 site and the other 25 neighboring sites remained so long in the region as there are archaeological sites dated between 8,000 AP and 4,000 AP years.

## 5. Concluding remarks

This research allowed to study the Taió region in detail, supported by pre-existing studies (Silva and Bortoluzzi 1987; Campos 1964), and adding information on the geology of the region.

In the field, the silex layer was a 35 cm thick, laterally continuous, in a sedimentary context, belonging to the Paraná Basin sedimentary sequence, this being the probable archaeological source area of the silex.

Petrographic analysis revealed that this silex is rich in bioclasts, derived from the concentration of carapaces of possible foraminifera, radiolaria and bivalves, typical of the marine environment.

The chemical composition, obtained by SEM analysis, indicates that the material is composed mainly of Si and O, most of the time, without defined structure, in botryoidal arrangements. However, there are subordinate amounts of C and Ca, corroborating the results obtained in the field, where the silex sample reacted positively to the presence of calcium carbonate. The presence of this mineral is justified because it also composes the shells and carapaces of marine fauna, which may have been almost entirely replaced by silica during the diagenetic process through which the whole sediment package of the Paraná Basin passed. Thus, it is suggested that the origin of the raw material used in the INDUMA SC-TA-19 site artifacts is the silex layer found in the northern region of Taió.

As a contribution to archaeology, we suggested that there are more archaeological sites on the higher ground (Schmitz et al. 2009) linked to the fact that the silex's archaeological source area is also located in higher regions.

### 5.1 Future Works

It is suggested that, in geoarchaeology, the petrographic information obtained in this work can be used to determine

whether people lived in places other than the region of Taió and the presence of this raw material at the regional level. It is also suggested that this tool be tested in other lithic artifacts, expanding the knowledge about the origin of this raw material, since silex is one of the most used materials by the native peoples.

A diachronic study, based on lithic sets of dated occupations, focused on silex use relative to other rocks is also suggested.

In the field of geology, a further study is suggested, seeking to determine which fossils are present, thus allowing to understand the environment present at the time of deposition and subsequent diagenetic processes, as well as a clear definition of the nesting rock of the silex layer thus confirming the lithostratigraphic unit.

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