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GEOCHEMICAL FINGERPRINTING OF NEOLITHIC LITHICS FROM CENTRAL PORTUGAL: INSIGHTS INTO EXCHANGE NETWORKS AT ANTA 1 DE VALE DA LAJE, TOMAR.

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ABSTRACT

Lithics are essential for assessing Neolithic contexts, acting as indicators of cultural, technological, and economic phases. Prehistoric communities sourced local and distant resources through trade or exchange to craft tools for agriculture, pastoralism, or prestige items. Funerary rituals were prevalent in shaping the landscape and occurred at designated sites, either natural or artificial, such as caves or megalithic monuments. In Portugal, the Anta 1 de Vale da Laje (VL1) is the oldest known passage grave in the Alto Ribatejo region. Despite extensive multidisciplinary research including archaeology, architecture, micromorphology, archaeobotany, phosphate analysis, pottery, sedimentology, and dietary studies, the origin of the lithic tools remains uncertain.

This study employs a multi-method approach —typological, geochemical (via pXRF), petrographic, and spatial distribution analyses —to establish possible provenance. It focuses on arrowheads and geometric microliths, comparing them with raw material flint samples from Rio Maior region stored at the Instituto Terra e Memória in Mação (ITM Rio Maior) for geochemical fingerprinting, aiming to identify resource sources and explore symbolic practices. A total of 186 samples were analyzed: 174 lithic artefacts and 12 flint samples from ITM Rio Maior.

The techno-typological results show a predominance of arrowheads with unbroken points, suggesting they were produced as burial offerings, probably sourced from local and distant locations.

Portable XRF detected Platinum Group Elements (PGEs), such as Ru, Rh, and Pd, indicating a local source. Petrographic and scanning electron microscopy (SEM) analyses identified flint and silcrete in the debitage residues, reflecting a procurement strategy based on local resources and/or exchange. Spatial analysis indicates long-term use by communities for over two millennia, with lithic tool distribution across stratigraphic layers suggesting occasional or seasonal occupation by highly mobile groups. Overall, the research deepens understanding of prehistoric behaviour and the evolving role of VL1 during the Neolithic in Central Portugal.

Keywords: Lithics. Neolithic, Geochemical fingerprinting, Anta 1 de Vale da Laje, Exchange networks.

RESUMO

Os líticos são fundamentais para avaliar os contextos arqueológicos atribuídos ao Periodo Neolítico, servindo como indicadores das fases culturais, tecnológicas e económicas. As comunidades pré-históricas tinham acesso a fontes locais e distantes através da troca de matérias-primas para produzir ferramentas para: agricultura, pastoreio ou outros itens de prestígio. Os rituais funerários, por exemplo eram predominantes na paisagem e ocorriam em locais designados, naturais ou antrópicos, como nas grutas ou em monumentos megalíticos.

Em Portugal, a Anta 1 de Vale da Laje (VL1) é a anta mais antiga conhecida na região do Alto Ribatejo. Apesar da extensa investigação multidisciplinar, incluindo arqueologia, arquitetura, micromorfologia, arqueobotânica, análise de fosfatos, estudo das cerâmica, analises de sedimentologia e estudos dietéticos (analises isótopicas), a origem das ferramentas líticas permanece obscura. Este estudo emprega uma abordagem multi-método — tipológica, geoquímica, petrográfica — para determinar a provável e/ou possível proveniência dos materiais identificados.

Este estudo concentrou-se nas pontas de seta e micrólitos geométricos, comparando-os com amostras de matérias -primas (sílex) da região de Rio Maior armazenadas no Instituto Terra e Memória (ITM, Mação); para aferir impressões digitais geoquímicas, com o objetivo de identificar as fontes de recursos, as práticas empregadas e/ou simbólicas. Foram analisadas 186 amostras: 174 artefactos líticos e 12 amostras de sílex matérias-primas de Rio Maior. A análise técnico-tipológica mostra uma predominância de pontas de seta com pontos ininterruptos, sugerindo que foram produzidas como oferendas funerárias, provavelmente de fontes locais e distante.

Da analise realiza com recurso ao XRF portátil detetou *Platinum Group Elements* (PGEs), como Ru, Rh e Pd, indicando uma fonte local. As análises petrográficas e de MEV identificaram sílex e silcretos nos resíduos de *debitagem*, refletindo uma estratégia de aquisição local e/ou trocas regionais. A análise espacial indica uso prolongado por mais de dois milénios, com a distribuição das ferramentas líticas em camadas estratigráficas sugerindo ocupação ocasional ou sazonal por grupos altamente móveis. De modo geral, a investigação aprofunda a compreensão do comportamento pré-histórico e do papel evolutivo da Anta 1 de Vale da Laje no Neolítico em Portugal.

Palavras-chave: Líticos. Neolítico, Impressões digitais geoquímicas, Anta 1 de Vale da Laje, Redes de intercâmbio.

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HREE- High Rare Earth Elements

ICP-AES - Inductively Coupled Plasma Atomic Emission Spectrometry

ITM Rio Maior Flint Samples – Earth and Memory Institute Rio Maior Flint Samples

LA-ICP-MS - Laser Ablation Coupled Plasma Mass Spectrometry

NAA – Neutron Activation Analysis

pXRF - Portable X-Ray Fluorescence

SEM - Scanning Electron Microscope

VL1 - Anta 1 de Vale da Laje

VP-SEM-EDS - Variable Pressure Scanning Electron Microscopy with Energy Dispersive Spectroscopy

vs - versus

GLOSSARY

Chert is composed of microcrystalline quartz (SiO2) that precipitated out of solution given appropriate temperature, pressure, and pH conditions, or in the presence of certain impurities. Clays, iron oxides, carbonates, and organic matter are the primary impurities in cherts, usually present between quartz grains. Only in very unusual circumstances can impurities substitute for silicon in the quartz crystal structure of chert.

Flint refers to all sedimentary rocks composed primarily of microcrystalline quartz, including chert, chalcedony, agate, jasper, hornstone, novaculite, and several varieties of semi-precious gems.

Silcrete refers to a strongly indurated material that resulted from surface or near-surface low-temperature silification of weathered bedrock, regolith and/or unconsolidated sediments. They are very brittle, with a lustrous conchoidal fracture, forming sharp edges suitable for tool use. Its qualities vary widely because of different underlying formation processes. In simple terms, silcrete is defined as a silicified soil.

In this study, the term flint is used to describe chert.

CHAPTER ONE

1.0 INTRODUCTION

1.1 Background to Study

> "*The finds in themselves do not speak to us directly: they are 'voices of silence', as André Malraux once put it, and it is we who must devise a language to allow us to interpret their message*" (L. Binford, apud Daniel and Renfrew, 1988, p. 157).
>
> "*There are two kinds of fossils – physical fossils (remains of early men), and cultural fossils (remains of tools made and fashioned by man)*" (C. Lyell, apud Daniel and Renfrew, 1988, p. 27).

This study contributes to the assessment of the provenance of raw materials used by Neolithic communities in the lower Zêzere valley, part of the wider Tagus basin, the largest basin on the westernmost edge of the Iberian Peninsula.

Provenance has been defined by several other authors (Mellaart, 1963; Jones, 2002; Renfrew and Bahn, 2004; Joyce, 2012; SAA, 2023b; Anderson, 2024). As Richter et al. (2013) explained:

> "The provenance analysis of stone tools plays a crucial role in interdisciplinary archaeological research. It seeks to determine whether Neolithic populations sourced the raw materials for their stone tools from nearby areas or if they were traded or acquired through expeditions from more distant regions. Another possibility is that the Neolithic people used pre-existing, finished stone tools that had been exchanged between different settlement areas" (p.194).

Provenance studies are important for assessing the range of spatial mobility and networking of human communities, which encompasses economic (catchment areas), logistical (mobility strategies and techniques), and cultural (from fashion to beliefs) dimensions. This spatial dimension structures the context of the cultural performance of communities, which encompasses many other variables, including those related to environmental characteristics and resources. However, terms like *provenience* and *context* have been used interchangeably with *provenance*. Provenance originates from the Latin word *provenire*, meaning "*come forth*, *originate*, *appear*, *arise*" (Online Etymology Dictionary, n.d.c). Synonymous with provenience, *provenance* refers to the "*findspot*" of an object (Joyce, 2012). In contrast, *context*, derived from the Latin *contextere*, meaning "*to weave*", the physical components, spatial relationships, matrix, and specific location where an object was discovered are described, as well as its relationships with other objects and features (Renfrew and Bahn, 2004).

According to Millar (2002), "provenance encompasses the full history and journey of an object or record from one place to another, creating a productive space to understand an item and the meanings it has accumulated over time" (p. 8). According to Pereira et al. (2016), "raw material provenance is a powerful tool to infer a wide range of traits related to past human population behaviour such as circulation, territory and economic patterns".

In the study of lithic industries, provenance encompasses factors such as the identification of source rocks, relief and climate in the source area, tectonic setting, transport history, and diagenetic modifications (Schieber, n.d). In archaeology, an assumption is made that some characteristics of the source of raw materials are carried over into the finished object, and that they are sufficiently diagnostic to allow differentiation between geographically distinct sources (Wilson and Pollard, 2001). Chemical fingerprinting refers to these characteristics such as trace element patterns, rare earth profiles, or isotopic ratios (Pollard et al., 2018). These chemical fingerprints of geological raw materials were initiated by (Göbel, 1842; Wocel, 1854; Damour, 1865; Helm, 1886) to determine the specific origin, or as a means of signifying mobility, exchange, or trade of the past peoples. One of the first theoretical considerations concerning the relevance of determining the provenance of archaeological materials by chemical means was made by Damour (1865,313):

> *"When one discovers, in fact, either buried under the ground, either in the caverns, or among the remains of ancient monuments, an object on which the hand of man has marked his work, and whose matter is of distant origin or foreign to the country, it is inferred that there has been transport of the object itself, or at least of the matter of which it is formed. Hence arises inductions on the relations which may have existed between different peoples, on their migrations, their industry, etc."*

This study focuses on the lithic collections of the megalithic monument “Anta 1 de Vale da Laje”, in Central Portugal, a key site to understand the neolithization of the southwest of the Iberian Peninsula (Figure 1).

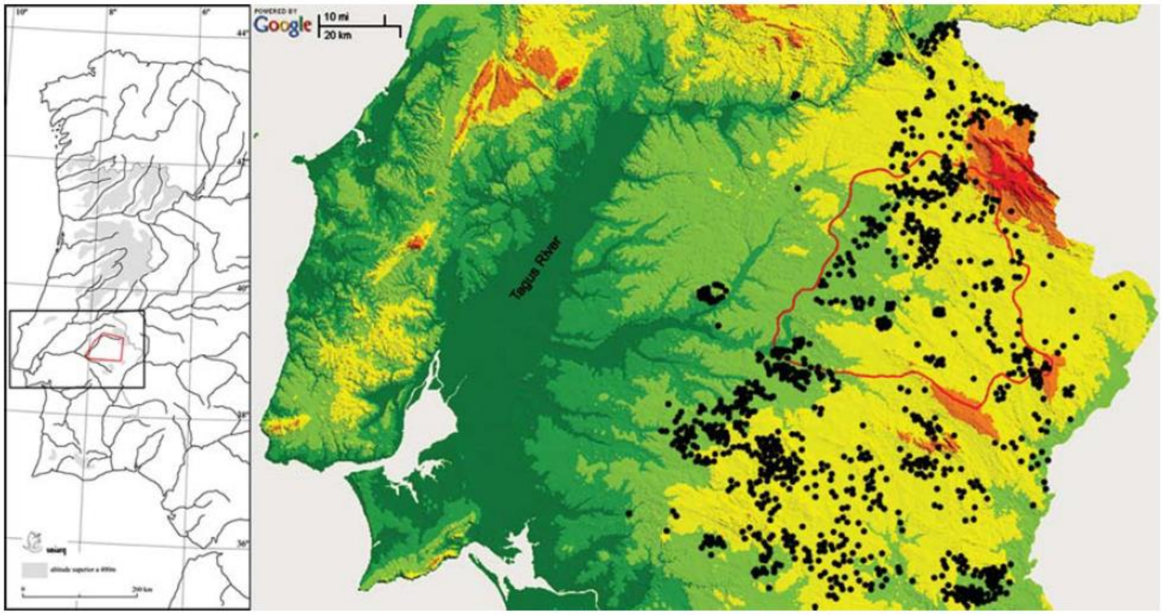


Figure 1: Megalithic Monuments in North Alentejo and Central Alentejo (Andrade, 2015).

Neolithization in Europe is studied using different analytical tools, e.g. radiocarbon dating, isotopic analysis and archaeogenetic analyses, to assess settlement patterns, burials, and material culture across the region. Studies provide insights into the local diet, intensity, complexity, and diversity in domestication, along with climate, subsistence economy, and social networks (Shennan, 2018). The Neolithic Period is often seen as a critical era marking the shift from foraging to farming, as ancient hunter-gatherer groups began cultivating crops (Larsen, 2011). This agricultural spread originated in the Near East and gradually reached Europe (Richerson et al., 2001). Worldwide, Europe's Neolithic transition has been documented (Ammerman and Cavalli-Sforza, 1984; Pinhasi et al., 2005), both globally and locally (Bocquet-Appel et al., 2009, 2012).

Several factors promoted this Neolithic expansion, including influence from waterways (Davison et al., 2006), non-farming environments (Patterson et al., 2010), rapid dissemination around the Mediterranean (Fort et al., 2012), and a slowdown in Northern Europe (Isern and Fort, 2010; Isern et al., 2012). In the Mediterranean, the Iberian Peninsula displays varied landscapes resulting from its connection to Africa and its humid oceanic Mediterranean climate, combined with the Atlantic influence. Notably, it was among the last regions to adopt agriculture (Zapata et al., 2004). In Portugal, the Neolithic economy shifted toward marine resources due to food shortages caused by sea level, climate, and vegetation changes (Lubell et al., 1994). Mesolithic hunter-gatherers exploited rich marine areas such as Muge in the old Tagus estuary, the Sado and Mira valleys, and the Alentejo coast. They practised diverse fishing and shellfish gathering methods, fostering sedentism and high population densities (Anderson et al., 2024). Additionally, Johnson (2014) noted that hunter-gatherer communities have been extensively studied based on subsistence, settlement, technology, and social organization.

During the Neolithic transition, human behaviour and economic structures evolved to better adapt to environmental changes. This adaptation involved utilizing natural resources such as rocks, plants, animals, and water within and beyond their landscapes. Many rocks were crafted into blocks for constructing megalithic monuments, tools, and weapons—serving daily needs, social status, trade, exchange, and mobility. The function and prestige of stone tools depend largely on the availability and characteristics of the raw material sources. Today, research focuses on lithic resource management, as stone tools are among the best-preserved artefacts from Neolithic sites (Brandl and Hauzenberger, 2018), offering vital insights into past human activities and relationships (Andrefsky, 2009). Provenance studies trace raw materials back to their sources, based on characteristic features transmitted into the finished objects. These features include trace element patterns, rare earth elements, or isotopic signatures, enabling understanding of local and regional sources. Provenance study is articulated by the chemical characteristics of certain geological raw materials that serve as a “fingerprint” in finished objects, enabling the determination of their specific origin or as a means of signifying mobility, exchange, or trade (Göbel,1842; Wocel, 1854; Damour, 1865; and Helm, 1886).

This study examines flint arrowheads, flakes, blades, bladelets, geometric microliths, debitage residues, and amphibolite cores recovered during excavations conducted between 1989 and 1993 at the Neolithic funerary site of Anta 1 de Vale da Laje (Tomar, Portugal) (Figure 2).

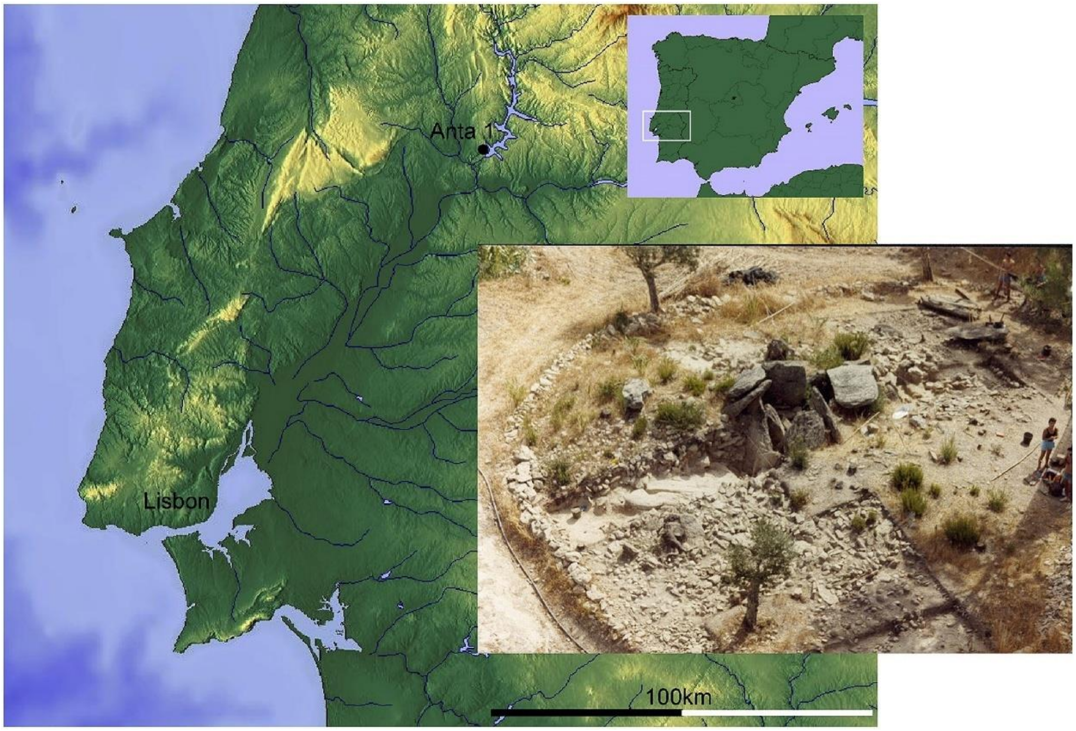


Figure 2: Anta 1 de Vale da Laje in the Iberian Peninsula and an Aerial View of the Excavation in 1991 (Stojavnoski et al., 2020).

It is a small passage grave located among the megalithic necropolises at an elevation of 167 meters in Vale da Laje, Serra’s Parish, Tomar Municipality, in Northern Portugal (Oosterbeek et al., 1992; Drewett et al., 1992). An attempt to identify the local and regional provenance of these flint tools, through geochemical analysis using portable X-Ray fluorescence (pXRF), to evaluate the community's self-sufficiency, the mobility and exchange routes during prehistoric periods. The geochemical data obtained were compared with the Rio Maior raw material site, which is less than 100km from Anta 1 de Vale da Laje.

Portable X-Ray Fluorescence (pXRF) is a non-destructive technique used on artefacts to determine the source of raw materials (Hunt and Speakman, 2015), while scanning electron microscopy analysis (SEM) is a versatile analytical tool employed to reveal the structural and chemical properties of archaeological materials (Ponting, 2004).

Pereira et al. (2016) used pXRF to study the lithic procurement strategies of hunter-gatherers at Pena d'Agua Rockshelter near Lisbon during the Epipaleolithic period. Their findings showed a clear similarity in Al and Si levels between archaeological and geological samples. Some geological samples exhibited enrichment in elements like Ca, Y, and Sr, whereas the archaeological artefacts showed enrichment in terrigenous elements such as K, Ti, Fe, and

Zr. They also noted that elements such as Mn, Mo, and U, detected at the site using pXRF, should be confirmed through other analytical methods.

1.3 Research Problem and Questions

The research question concerns the recognition that megalithic communities prioritised the intensification of resource exploitation, complemented by extensive interaction and broad catchment strategies. These components of human activity significantly contributed to long-distance interactions.

Previous research has demonstrated several indicators of long-distance interactions, ranging from architectural archetypes (Scarre and Oosterbeek, 2021) to dietary evidence (Stojanovski et al., 2020).

Research on geological raw materials demonstrated that local resources were probably utilized; despite this, the source of construction materials for the passage grave was local and, in fact, from within a very short range of less than 300 meters from the monument (Moleiro, 2015). The source of the lithic tools is unknown.

Complementing the above, this study addresses the question of the provenance of raw materials used for producing mobile artefacts, namely tools.

1.4 Aims and Objectives

This study pursued two major aims:

(1) To apply geochemical fingerprinting techniques to the lithic assemblage;
(2) To interpret findings within Neolithic exchange frameworks and their socio-economic contexts.

The objective is to obtain a preliminary assessment of VL1 lithic artefacts and then frame it within the knowledge of the aforementioned frameworks and contexts.

1.5 Scope, Delimitation and Significance of Study

This study focuses on the typological analysis of lithic assemblages and their geochemical compositions. The study provides a characterization of the artefacts, identifying main clusters and, also, a discussion on their eventual relation or not to the Rio Maior main flint sources located in the Estremadura limestone complex, known to be a large source of flint in the

Atlantic coast. This study did not consider the technological analysis of the lithic tools and the context.

This study contributes to the knowledge of the state of the art of VL1 for the interpretation of prehistoric behaviour, thereby providing insights into self-sufficiency, degree of mobility, symbolism, and social networks of flint procurement. The findings may form the basis for future research related to functional analysis of lithic tools, bioarchaeology for provenance mobility, palaeoclimatology, and palaeoecology, among others.

The dissertation is organized through six chapters concerning:

- Introduction (chapter one),
- The characterization of the archaeological and geological context (chapter two),
- The description of the studied materials and methodologies used (chapter three),
- The results obtained (chapter four),
- A discussion on them (chapter five) and,
- A final conclusion (chapter six).

CHAPTER TWO

2.0 ARCHAEOLOGICAL AND GEOLOGICAL CONTEXT

2.1 The Neolithic in Central Portugal: Mobility, Trade, and Monumentality

Several authors have interpreted the neolithization of Western Iberia as the adoption not only of new technologies but of a new ‘Neolithic way of life’ (Guilaine and Veiga Ferreira, 1970; Arnaud, 1982; Zilhão, 2001; Carvalho, 2008). The trigger of the transition might have been concerns about food insecurity (Lubell et al., 1994), specifically following the dry and cold climatic oscillation of 8.2 Ka BP, which forced growing mobility and other novel human strategies, including food production. Several studies have been able to demonstrate the high degree of mobility among Neolithic communities (e.g. Guiry et al. 2016), which would grow and expand through the Middle and Late Neolithic and the Chalcolithic, when it becomes a core component of the economy of marine or aquatic resources. Furthermore, there are many dimensions to the Neolithic, some of which are discussed below:

Renfrew (1969) proposed that areas within 300 km of a raw material source constitute a “supply zone,” as observed in Near Eastern obsidian studies. Renfrew (1977) defined the Law of Monotonic Decrement (LMD): “In circumstances of uniform loss or deposition, and in the absence of highly organized directional (i.e., preferential, non-homogeneous) exchange, the curve of frequency or abundance of occurrence of an exchanged commodity against effective distance from a localized source will be a monotonic decreasing one (p. 72).”

Jones et al. (2003) proposed that local materials located farther from their source tend to be in altered or reworked states because of repeated use of tools. In contrast, non-local materials might indicate opportunistic collection efforts by mobile groups, as suggested by (Binford 1980; Leach, 1988; and Smith, 2010). Understanding these contrasting models is essential for analysing mobility patterns and the exchange of materials.

Long-range exchange and raw materials sourcing were exhibited during the Neolithic period, as highlighted by several authors, some of whom, including Odriozola et al. (2010) opined that the rarity of products is an indication of a smaller number of nearby sources of raw material. They further suggested that the variscite for beads production used in the megalithic funerary contexts across Europe during the Neolithic and Bronze Age were of long-distance contacts and trade routes based on XRF, XRD, and FTIR analyses.

Aubry et al. (2012) showed that Upper Palaeolithic hunter–gatherers in the Côa Valley (Central Iberia) engaged in long-distance raw material exchange to obtain lithics, as revealed by GIS analysis.

Barrientos and Sanjuán (2021) stated that the construction of megalithic monuments revealed the level of social cohesion, mobility to source raw materials within the locality and/or far distant, technological and cognitive power for the choice of materials and their placement to build megaliths during the Neolithic period.

Zilhão et al. (2021) noted that the obsidian products found and widely distributed across Western Europe during the prehistoric period were sourced via long-distance exchange routes and trade from the Tyrrhenian Islands based on the EDXRF analysis of Early Magdalenian finds from La Boja, southeastern Spain.

Costa et al. (2021) found through pXRF, VP-SEM-EDS, and LA-ICP-MS analysis of arrowheads from the Zambujeiro and Mitra 2 dolmens in Évora that the raw materials originated from long-distance trade routes across southern, eastern, and western Southern Iberia during the Neolithic and Chalcolithic periods.

Binford (1980) opined that the "most viable adaptive strategy in uniform environments is a *foraging mode of production* in which consumers maintain a high level of residential mobility, moving to new resource procurement areas as local productivity declines".

Smith (2010) defined the concept of lithic conveyance zones—areas to which Indigenous groups would return regularly to collect stone materials. In European regions, Thorpe and Williams-Thorpe (1991) noted that the sourcing of raw materials for short distances is within a range of 1 to 2 km. Marks et al. (1991) indicated that in Portugal, mobility and the procurement of raw materials took place within the siliciclastic formations of Upper Cenomanian flints in the sedimentary Basin of the River Tagus (TSB), as evidenced by archaeological sites located less than 5 km from these flint sources in the framework of the study of the Upper Paleolithic of the Rio Maior basin.

Jorge (2014) investigated the lithic tools recovered from the settlements and burial places on the Mondego Plateau in central-northern Portugal during the Late Neolithic using a multiple approach based on the material culture approach with a landscape-scale, and comparative artefact studies and discovered that the flints were sourced within the regions of short distance between the range of 1km to 15 km.

According to the osteological data reported by Silva (2002, 2003a), greater mobility was indicated in the individual hip joint and proximal area of the femur studied as opposed to fully sedentary agriculture populations during the Middle-Late Neolithic/Chalcolithic periods due to rearing animals and/or regular visits to hilly regions. This fact is also evidenced in the livestock (sheep and goat) remains and game resources found in the archaeological records in the Estremadura and Algarve regions of Portugal. The Isotopic analysis of the strontium ratio (^{87}Sr/^{86}Sr) in the dental enamel of the population also indicated long-range mobility (Hillier et al., 2008; Hillier et al., 2010; Waterman et al., 2014). Boaventura et al. (2014) reported that greater mobility was observed in the female bones of the agricultural communities of Estremadura and Algarve coastal regions during the Middle Neolithic-Late Neolithic/Chalcolithic Periods in Portugal. Human mobility is also echoed in livestock remains and game resources.

VL1 is a testimony of short-range and seasonal mobility.

Binford (1980) proposed that people typically gathered resources over short distances in an opportunistic way, often during seasonal mobility.

Moleiro (2015) reported that VL1 dolmen construction materials were sourced just 100 m away from the monument, based on an archeopetrographic study. These findings suggest practical and efficient land-use strategies rooted in the region.

Stojanovski et al. (2020) reported seasonal pastoralist mobility to the VL1 monument during the Neolithic based on milk residue from pottery. Oosterbeek (2025) noted that agro-pastoralists during prehistory in Portugal were diverse in cultural materials, morphology, and mobility, exploiting marine and terrestrial resources along the Tagus tributaries and Alentejo plain. These patterns reflect a landscape of intensively used micro-regions connected through seasonal and subsistence-based mobility.

In summary, mobility is a demonstration that movement is an essential part of our lived experience, whether it is in terms of the regular journeys, resources and raw materials are procured, seasonal gathering trips, or occasional forays to more distant lands. The Neolithic period is characterized by the movement of human and animal remains, monuments, landscapes, seascapes, and lithic and other artefact assemblages for the recognition of social associations, community relations, spiritual and religious practices, and economic activities (Leary and Kador, 2016).

Megalithic monumentality is the outcome of the relationship between different regions, in which concepts and resources are circulated in multiple directions. In Portugal, the megalithic funerary practices initially started as cave burials, as observed in the limestone region of Portuguese Estremadura, such as Gruta do Caldeirão and Nossa Senhora das Lapas, during the Early Neolithic and beyond with individual burials to collective burials and later evolved into megalithic tombs during the 19th century. One common funerary practice from the Early Neolithic to the 19th century is the placement of the dead on the burial space floor rather than pits or graves. In Central and Southern Portugal, the erection of individual and stone settings of standing stones was practiced in the Algarve and Alentejo regions during the Early Neolithic period to the 6th millennium BC, as development of megalithic monumentality (Scarre, 2020). The Beira and Alentejo regions were “classic” areas for the study of megalithic monuments in Portugal from the middle 19th century (Carvalho, 2024). Notably, Alentejo region of Portugal is famous for its high number of Neolithic-age megalithic monuments, such as tombs (dolmens or antas) and ceremonial features like standing stones (menhirs) and stone circles (cromleques) (Pope and Miranda, 1999).

Megalithic societies are characterized by social and economic disparities between neighbouring human groups regarding access to raw materials and other resources, coupled with the sharing of cultural materials such as flint blades, geometrics, polished stone tools, pottery, and personal ornaments (Carvalho, 2024). Throughout many culturally defined periods, funerary landscapes had both a social and symbolic impact (Boas, 2024).

Jones et al. (2003) proposed that local materials are more likely to be found farther from their source in reworked or broken forms, due to increased 'tool-using events'. In contrast, other proponents argue that non-local materials appear at sites because of opportunistic gathering by highly mobile groups (Binford 1980; Leach 1988).

2.2 Overview of Flint Usage in the Portuguese Neolithic

In Portugal, the western façade is the richest and most important flint supply zone. Primary and/or secondary flint sources were identified in the Southern façade (the Algarve coast). The quantity, accessibility, variety, quality and size of the western façade flints are exceptional than other parts of Portugal. These flint sources served as raw material to produce bifacial artefacts during the Neolithic/Chalcolithic (Forenbaher, 1999). Lithic production in Portugal from the Late Neolithic to Chalcolithic was performed by craft specialists and on an individual household. These flints were fashioned into tools and were used in their settlements and burials variously for utilitarian purposes, hunting, warfare, and for prestige (Forenbaher, 1999). Massive flint exploitation, extraction, production sites and large deposits of flint artefacts occurred in west Central Portugal but were absent in the South (Silva, 1993b). Flint is valued due to its characteristic features that include good quality, fine-grained, and rare internal flaws. It has a wide range of shades from opaque to semi-translucent. These characteristics are typical of the Cenomanian silicifications (Cretaceous) of the Portuguese Estremadura, where it occurred a secondary source on Miocene deposits (Aubry et al., 2014; Matias, 2016).

Chronologically, flint procurement strategies and lithic artefact production in the Iberian Peninsula are summed up into two according to Andrade and Matias (2011):

> (1) "occasional exploitation sites, used in the framework of seasonal group movements to satisfy immediate needs, and related to the advent and affirmation of the first farming communities (Neolithic)".
>
> (2) "permanent exploitation sites specialized in artefact production, determined by permanent raw-material procurement needs, and related to the consolidation of stable farming communities (Chalcolithic)."

Therefore, Jimenez Lorente et al. (1999) analysed and discoursed that the archaeological sites imply that:

> (1) "that they are exploitation sites located in the resource procurement area of one or more settlements and where the shaping of flint blocks and occasional manufacture of blanks and tools was undertaken".
>
> (2) "that they are exploitation sites where flint blocks were shaped into core pre-forms, with subsequent reduction taking place at settlements located nearby"

2.3 Previous Research at Anta 1 de Vale da Laje

The set of native archaeologists from the Centre for the Study and Protection of Tomar's Cultural Heritage (C.E.P.R.T.) first discovered Anta 1de Vale da Laje (Drewett et al., 1992). A reconnaissance survey of the site was carried out by Luiz Oosterbeek and Ana Rosa Cruz in the late 1980s. The earlier excavations were carried out in 1989,1990, and 1991 respectively on focused areas within three months undertaken by various groups of people that included institutions such as Granada and London Universities, E.S.T.T. students, sponsors such as Youth Institute, European Communities' Bureau Erasmus, Tomar City Council, Tomar Infantry Regiment coupled with the Nun Alvares Pereira Secondary School and the cooperation received from the land owner – Mr José Caetano with his wife.

The mound was first excavated in 1989, purposely for its stratigraphical classification. In 1990, the stages of construction and modifications of the monument were further discovered during the excavation of the mound under the co-supervision of Dr Peter Drewett. The structural features such as the onset of the lithics, paleosol, and buttresses, among other features were specifically examined during the 1991 excavation, co-supervised with Dr João Coroado for geoarchaeology (Oosterbeek et al., 1992). Dr Paulo Felix conducted the survey and drawings of the site. The archaeological finds, plans, drawings, photographs and other records are in custody of the prehistory sector of the Escola Superior de Tecnologia de Tomar of the Polytechnic Institute of Tomar (Drewett et al., 1992).

Oosterbeek's (1994) quest to define the start of the Neolithic period in Iberia led to the exposure of the rich finds in North Ribatejo, Portugal. North Ribatejo was considered based on the absence of plains, and the presence of flood areas for arable farming, forest, and good alluvial soils. It is composed of Precambrian and Paleozoic rocks in the northeast, a Mesozoic complex in the northwest consisting of limestones in the northwest and several inverse faults bearing north-south, and Cenozoic deposits dated Miocene-Holocene in the south. The lithologic units of North Ribatejo contain limestones (with flint) in the northwest, schists and granites in the northeast, the south is dominated by deep alluvium deposits coupled with sandstones, while quartz and quartzite pebbles dated Plio-Pleistocene are on the uppermost part of these rock units. The hydrographic system was mainly controlled by the Tagus and Zêzere Rivers, while the karstic relief was formed due to the Nabão River flow system. Archaeological sites are not too far from these sources of water. The agricultural potential of northern Portugal is very good and highly diversified. All these natural resources have probably attracted prehistoric people to the area.

Oosterbeek (1994) recovered finds that include lithics, pottery, bone, ornaments, and metal, with lithics dominating the artefacts. Studying eight sites from North Ribatejo (Gruta de Nossa Senhora das Lapas, Amoreira, Juncais de Baixo, Anta 1 de Vale da Laje, Gruta do Cadaval, Gruta dos Ossos, Gruta do Morgado and Povoado da Fonte Quente), he established the context of transition and change that led to megalithism. The excavations at Anta 1 de Vale da Laje revealed stratigraphy from the Neolithic to the Early Bronze age, with at least their main occupation phases, illustrated by architecture (Neolithic construction, Chalcolithic reconstruction and monumentalization, and early Bronze age reuse) and by changes in artefact assemblages (lithics and ceramics).

Layer C is the monument construction level; the entrance and exit were made of gneiss materials with intercalations of yellowish silt-clay and buttresses were also emplaced. It is dated Neolithic Age.

Layer B: structural modifications in this layer include the addition of a mound made with a stone and a lithic belt composed of quartz and quartzite pebbles. This is the burial layer as evidenced by human bones such as adult fragmented skull caps and a minor of unknown sex. It is dated Chalcolithic Period. In the main occupation layer (B), the monument was reorganized, the mound was covered with stones to look like a cairn, adding a kerb around it and some other complex structures by the entrance, including a paved *atrium* and a stone removable “door”. Both the lithic and the ceramic assemblages were abundant.

Oosterbeek compared and established five-stage models to describe the Neolithic process using the context of cultural-chrono-stratigraphical to illustrate his theoretical views, including evolutionism, diffusionism, and his own dialectic approach. Oosterbeek observed that the linear evolutionist model described Anta 1 de Vale da Laje as a land-marker of megaliths based on the availability and accessibility to the natural geological resources of amphibolite within the Tagus Valley coastal area. The fifth model stressed the unequal and combined development of the Neolithic in the Iberian Peninsula, Anta 1 de Vale da Laje standing for one of the cultural traditions of the epoch, primarily related to the inland Alentejo plain and the Tagus River basin, rather than to the coastal areas (Oosterbeek, 1994).

Several disciplinary studies were undertaken. For instance, sedimentological assessment by Garcia's (2000) granulometric and statistical analyses defined ten stratigraphic units for VL1 from the posterior analysis of the deposit obtained from the northern part of the site with visible and clear structural features. The author concluded that the sediments comprised micaceous sand-silty to blocks across the different layers, very dark grayish brown to olive brown, bioturbated, possibly originating from the weathering of the gneissic rocks, coupled with rich carbon remains.

Based on micromorphological assessment of the stratigraphic layers, Adewumi (2019) reported three (3) evolutionary phases of site construction and six (6) periods of activities related to human and natural modifications before, during, and after the construction of megalithic monuments. Bioturbations of excremental origin, possibly due to slaking or mass movement were observed at this site. Adewumi (2025) micromorphological data revealed that there were in situ changes in the VL1 sediments due to the predominance of weathering and alterations, as observed in the recurring sheetwash deposits with varying laminations, indicating different rainfall intensities and sediment loads.

Migliavacca (2000) reported the presence of gneiss pebbles, quartzite clasts, unconsolidated silt sands and deeply bioturbated sediments related to the samples analyzed, stressing the increase of phosphates in the Chalcolithic, probably related to animal herding. The phosphate analysis indicated a high phosphorus content of 45.8% in the samples collected within the monument, below the monument slabs, and beneath the slabs attributed to the Neolithic, thus reflecting the funerary practices during the site's earliest phase. Depleted phosphorus values were observed during the later construction phase of the monument, indicating human activities (Migliavacca, 2000).

In the framework of artefacts analysis, Coroado and Cabral (1995), multivariate analysis of 58 clay samples from Maxial, Vale da Bairrada and VL1 using the ICP-AES confirmed that the clays and ceramics of the VL1 (from layer B) were dominated by amphiboles and plagioclase like the lithologic composition on the right bank of the Zêzere River. He confirmed that these vessels served different purposes in the prehistoric period. He further suggested the correlative geochemical study of the clays from these areas - Tomar, the western part of Maxial settlement, areas with the largest concentrations of amphibolite, amphibolite outcrops, north-west pegmatite area dominated by potassium feldspar to determine the qualitative and quantitative aspects of the deposit's mineralogy. Coroado and Cabral (1995) opined that the clays for ceramic production in Anta 1 de Vale da Laje and Povoado do Maxial were sourced within the locality - Tomar clays using the ICP-AES atomic emission spectrometry method of analysis. Fuying (2008) established that VL1 and Gruta do Cadaval contained similar mineralogical compositions of quartz, mica, and feldspars based on XRD and NAA pottery analysis. However, VL1 cluster analysis contained more clusters and resemblance; the Fe contents of the ceramics were indicative of probably being formed *in situ*, and abundant plagioclase was also present. Moreover, he indicated that the acidic nature of VL1 soil could be traced to the presence of HREE, Ga, Rb, Cs, K_2O, Ta and U.

Stojanovski (2017) confirmed the good preservation of lipid molecules in the pottery matrix of VL1 despite its soil acidity and noted that there was a shift in the economy to intense agro-pastoralism due to the wide consumption of dairy products during the Neo/Chalcolithic period. Stojanovski (2020) concluded that this site recorded the highest recovery rate for lipid residue from archaeological pottery in Europe. Layer B contained diverse pottery shapes while layer C contained only bowls with carinated types common to both layers. VL1 site pottery assemblage showed diversity in size and shape, with bowls having 75% of the entirety of the shapes analyzed which reflected that the cultural activity mostly used bowls either for communal food or drinking for a large populace during festivity, burial ceremony, and annual event. In terms of decoration, most pottery of VL1 is not decorated except for "almagra" (red painted vessels), notably common in the central and southern areas of the Peninsula.

Stojanovski et al. (2020) recognized four categories of pottery containers (dishes, bowls, lamps, and rims) for various purposes. They used compound-specific radiocarbon to establish the consumption of meat and dairy products by these early people during the Neolithic and that the animals' diets reflected stable carbon isotope values of fatty acids probably due to one or all these factors - the salinity of the Tagus estuary, acute aridity, and significant input of Carbon 4 plants in the animal diet. Also, they established that the site was used for social gatherings due to pottery litter in the place.

In the context of paleoenvironmental reconstruction, the anthracological study of over 600 fragments of charcoal attributed to 18 different plant taxa were recovered from Anta 1 de Vale da Laje such as *Ericaceae* (*Erica* type *arborea* (white heather); *Arbutus unedo* (arbutus); *Calluna* sp. (heather-vulgar); *Erica* sp. (heather) and *Cistaceae* (steva); *Rhamnus / Phillyrea* (aderno); *Leguminosae* sp. (vegetables); *Ligustrum* sp. (common alfena); *Olea* sp. (Oliveira); *Thymelaeaceae* sp. (trovisco); *Cornus* sp. (sanguinho-legítimo) and *Pinus* sp. (Pine) as open vegetation and belonging to Neolithic, Chalcolithic, and Bronze Ages. The sediment from the Dolmen interior yielded the importance of shrub plants, and thickets have been a marker species throughout the different phases of occupation at the site. Thermophile vegetation is common due to the Mediterranean climate. Oak forests have been lost due to natural fires and agropastoral activities, supporting the growth of temperature-loving plants on the site (Allué, 2000). Moleiro (2015) dated the VL1 site 6th and 3rd millennium BC based on archaeobotanical remains.

Ferreira (2020) analysed two stratigraphical layers - F27 East and H26 West profiles. From the uppermost layer of the F27 East profile at a depth of 10cm, 13 pollen taxa were recovered including the arboreal taxon *Quercus* sp. (perennial type, with increase); Shrub – *Ericaceae* (increases) and others such as *Cerealia, Chenopodiaceae*, *Apiaceae*, and *Ranunculaceae*. Likewise, at a depth of 15cm, 13 pollen taxa occurred, including the reoccurrence of arboreal taxa- *Pinus* sp and evergreen *Quercus,* coupled with shrubs and herbaceous species such as *Chenopodiaceae*, *Thymelaeaceae, Brassicaceae, Ranunculaceae, Ramnaceae*, and *Cerealia* (novel taxon discovered). At a depth of 25cm, between 4 to 8 taxa were recovered consisting of arboreal type - *Pinus* sp.; shrubs - *Ericaceae;* herbaceous - *Poaceae*, *Asteraceae*. At a depth of 30cm (the deepest and oldest stratigraphical layer with the highest pollen diversity and concentration) fourteen (14) pollen taxa comprising of *Pinus* sp. (most abundant), deciduous - *Quercus* sp. (rare), evergreen *Quercus, Oleaceae*, *Fraxinus;* shrubs – *Ericaceae;* herbaceous - *Poaceae*, *Asteraceae*, *Brassicaceae,* and *Thymelaeaceae* (all mostly abundant). The uppermost layer of the stratigraphical H26 West profile at a depth of 15cm contained an outstanding pollen concentration and taxa of 1639 grain/gr. sediment and 8, respectively. The herbaceous taxon *Thymelaceae*, *Brassicaceae*, and *Asteraceae* (had highest concentration of 1116 grain/gr. sediment); the arboreal taxon *Pinus* sp.; and the shrub *Ericaceae*. The deepest and oldest layers were impoverished and rare in pollen taxa and concentration.

The uppermost layer (at depth 10cm) of the stratigraphical layer - F27 East profile contained 754 NPP/gr.sediment with the increase of fungal taxa such as *Glomus*, *Tilletia*, *Polyadosporites*, and *Pluricellaesporites*; zoo remains, and undefined taxon– *Pseudoschizaea*. The intermediate layer from depths 15-25cm contained a marked presence of fungal taxa such as *Hipha, Polyadosporites,* and *Pluricellaesporites* coupled with the decrease in zoo remains. The deepest and oldest depth at 30cm contained the richest non-pollen concentration of 600 NPP/g.sediment influenced by the presence of zoo remains and highly diversified pollen taxa with a marked abundance of fungal taxon species such as *Polyadosporites* and *Exesisporites*. The uppermost layer of the H26 West profile at a depth of 10cm contained the highest pollen concentration of 639 NPP/gr.sediment with 9 pollen taxa represented by fungal taxon species such as *Polyadosporites, Pluricellaesporites* and *Tilletia;* zoo remains, and *Pseudoschizaea* from the undefined group. At the intermediate depth of 25cm, there is a great decrease in the values of the pollen concentration and taxa with the fungal taxon *Exesisporites* as the dominant species. While the two oldest and deepest depths – 35 and 45cm contained very poor pollen concentration and taxa. However, it can be concluded that during the Quaternary period, there were nomads with herbivorous animal presence and pastoralism as a subsistence occupation due to the abundance of fungal taxon species recorded at VLI (Kuoppamaa, 2025).

2.4 Anta 1 de Vale da Laje: Site Description and Lithic Assemblage

Architecturally, Anta 1 de Vale da Laje is a pentagonal chamber monument with a small passage. The memorial measures 5.8m externally and internally 4.8m with a height of 1.75m (Figure 3). The chamber is 2.2m wide. The corridor covers 56% of the area and is about 0.8m high, narrowing down towards the entrance, covered with a capstone 0.6m lower than the chamber. The oval-shaped tumulus (which looks like a cairn) was built on an earth composed of a thick stone shell measuring about 9 to 10m east-west and north-south, respectively (Oosterbeek et al., 1992). The architectural design of *Poço da Gateira* and *Gorginos* 2 is similar to VL1, both passage graves dated ca.5th millennium BC through thermoluminescence, probably indicating the earliest stages of megalithic monuments construction in Portugal (Leisner and Leisner, 1951; Whittle and Arnaud, 1975). Based on its architecture and archaeological finds, the site showed different periods of monument construction, usage and modifications (Almeida and Oosterbeek, 2019).

Figure 3: Anta 1 de Vale da Laje (VL1) Megalithic Tomb (Photo: Nelson J. Almeida, IPT).

monuments in the Vale da Laje small valley on the right coast of the Zêzere River. It was built on the highest topography of 167m, thus enhancing its visibility above other features within the vicinity. The Dolmen was built directly on a gneiss outcrop, while the mound was constructed with quartz and quartzite materials, which had several stages of construction and modifications. These geological materials were procured within the neighbourhoods at less than 100m. The soil is very acidic (Oosterbeek et al., 1992). Anta 1 de Vale da Laje monument is well preserved and of great importance in the archaeological context of the central part of North Ribatejo, probably the oldest (Drewett et al., 1992). In Europe, raw materials sourcing was within a 1 to 2km range, as evidenced in Anta 1 de Vale da Laje, whose raw materials were procured within a distance of 100m from the monument (Thorpe and Williams-Thorpe, 1991).

The local community people of the vicinity of Anta 1 de Vale da Laje (Tomar), Anta da Lajinha (Mação), Anta da Foz do Rio Frio (Mação), and Anta do Penedo Gordo (Gavião) harnessed the availability of geological resources such as lithic materials within their reach for construction activities. This intentionality showcased their technological knowledge, social cohesion and relationship within the landscape (Oosterbeek et al., 1992; Moleiro, 2015). Adewumi (2020) confirmed different phases of human occupations, natural and human-induced modifications at the site through soil micromorphology. Stojanovski (2020) noted that the VL1 site was probably often visited by pastoralists, mobile foragers, and the entire community during seasonal changes and other social activities because settlements were scarce in the Portuguese landscape during the prehistoric period. This opinion was further supported by (Oosterbeek, 2001; Scarre et al, 2011; Cerrillo-Cuenca and González Cordero, 2014).

Stratigraphically, according to Oosterbeek et al. (1992), twenty-nine units were identified but were composited based on the matrix into four layers. The layers include:

Layer D: - the foundational level for the Dolmen, made of paleosol. It contains silty-clayey materials with rich coals and is relatively undisturbed.

Layer C: - the monument construction level. The passageway flooring is made of gneiss pebbles, buttresses were placed, and the outer edge is constructed with blocks of gneiss. It contains yellowish silt-clay intercalated with a few pebbles. It dates to the 4th Millennium, with archaeological finds characterized by the Neolithic Age. Perishable materials used in later construction phases on this layer have been destroyed. Layer C surface ceramics are rough and friable. It contains strange fragments and oxidized fire.

Layer B: - Minor modifications were made at this level to some of the structures, such as the mound made with a stone (shaped like a cairn), a lithic belt built at the outer edge of the mound built with quartz and quartzite pebbles. This layer contains a brownish silt-sand deposit with abundant gneiss pebbles. It dates to the 3rd Millennium and is referred to as the Chalcolithic Period. Burials were made on this layer, at a height of 5cm, attached to the mound, different parts of human bones were recognized with fragmented skull caps embodying an adult and a minor of unknown sex. Other finds include schist plates decorated with wolf tooth of proximal peroration (ocular), green pendants with conical perforation, green discoid and tubular beads. Layer C ceramics include dozens of vessels of six main types.

Layer A: - It is a very disturbed layer made of loose deposits. It contains abundant plant roots and ceramics. It is referred to as the Bronze Age based on finds of medium-low carina shiny-spatulated surface vessels.

Other previous studies on stratigraphy include: Cruz (1997) identified four lithostratigraphic layers named A-D with different sediment colour variations, composition and textures, and archaeological finds probably dated Early Bronze Age to Languedocian? Stojanovski et al. (2020) labelled VL1 stratigraphy with layers A-C above containing archaeological finds and the layer below with no archaeological finds as D. The monument was constructed on layer C, intensified usage on layer B, while the disturbed layer is A dated Copper Age - Recent based on its remains.

According to Oosterbeek et al. (1992), layer C contains tools made of lithics such as flint microliths (trapezoids) and very fine polished amphibolite axes. Layer B contains abundant remains of various types of retouched and unretouched blades, notches, scrapers, dozens of different types of arrowheads made of flint, the chisels, adzes and axes were made of amphibolite and greywacke, cores of lamellae made of translucent quartz, and flakes made of flint, translucent quartz, and quartzite. The uni/bifacial macro tools (choppers and chopping tools) discovered in this monument are also found in *Maxial* village and *Beiras* megalithic monument (Oosterbeek et al., 1992). VL1 is a mosaic of the three rock types: schists, amphiboles, mica schists, greywackes, quartzites, carbonate rocks, and gneisses. It has low permeability due to porous formations of schists, greywackes, mica schists, and gneisses, among others and dated Precambrian (Cruz, 1997). Moleiro (2015) recorded that the gneissic slabs used for the monument construction were acquired from nearby gneiss outcrops. Oosterbeek et al. (2017) reported archaeological finds made of flints (arrowhead, blade, fragment, shards, scrapper, chip), quartz (flake, fragment, chip), quartzite (flake, fragment, chip), and shale (fragment) that supported previous dating of VL1 as Neolithic/Chalcolithic.

Almeida and Oosterbeek (2020) carried out an extensive excavation around the tumulus on a grid of 2m by 2m for the comprehensive analysis based on sedimentology and micromorphology, among others, to elucidate stratigraphy and chronology. Based on the archaeological finds, the Neolithic/Chalcolithic age was confirmed. Stratigraphically, ten layers of different colours and compositions were defined. The archaeological finds were made up of flint, amphibolite, quartz, and quartzite.

2.5 Geology of the Study Region

The Iberian Peninsula represents a subcontinent characterised by a diverse array of geographic, landscape, geologic, and climatic features, encompassing an estimated area of approximately 582,860 km². The peninsula's predominant feature is an extensive central plateau, covering roughly 211,211,000 km² and standing at an elevation of about 600 meters. Within the Iberian Peninsula, a variety of landscapes can be observed, including mountains, which are more prevalent in the northern regions compared to those in the southern regions. Significant plains are primarily located in the Tejo region in central-southern Portugal and the Quadalquivir Valley in southwestern Spain (Ribeiro et al., 1987; Daveau, 1995). The three principal geomorphological features of Iberia consist of Massif Precambrian and Cambrian rocks, including schist, granite, and quartzite; secondary and early Tertiary limestone deposits, which define the Mediterranean landscape features; and the Tagus Basin, filled with sedimentary deposits dating to the Ceno- Anthropozoic era (Oosterbeek, 1994). The climatic variability of the peninsula is attributed to the influences of Atlantic winds and currents, along with the continental climate of the Meseta, which is typified by dryness and elevated temperatures during the summer months (Oosterbeek, 1994).

Geologically, the Portuguese territory belongs to the Hercynian terranes of the Iberian Peninsula, and these terranes majorly contain formations of igneous and metamorphic rocks that lack flints and are dated Precambrian and Paleozoic age (Alvarado, 1980; Ribeiro, 1980). The development of the western Iberian margin is associated with the initial opening phases of the North Atlantic from the Late Triassic to the Early Cretaceous, which was influenced by inherited structures from the Variscan basement (Kullberg et al., 2006). Along the Atlantic façade, there exist abundant sedimentary rocks (limestones, sandstones, and shales dated Jurassic and Cretaceous) in the "Western Façade" that extends along the Atlantic seaboard between Lisbon and Coimbra of 40 km wide and 200 km long. The smaller terrane "Southern Facade" extends along the southern coast of the Algarve. In between the western and southern facades, there is the occurrence of smaller terranes of Serra de Arrábida (west of Santarém) and parts of Grânola (Ribeiro, 1980; Serviços Geológicos de Portugal, 1968). Central and Western Portugal are covered by Neogene alluvial deposits, probably of Miocene and Pliocene age. In Portugal, the primary agent of transportation and redeposition is by river action and primary flint sources are deposited near the Atlantic coast. From the primary sources, flint was eroded and transported towards the western part of the ocean and was deposited. This can be observed in the western part of the alluvial basin of the Tejo as secondary flint sources and at the easternmost Jurassic sedimentary terrane of the Tejo near Santarém (Forenbaher, 1999). Portuguese flint sources are found in the Western Façade and other alluvial areas (Shokler, n.d.) Figure 4.

The Central-Iberian Zone of the Iberian Massif mainly comprises coarse-grained Armorican metaquartzites from the Lower Ordovician, which form narrow ridges and are resistant to erosion (McDougall et al., 1987). These rocks are also part of the composition of the Douro and Tagus River deposits. Rarely, conglomerates, sandstones, and sandy mudstones are present; these are consolidated fluvial deposits from the Late Cretaceous to the Palaeocene in the Douro basin (Mediavilla and Dabrio, 1986; Blanco et al., 2008). The development of the western Iberian margin, associated with the initial opening phases of the North Atlantic from the Late Triassic to the Early Cretaceous, was influenced by inherited structures from the Variscan basement (Kullberg et al., 2006). Quartz appears in different mineral forms, such as hydrothermal vein fillings in joints and fractures caused by Variscan tectonics, and as a secondary mineral in detrital deposits. Flint occurs in three primary depositional settings: firstly, as syn-diagenetic siliceous precipitates formed in open or barrier reefs on the continental shelf within the Mesozoic stratigraphic sequence of the Lusitanian basin; secondly, within Miocene lacustrine environments on the northern and southern edges of the Central Mountain System (Bustillo, 1976; Bustillo and Pérez-Jiménez, 2005; Armenteros, 2000); and finally, as silcrete, an amorphous silica that transforms into opal-CT, found in scattered outcrops across the northern and southern Meseta (Molina Ballesteros et al., 1997; Blanco et al., 2008) and in the Lusitanian basin (Proença Cunha, 2000).

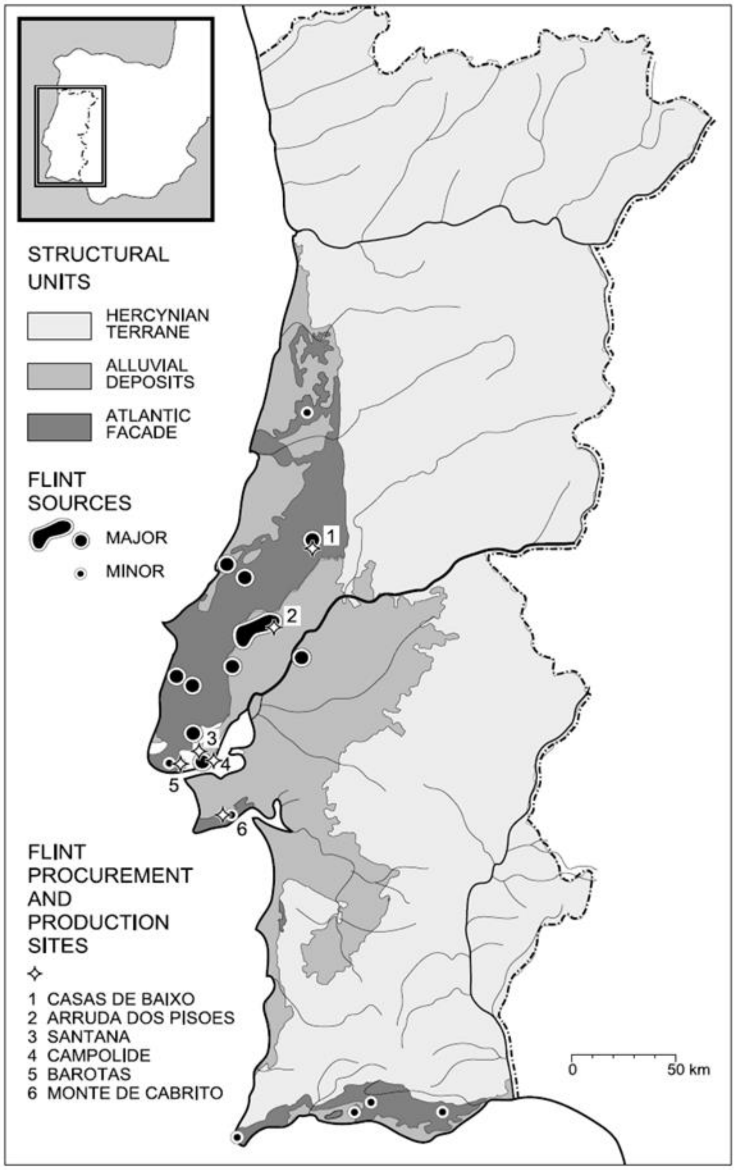
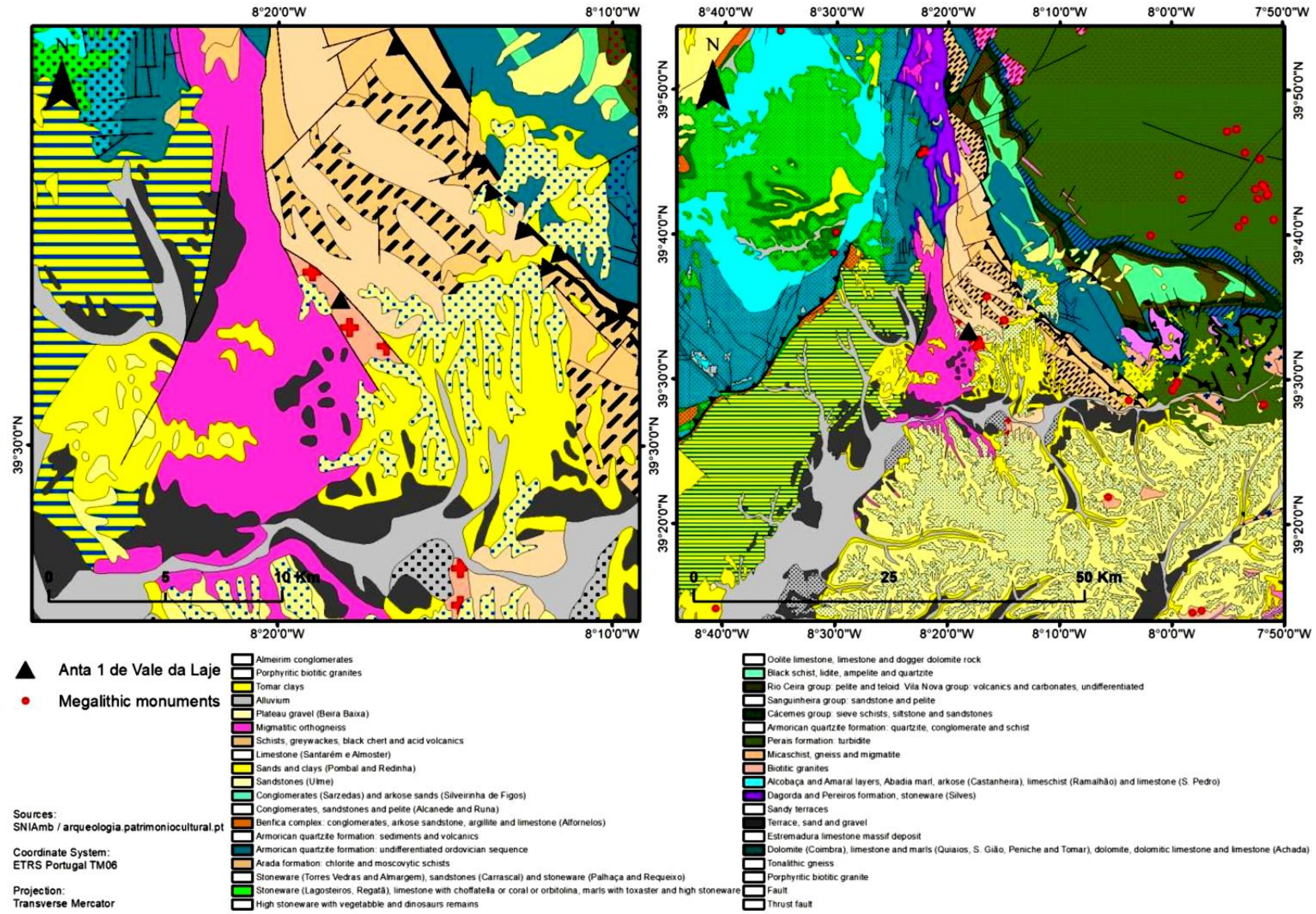


Figure 4: Main Geological Structural Units, Flint Sources and Flint Procurement and Production Sites in Portugal (Forenbaher,1999).

In Western Iberia, geological factors restrict flint sources to specific areas, most notably the western Atlantic façade of Central Portugal – the modern province of Estremadura (Forenbaher, 1999). In Iberian Massif sites, fine-grained siliceous rocks are present, but flint and silcrete are not naturally available. It is noted that flint and silcrete originated from the Lusitanian, Tagus and Douro Basins (Aubry et al., 2016). In this context, the local geology of these basins likely contributed to the formation of silcrete (Figure 5). Flint and silcrete come from the Lusitanian, Tagus, and Douro Basins (Aubry et al., 2016). Together, the Douro and Tagus River Basins cover about one-third of the Iberian Peninsula and feature a Mesozoic marine basin (Lusitanian), a Cenozoic basin in the continental crust (including Duero,



Sources:
SNIAmb / arqueologia.patrimoniocultural.pt

Coordinate System:
ETRS Portugal TM06

Projection:
Transverse Mercator

Figure 5: Map Showing the Regional Geology with the Location of Anta 1 de Vale da Laje Site (Map: Luis Costa).

Western Mountains (Ribeiro et al., 1979; Kullberg et al., 2006; de Vicente et al., 2011). Prior research has identified abundant, large, and uniformly distributed siliceous nodules across these basins. Aubry et al. (2012) reported that lithic raw material sources are accessible in both primary and secondary deposits within the Douro and Tagus basins.

The VL1 lithology corresponds to the Alto Ribatejo region, which comprises limestones and marls (with a weak presence of flint), along with dendritic deposits filling riverbeds, as well as schist, gneiss, and granites to the east of the monument. The VL1 environment includes more schist, greywacke, quartzite, and granitic rocks, and the dendritic drainage basin consists of clay, silts, sands, and pebbles. The region's Quaternary deposits encompass Holocene alluvial sediments, Pleistocene wide fluvial terraces, karstic cave fillings from the Limestone Massif, and dendritic coverings (Adewumi, 2019) (Figure 6).

Silcrete is broadly characterized as a sedimentary rock in which substantial amounts of authigenic silica either accumulated in or replaced surface and near-surface deposits, forming an indurated mass (Summerfield, 1982; Milnes and Thiry, 1992; Nash and Ullyott, 2007). Summerfield (1983) simply defined silcrete as a silicified soil. It is a fine-grained siliceous rock recognized by conchoidal fracture, producing sharp edges suitable for tools. Its properties vary considerably due to different formation processes (Will, 2021). During the Pleistocene and Holocene, silcrete was widely utilized globally for toolmaking (Wragg-Sykes and Will, 2017). The collection and utilization of silcrete are linked to specific techno-complexes, heightened “mobility, technological innovations such as heat treatment, standardized techniques like backing, bifacial, and pressure flaking, and even symbolic behaviours (Singer and Wymer, 1982; Ambrose and Lorenz, 1990; Wurz, 1999; Henshilwood et al., 2001; McCall, 2007; Mourre et al., 2010; Lombard et al., 2012; Schmidt et al., 2013; Mackay et al., 2014; Will and Mackay, 2017).

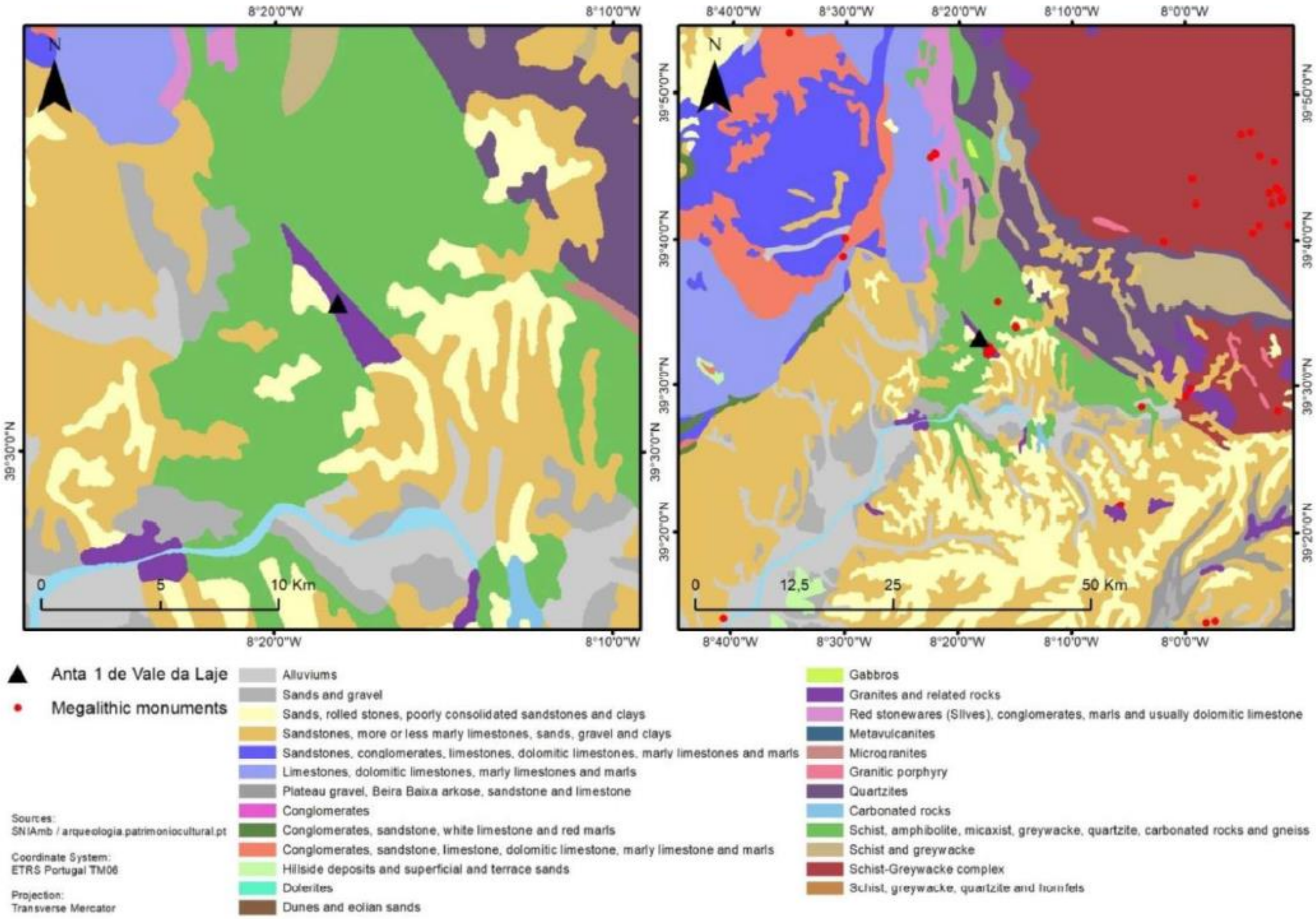


Figure 6: Map Showing the Regional Lithology with the Location of Anta 1 de Vale da Laje Site (Map: Luis Costa).

In Central Portugal, the North Ribatejo (Alto Region) is a sub-region along the banks of the Tagus Basin, covering approximately 2,500 km². It is where the Tagus, Zêzere, and Nabão Rivers converge (Oosterbeek, 1997; Cruz, 1997). Pereira et al. (2016) reported that colleagues presented some chert and silcrete samples from Spain, France, the United States, South Africa and Germany and have been kept in the LusoLit to serve as control for geochemical provenance and noted that these samples "will not" cluster with Portuguese silcrete samples.

Flint served as a crucial resource for prehistoric communities across Europe. The patterns of how specific raw materials were distributed and utilized in various cultural settings indicate that different materials held distinct values. Consequently, raw materials became a defining aspect of community life and, on a larger scale, helped illustrate interactions between communities and across regions. Studying flint involves several facets, including determining its origin, extraction methods, knapping techniques, and its various functions. However, all these investigations begin with understanding the nature, distribution, and availability of flint resources in each area (Gurova et al., 2021).

Conclusively, to understand the geological sources of flaked lithic raw materials to reconstruct prehistoric lithic economies, researchers have developed models of group behaviour, exchange networks, and craft specialization by analyzing the geographic location and distribution of raw material procurement areas (Church, 1994) coupled with range of provenance analysis techniques to determine the geological origins of lithic artefacts (Shackley, 1998).

2.5.1 The Geology of Rio Maior Basin

The Rio Maior Basin is situated in the central-western region of Portugal, encompassing the southeastern portion of Sheet 26D (Caldas da Rainha) and the northeastern portion of Sheet 30B (Bombarral) Zbyszewski (1996). It is elongated (NNW–SSE) direction measures 7.5 km in length and 3 km in width (Murray, 2019).

Stratigraphically, it comprises a thick basal unit of sand, overlain by alternating layers of diatomite and lignite. These sedimentary sequences are overlaid by Pliocene–Pleistocene and more recent alluvial deposits (Carvalho and Pereira, 1973).

Diatomite and lignite form the main superficial deposits covering about 3 km long and 1 km wide. Sand deposits exist on the western and southern margins of the basin between the Fonte da Bica and Azinheira areas. This sand deposit occurs as a small, asymmetrical syncline, with the western sand strata aligning northeastward at angles 10° to 50° (Flores, 1996), Figure 7.

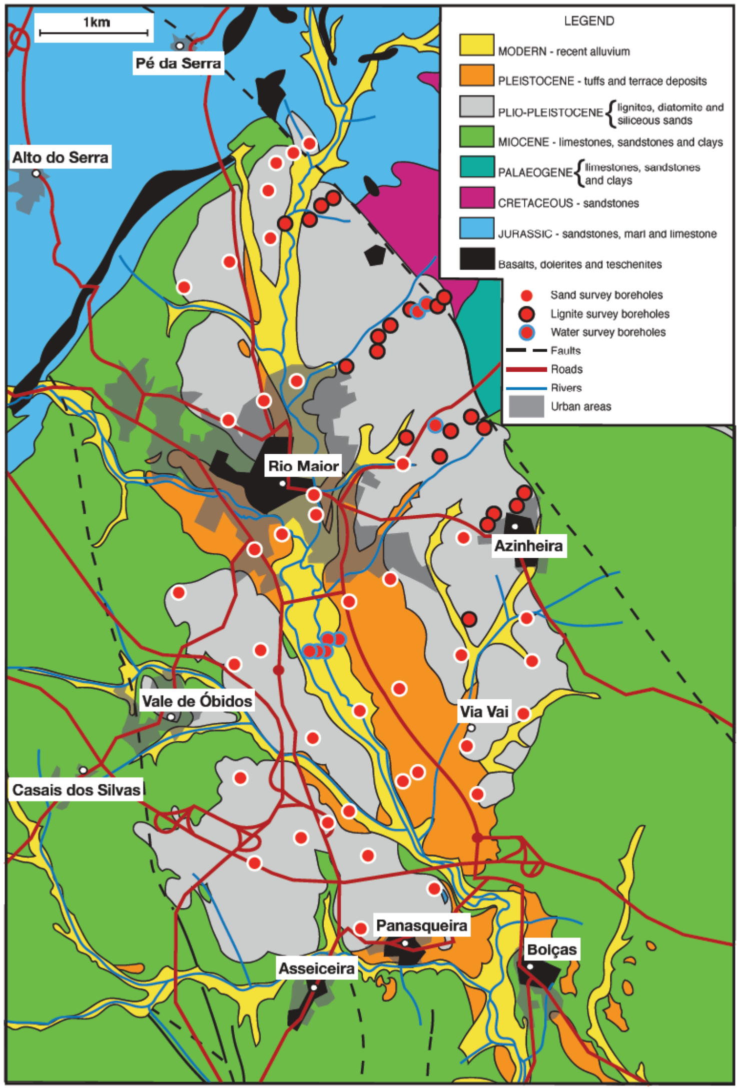


Figure 7: Geological Map of Rio Maior (Murray, 2019).

In terms of structures, it is characterised by two major faults orienting approximately 40° NW coupled with other faults oriented at 30° NE and 50° NW (Antunes et al., 1992). On the eastern margin of the basin, the Cidra Fault demarcates the contact between the diatomite–lignite sequence and the Mesozoic limestone of the Extremadura Massif. The evaporitic deposition that took place during the Late Triassic -Lower Jurassic formed the salt deposits in the Rio Maior (Uphoff et al., 2002; Calado and Brandão, 2009). The open-marine deposition that influenced the Lusitanian Basin from the Lower Jurassic to the Middle Jurassic (Mouterde et al., 1971; Mouterde et al., 1979) caused the deposition of the limestones around Rio Maior (Montenat et al., 1988). Rio Maior Flint is characterized by very fine-grained red flint of usually very high quality; however, “no microscopic analyses or chemical tests have been carried out”, as commented by (Bicho, 2004b).

Figure 8 shows the geographic locations of the Anta 1 de Vale da Laje site and the reference known raw material source of the Rio Maior geological flint, where some rock samples were collected and stored at the Earth and Memory Institute, Mação, referred to as ITM Rio Maior flint samples in this study.

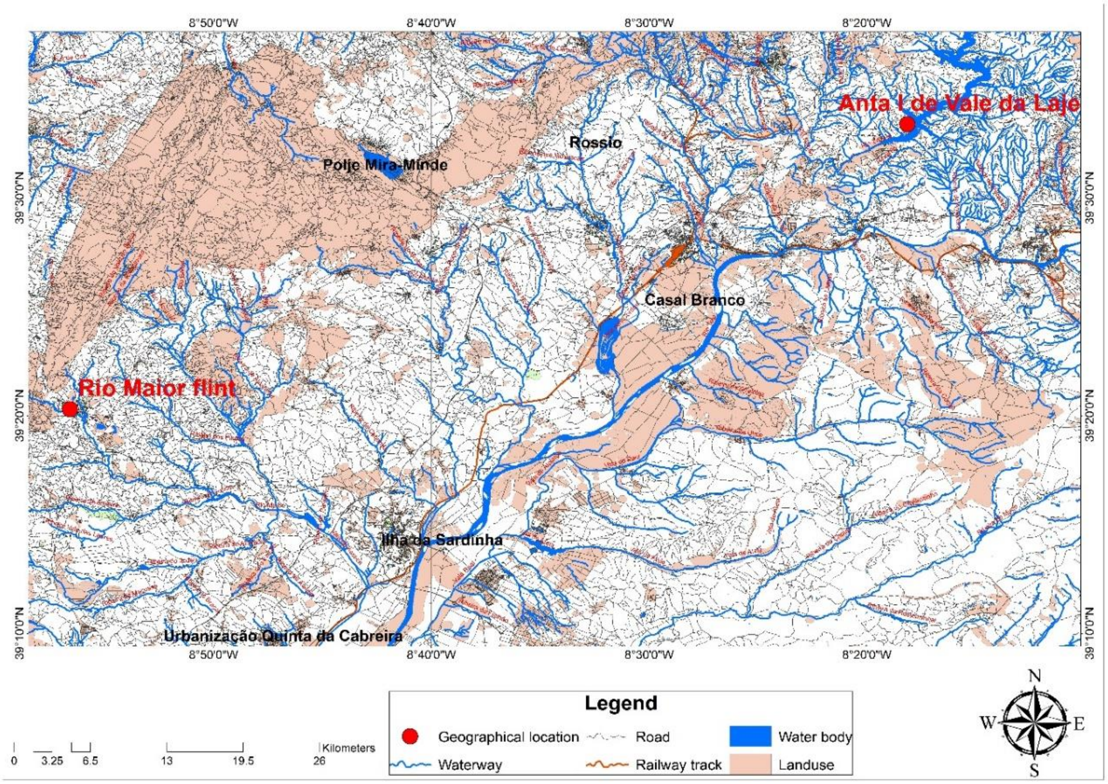


Figure 8: Geographical Locations of Anta 1 de Vale da Laje Site and Rio Maior Flint Area.

CHAPTER THREE

3.0 MATERIALS AND METHODS

3.1 Overview

This outlines the materials and methods used in geochemical fingerprinting of Neolithic lithics from Central Portugal: insights into exchange networks at Anta 1 de Vale da Laje, Tomar. The method encompasses lithic sample selection, Rio Maior flint samples, and others (petrographic and microscopic analysis of the debitage residues, analytical techniques, data processing and statistical analysis, provenance and source comparison).

3.2 Lithic Sample Selection

Lithic sample selection is a methodological technique in archaeological research used to analyse lithic tool assemblages. It helps assess the quality of raw materials, understand technological processes, and infer cognitive strategies employed by prehistoric peoples to fulfil their functional needs. In this study, lithic sample selection was applied to examine the lithics and diversity of artefacts recovered from the VL1 funerary context. This method contributes to broader research objectives, including exploring Neolithic exchange networks and determining the provenance of materials in Central Portugal. A reconnaissance visit was paid to VL1 before the lithic selection to study the characteristics of the site (Figure 9).

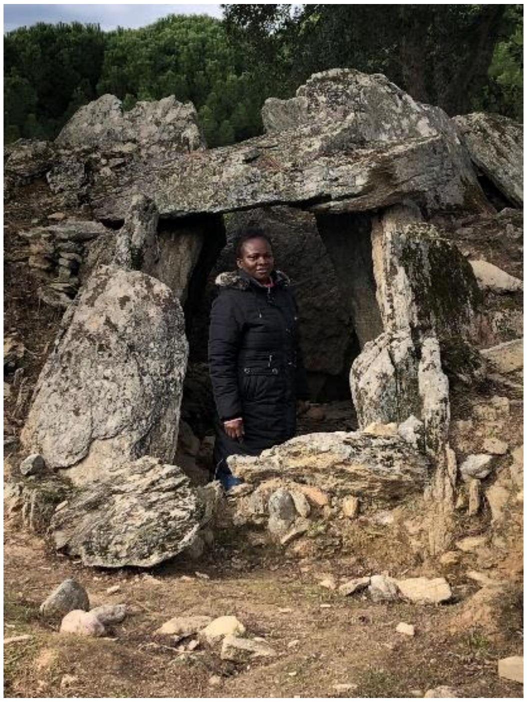


Figure 9: A Visit to the Anta 1 de Vale da Laje Megalithic Monument Site.

A total of 300 samples were studied, comprising 278 lithic artefacts from the VL1 site and 12 Rio Maior flint samples. The lithic assemblage consisted of flint (arrowheads, blades, bladelets, geometric microliths, debitage residues, and ITM Rio Maior raw material samples) and amphibolite (hand axes, pebbles, choppers/chopper cores, and flakes). All artefacts recovered are well preserved. Each artefact was measured in centimetres for length, breadth, and thickness using a ruler for larger items and a calliper for smaller ones. The weight was recorded in kilograms with an electronic scale (model KERN 440-53). Measurements were logged in MS Excel, along with details like colour and overall appearance. Colour identification was performed visually with the Munsell Soil Colour Chart (2019 production). The arrangement of all the lithic artefacts and photographs was taken using a Canon GIX, with a standardised measuring ruler placed beneath each artefact for the assemblage and each artefact (Figures 10 and 11, respectively). The assemblage was examined and classified based on raw material and physical features (colour, shape, size).

Drawings of each artefact were created by positioning the distal end toward the researcher and the proximal end oriented away, capturing dorsal, side, and ventral views. The purpose was to describe features such as typology, archaeological characteristics, rock texture, and other attributes to distinguish different flint types petrographically.

The arrowheads typology was identified and classified according to Forenbaher (1999) using the shape of the base and the edge shape, with minor modifications such as the presence/absence of the point/tip. Other features observed and noted include the serration on the edges of the arrowheads, some are slender, while some are embossed.

It was observed that the arrowheads were serrated at the edges. Some of the arrowheads are slender, and some are embossed. Most of the arrowheads show colour variations from monochromatic to polychromatic. The monochromatic colours are red, white, brown, gray, and black; polychromatic colours are reddish brown, white/brown, white/brown speckles, brown/white speckles, reddish white, white/black speckles, reddish black, white/light brown/yellow, and brown/gray.

The blades were arranged based on size, with the width doubles the length, while the bladelets were sorted so that the length equals the width.

The geometric microliths were arranged based on their shape (triangles, trapezoids, crescents, miscellaneous, and indeterminates) based on Bordes' classification (1961). Most of the geometric microliths show colour variations from monochromatic to polychromatic.

Figure 10: The Arrangement of all the Lithic Artefact Collection.

Figure 11: Photograph of all the Lithic Artefact Collection.

The monochromatic colours are red, white, brown, gray, and black; polychromatic colours are reddish brown, white/pink, whitish gray, whitish brown, whitish red, reddish brown/white, brown/red/white, brown/light gray/white, reddish brown/white/yellow, and reddish brown/white/dark brown.

The debitage residues were sorted based on their sizes and with evidence of removal of blanks such as flakes, blades, and bladelets, among others.

The hand axes were identified and classified based on complete (without broken edges) and incomplete (with broken edges).

The pebbles were identified based on the roundness and sphericity of their shape.

The choppers and chopper cores were identified based on the evidence of their sphericity and with evidence that it has been used for knapping.

The flakes were identified based on the semi-circular shape of the samples. Some of the edges of the bigger flakes show evidence of usage because they are worn out, while the small flakes do not show evidence of usage.

However, for this study, only the arrowheads (88), geometric microliths (74), debitage residues (12), and the raw material from the ITM Rio Maior raw material samples (12) were investigated for a geochemical analysis provenance study.

The inventory of these lithic assemblages was undertaken by the researcher during the analysis (Figure 12). The arrowheads and geometric microliths were analyzed for this study because they were valued for their utilitarian uses in settlements and at burial sites for symbolic funeral and ritual values during the Neolithic period. Rio Maior flint samples serve as a known source of flint whose value has been dated to the Portuguese archaeological and historical development since the Upper Paleolithic.

Figure 12: Inventory of the Lithic Artefact Collection.

3.3 Rio Maior Raw Material Flint Samples

The Rio Maior flint samples support the possible geochemical provenance analysis as a well-characterized reference collected by the Earth and Memory Institute, Mação. renowned for its flint deposit, Rio Maior was exploited to create lithic tools during the Upper Paleolithic. Its samples serve as a raw material source known reference for the geochemical analysis of the VL1 study samples. Including Rio Maior flint helps identify likely raw material sources for the lithic tools at the VL1 funerary site during the Neolithic period. It is located approximately 86.4 km southwest of VL1. Rio Maior significantly contributes to the understanding of the use of flint lithic industries in Portuguese archaeology (Heleno, 1944;1956).

Twelve samples collected from the Rio Maior flint outcrops were described and analyzed. Their physical features, especially colour, were observed directly due to their large size. The pXRF results provide a baseline for geochemical comparison and provenance attribution.

3.4 Petrographic and Microscopic Analysis of the Debitage Residues

Petrographic analysis and micropaleontology are both used to evaluate intra-source variability, but they require considerable skill and time (Herz, 2001). The petrographic and miscroscopic examination of debitage residues was conducted at the Geochemistry Laboratory, University of Ferrara, Italy, under the assistance and training of Prof. Bertola. For this study, the twelve debitage residue samples were selected based on the colour representation observed in the arrowheads and geometric microliths. The colour shades include reddish-brown, brown, and gray. The samples were first described based on colour, size, shape, texture, cortex, and their techno-typological features. Photographs of each debitage residue were taken using a photography light box (DUCLUS 30 by 30) Figure 13, the sample was placed inside the light box with a standard measured ruler set under it (Figure 14) respectively, while the dorsal and ventral sides of the sample were taken separately and labelled accordingly to avoid mixing samples. The samples are made up of retouched, unretouched artefacts, and cores. The petrographic analysis was performed with an OPTIKA binocular stereomicroscope equipped with a Moticam 2500 digital camera (5.0 megapixels, USB 2.0) and connected to a desktop. Each labelled sample was put in a petri dish containing water to reduce light reflection from the microscope and placed on the stage plate. The eyepieces were adjusted to my view, while the diopter was adjusted accordingly. The focus knob was adjusted several times to obtain the characteristic distinguishing features of the sample, such as the colours, microscopic animals and plants. The zoom knob was used to set the magnification. The sample was first viewed under a lower magnification, and eventually, the image of the sample was captured under a magnification of 10. The dorsal and ventral sides of the sample images were captured and labelled per sample. Macroscopic analysis using a binocular stereomicroscope involved examining colour, transparency, grain size, texture (Dunham, 1962), sedimentary structures, skeletal and other bioclastic components, non-skeletal elements, cortex type, and the likely depositional environment. The aim was to describe their microscopic flint characteristics (mineralogy, limestone relics, archaeological description, fossil assemblages, relative proportion of siliceous and other components, etc.) (Figure 15).

Figure 13: The DUCLUS Photographic Box.

Figure 14: Photograph of the Debitage Residue Collection.

Figure 15: Examining the Debitage Residues Under Stereomicroscope.

3.5 Analytical Techniques

The analytical techniques employed involve techno-typological and non-destructive geochemical analyses to characterize both the archaeological and geological samples. A non-destructive pXRF analytical technique was employed for the geochemical composition of lithic assemblages and Rio Maior flint samples. pXRF

3.5.1 Techno-typological Analysis

The arrowhead assemblages were classified based on typology according to Forenbaher (1999), with minor modifications related to the typology with point and without point. The arrowheads belong to a single typology (triangular) but exhibit various edge shapes to the shapes of their bases. The identified typology includes concave-based, straight-based, leaf-based, tanged-based, cruciform, rhomboid, halberd, and indeterminate. The geometric microliths include trapezoids, triangles, crescents, miscellaneous, and indeterminates. The Rio Maior flint samples were not techno-typologically classified since they are rock samples.

3.5.2 Geochemical Analysis: Portable X-Ray Fluorescence (PXRF)

The instrument used is the pXRF Bruker S1TITAN 500 Graphene window silicon drift detector (SDD), Figure 16. This was done with assistance from Prof Duarte from the Polytechnic Institute of Tomar and Virginia Latto (ITM). The samples were already arranged on a large table according to their different groups. Each sample was exposed and put on its labelled nylon. The pXRF machine was placed on the sample by taking each sample of the lithic assemblage and exposing it to X-Ray waves. To avoid contamination, the unlabeled side of each sample was scanned and recorded. The instrument was operated in Rh target tube with a (2 W, 15-40 kV, 5-100 μA) and at a 30-second exposure time. Each artefact gave diverse compositions ranging from five to nineteen elements. The elements detected by the machine for the assemblage include silicon (Si) with the highest percentage and nickel (Ni) are common to all the artefacts. Other elements platinum group elements (PGEs)- ruthenium (Ru), rhodium (Rh), palladium (Pd), and others such as indium (In), and iron (Fe) are present in nearly all the artefacts, while the remaining elements such as zircon (Zr), cobalt (Co), copper (Cu), magnesium (Mg), vanadium (V), aluminium (Al), titanium (Ti), tantalum (Ta), sulphur (S), manganese (Mn), arsenic (As), cadmium (Cd), mercury (Hg), phosphorus (P), zinc (Zn), bismuth (Bi), lead (Pb), yttrium (Y), molybdenum (Mo), and platinum (Pt) are scantly/rarely present in few or more of the samples. All the elemental composition values are measured in weight percentage (%).

Figure 16: The Portable X-Ray Fluorescence Spectrometer

3.5.3 Scanning Electron Microscope (SEM)

The use of scanning electron microscopy (SEM) for the study of archaeological materials such as bone, dentition, textile fibers, hair, plant remains, and stone tools was encouraged by

Brothwell (1969). SEM is a highly versatile analytical instrument that utilizes electrons and their interactions with a sample to provide detailed information about the sample. Its use in archaeology provides information that ranges from structural to chemical (Ponting, 2004). SEM is advantageous due to its complementary usage with optical microscopes. The selection of SEM for analyzing microscopic surfaces in archaeology depends on various factors. Its magnification range is above the optical limit of approximately 1000 times, providing a nearly continuous spectrum. Additionally, SEM offers a substantially greater depth of field- about 300 times that of optical microscopes- and high resolution, around 200 A. (Olsen, 2023). SEM displayed mineralogical textures in natural geological flints and flaked artefacts from prehistoric sites in the Massif Central (France) (Fernandes et al., 2007).

For this study, the SEM model ZEISS EVO MA 15, coupled with an Energy Dispersive X-Ray Spectroscopy (EDS) system (Aztec Oxford apparatus, SDD detector, WD 8.5 mm, EHT 20 kV), equipped with a LaB6 filament as electron source, was employed for the petrographic and microstructural characterisation of the selected debitage residues. The debitage residues were examined under the SEM to complement the pXRF results. Each of the debitage residues was scanned. Samples analyzed were classified into two groups (flints and silcretes) based on the observed structures and features.

3.6 Data Processing and Statistical Analysis

Data processing and statistical analysis were performed by using MS Excel, Principal Component Analysis (PCA), and Spatial Analysis to make inferences and provenance interpretations of the source material as compared with the known source - Rio Maior flint samples.

Principal Component Analysis (PCA): To identify clusters and reduce dimensionality.

Spatial Analysis is used to make inferences from the various stratigraphic levels about the technology, occupational history, subsistence economy, and symbolic practices during the different historical phases when the site was used.

3.7 Provenance and Source Comparison

Biplots have been used in archaeometry to display PCA results, illustrating the relationship between variables and their interpretation (Baxter, 1992; Neff, 1994; Gower and Hand, 1996). Principal Component Analysis (PCA) is employed to visually explore the relationship between cases through a graphical form (Jolliffe, 1986; Jackson, 1991; Baxter, 1994a;

Shennan, 1997). The origin of flint and silcrete artefacts was evaluated to see if they came from the Rio Maior flint. This source attribute relied on a combination of geochemical analysis, as detailed below. In archaeometry, cluster analysis is extensively employed for multivariate statistical techniques (Pollard, 1986; Everitt, 1993; Baxter, 1994a; Shennan, 1997).

Source Attribution Criteria

Artefacts were considered to originate from the Rio Maior source if their geochemical signatures satisfied the following conditions:

- Binary Plot Analysis:

- ➢ *Silicon (Si)* vs. *Nickel (Ni)* ratios for the arrowhead typology cluster with the Rio Maior flint reference samples.
- ➢ *Rhodium (Rh)* and *Palladium (Pd)* ratios cluster with the Rio Maior flint reference samples.

- Principal Component Analysis (PCA):

- ➢ The PCA for arrowhead typology showed a strong correlation with the known Rio Maior flint samples, indicating compositional similarity in multivariate space.

CHAPTER FOUR

4.0 OVERVIEW: TYPOLOGICAL AND GEOCHEMICAL RESULTS

This chapter outlines the steps taken to study and compare the arrowheads and geometric microliths from VL1 with the raw material source ITM Rio Maior flint samples. The goal is to determine whether these lithic artefacts were sourced from Rio Maior flint or otherwise. A total of 300 samples were examined, which include 278 lithic artefacts from the VL1 site and 12 ITM Rio Maior flint samples for this study.

4.1 Classification and Grouping of Lithic Types

Classification systems help archaeologists organize collections of cultural materials. Various typologies are used for artefact groupings, ranging from lithics to other materials from the historic period. Attributes used in classification include technological features and morphological details, such as concave versus convex tool edges, among others (Bell, 1958; Kleindeinst, 1967; Bordes, 1979; Luscomb, 1992; Gopher, 1994; Rosen, 1997). The total lithics assemblage analyzed for this study is presented in Table 1.

Table 1: Quantitative Analysis of the Lithics Assemblage

S/N	Lithics Assemblage	Quantity
1	Arrowheads Assemblage	88
2	Geometric microliths	74
3	Debitage residues	12
4	ITM Rio Maior Flint Samples	12
	TOTAL	**186**

The analyzed sample assemblage comprises arrowheads (88), geometric microliths (74), debitage residues (12), and ITM Rio Maior flint samples (12).

4.1.1 Arrowheads Typology

In this study, the arrowhead typology was classified into different types based on the shape of the base and the edge shape, as identified by Forenbaher (1999), with minor modifications for the presence/absence of a point/tip. All the arrowheads belong to a single typology (triangular). The identified arrowhead typologies are concave-based, straight-based, leaf-based, tanged-based, cruciform/side appendages, rhomboid/rombus eye, halberd, and indeterminates. Some of the representations for the arrowheads' typology are shown in Figures 17-24. The various occurrences of the arrowheads typology are indicated in Figure 25.

Figure 17: Concave-based Arrowheads Typology

Figure 18: Straight-based Arrowheads Typology

Figure 19: Leaf-based Arrowheads Typology.

Figure 20: Tanged-based Arrowheads Typology

Figure 21: Cruciform/Side appendages Arrowheads Typology

Figure 22: Rhomboid/Rombus eye Arrowheads Typology

Figure 23: Halberd

Figure 24: Indeterminates Arrowheads Typology

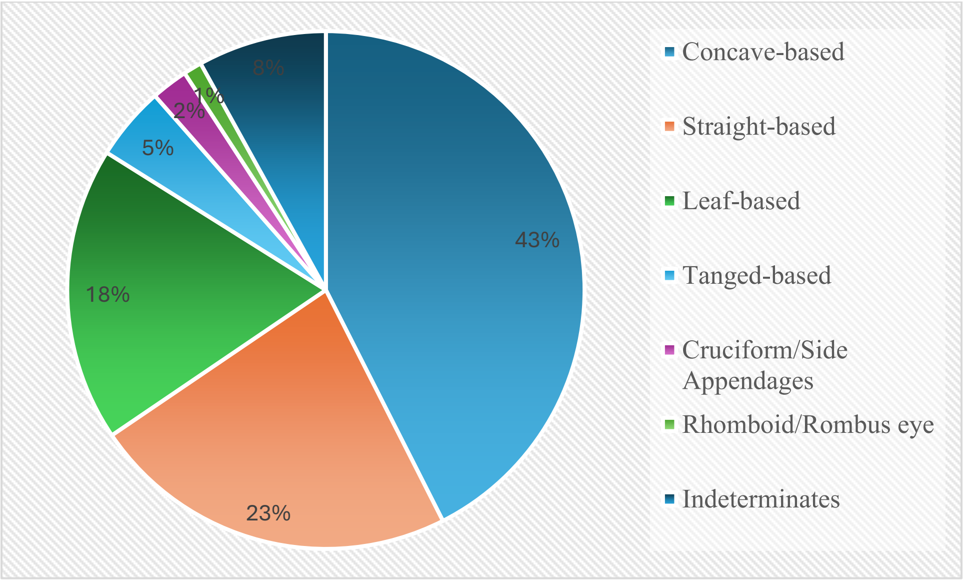


Figure 25: The Percentage Composition of the Arrowheads Typology

This includes concave-based (42%), straight-based (23%), leaf-based (18%), tanged-based (5%), cruciform/side appendages (2%), rhomboid/rombus eye (1%), and halberd (1%), indeterminate (8%).

The arrowheads are further classified based on the presence or absence of points (Figure 26). Some of the representations for the arrowheads typology with and without points include concave-based with points and without (Figures 27a and 27b); straight-based with points and without point (Figures 28a and 28b); leaf-based with points and without points (Figures 29a and 29b); tanged-based with points and without point (Figures 30a and 30b); cruciform/side appendages with points only (Figure 31); rhomboid/rombus eye with point only (Figure 32); halberd without point (Figure 33); and indeterminate with points and without points (Figure 34).

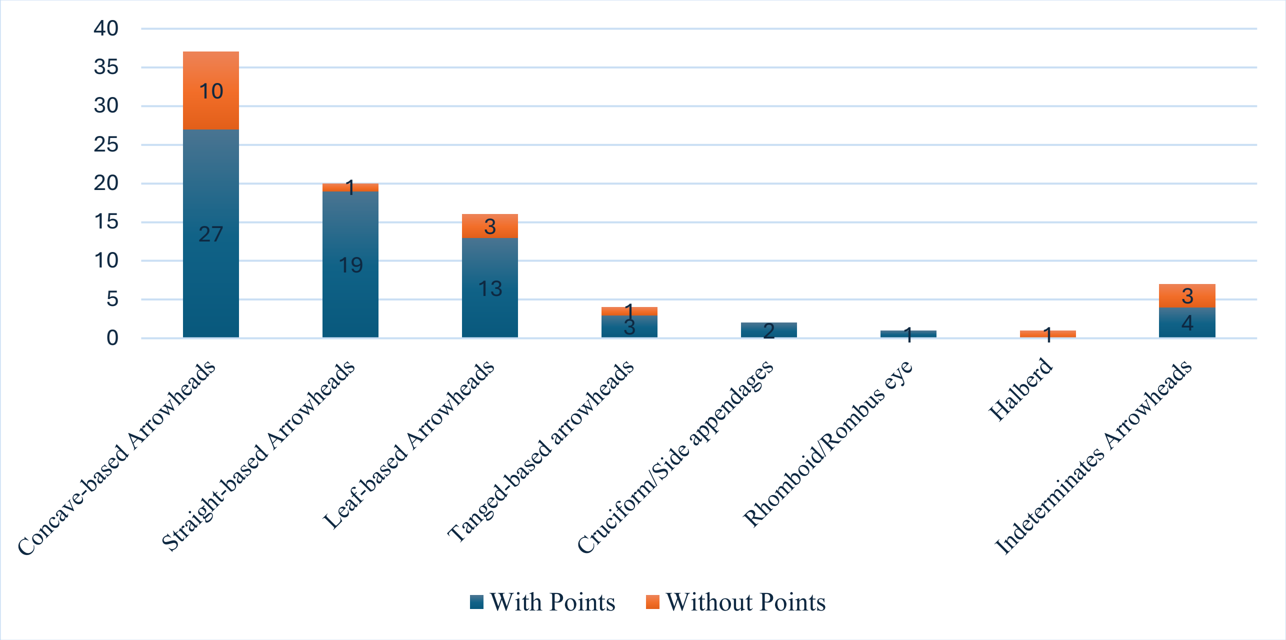
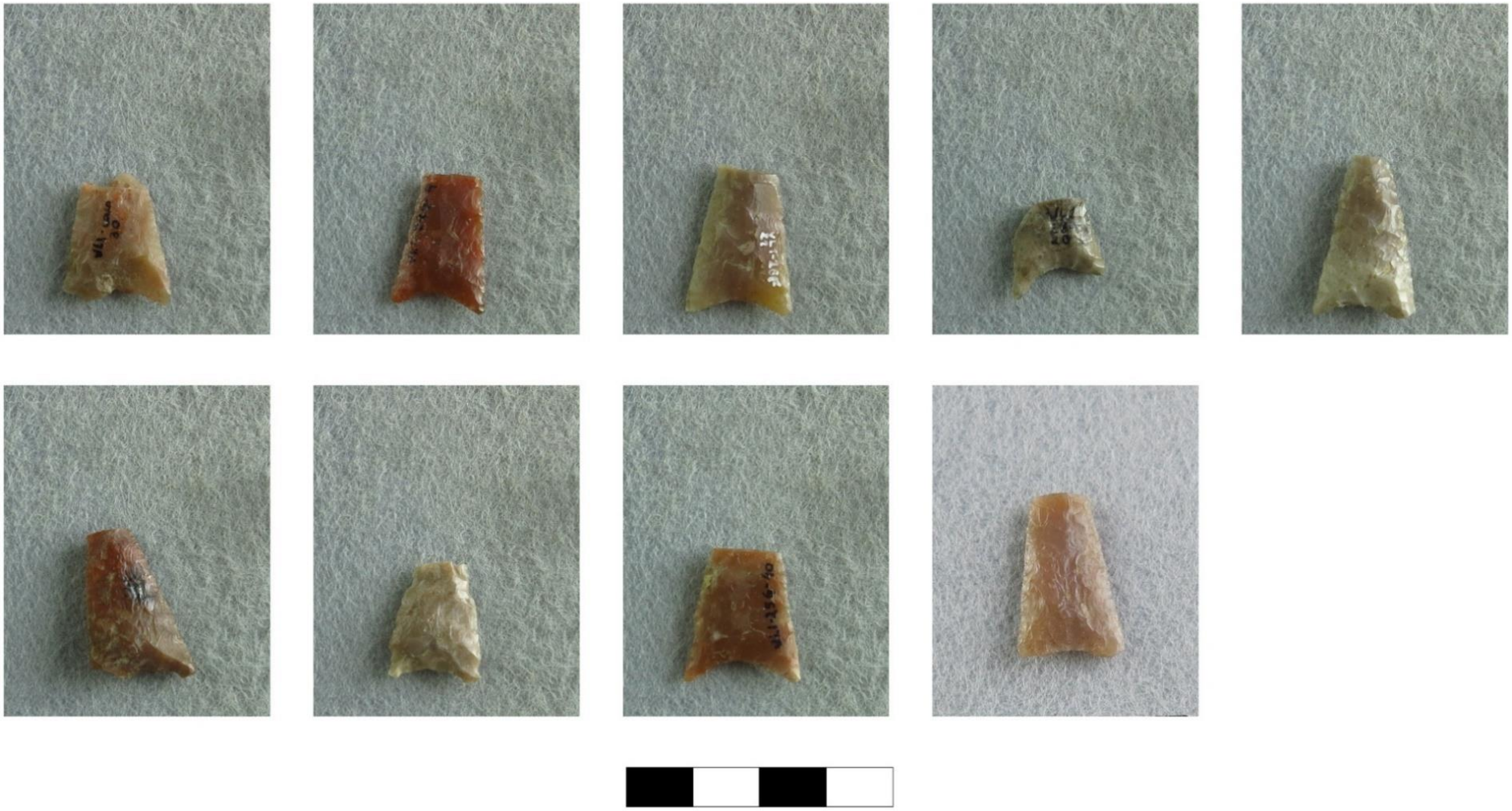


Figure 26: Arrowheads Typology With Points and Without Points

Concave-based with points (27), without points (10); straight-based with points (19), without points (1); leaf-based with points (13), without points (3); tanged-based with points (3), without point (1); rhombus only with point (1); cruciform only with points (2); halberd only without point (1); and indeterminates with points (4), without points (3).

27a: Concave-based Arrowheads With Points



27b: Concave-based Arrowheads Without Points

Figures 27a-b: Concave-based With Points and Without Points

28a: Straight-based Arrowheads With Points

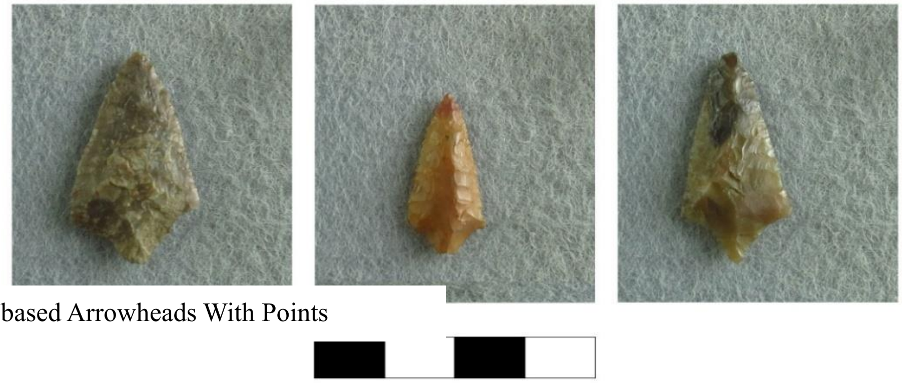
28b: Straight-based Arrowhead Without Point

Figures 28a-b: Straight-based Arrowheads With Points and Without Points.

29a: Leaf-based Arrowheads With Points

29b: Leaf-based Arrowheads Without Points

Figures 29a-b: Leaf-based Arrowheads With Points and Without Points



30a: Tanged-based Arrowheads With Points

30b: Tanged-based Arrowhead Without Point

Figures 30a-b: Tanged-based Arrowheads With Points and Without Point

Figure 31: Cruciform/Side appendages Arrowheads With Points

Figure 32: Rhomboid/Rombus eye Arrowhead With Point

Figure 33: Halberd Without Point

Figure 34: Indeterminates Arrowheads With Points and Without Points

4.1.2 Geometric Microliths

In this study, the geometric microliths were classified into different types based on their techno-morphological attributes such as the plan shape, the shape of the edge, and the use of the micro-burin technique (Tixier, 1963; Henry, 1995; Olszewski, 2001). The geometric microliths consist of five categories: triangles, trapezoids, crescents, indeterminates and miscellaneous. Some of the images are displayed in Figures 35-39. The geometric microliths compositions include triangles (27%), trapezoids (21%), crescents (16%), miscellaneous (20%), and indeterminates (16%) Figure 40.

Figure 35: Triangles Microliths

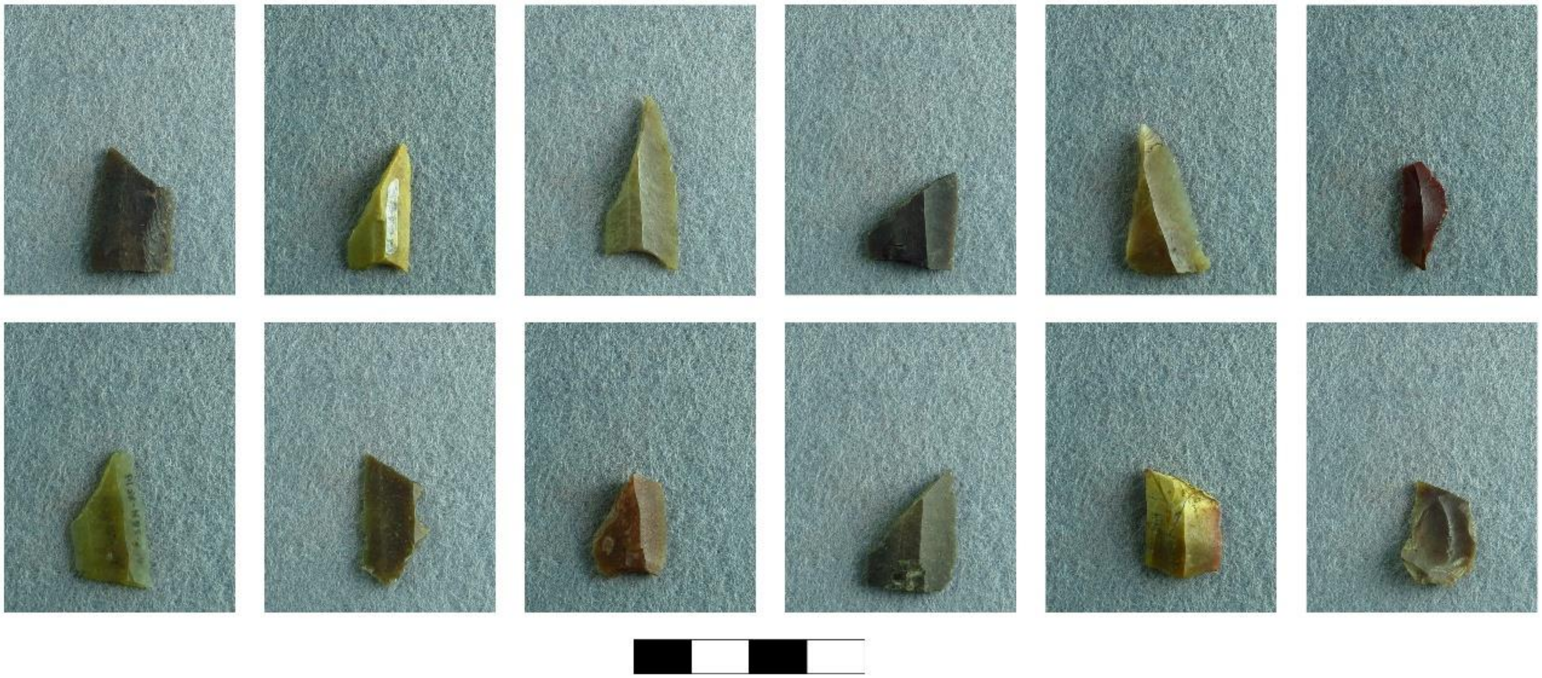


Figure 36: Trapezoids Microliths

Figure 37: Crescents Microliths

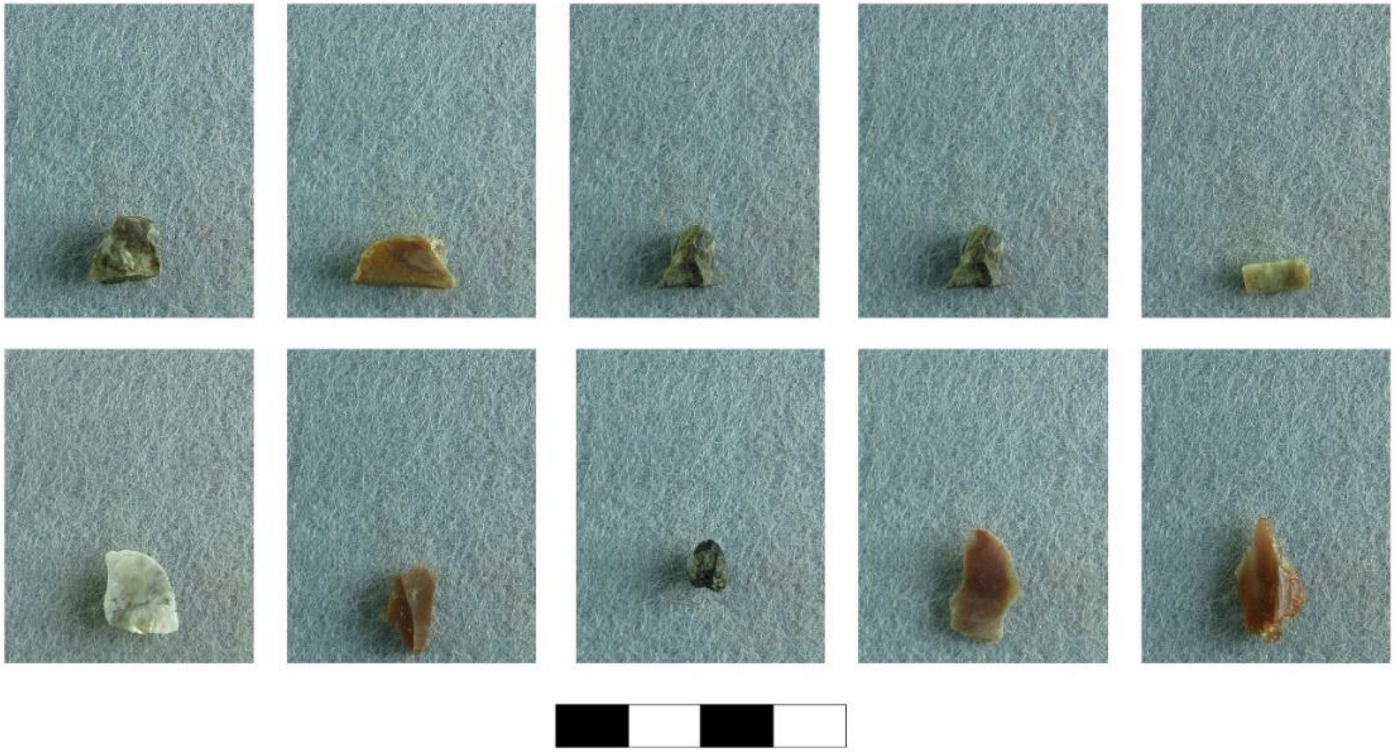


Figure 38: Indeterminates Microliths

Figure 39: Miscellaneous Microliths

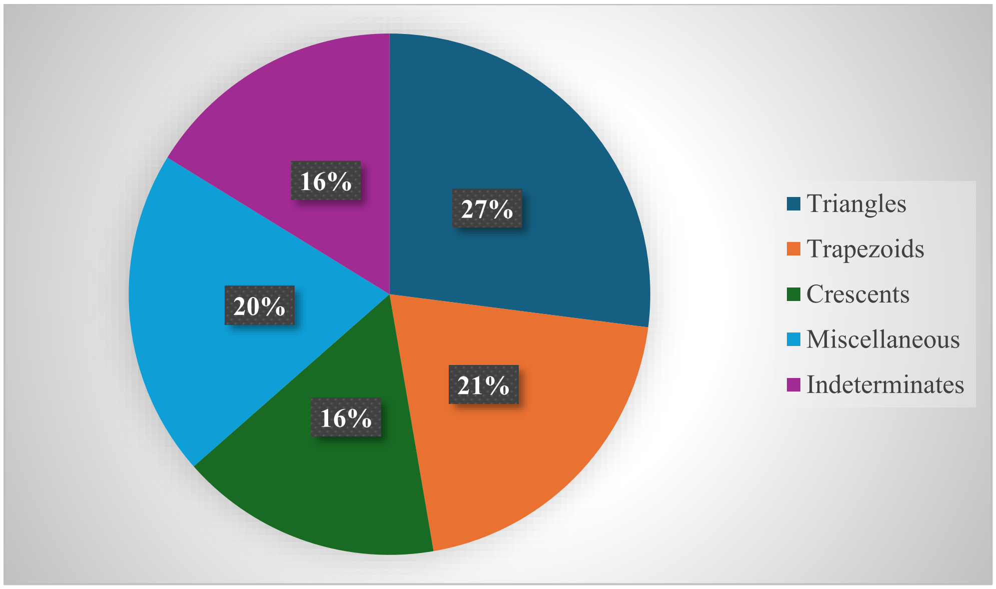


Figure 40: The Percentage Composition of the Geometric Microliths

4.1.3 Debitage Residues

The debitage residues were identified based on several blanks that had been removed from them, as described by Kadowaki et al. (2022, 2024). The images of the debitage residues are displayed in Figure 41, and the techno-typological analysis in Table 2.

Figure 41: The Debitage Residues

Table 2: Debitage Residues and Techno-typology

S/N	Artifact number	Raw material	Product	Technology-typology
1	M 23 S/C 3	Silcrete 1	Thick laminar flake	Lateral Scraper
2	REC SUP S/C 2	Silcrete 2	Cortical Flake	Unretouched
3	24 G S/C 891	Flint 1	undetermined	Multi-platform bladelet core
4	27 F Nivel 5 S.C. 31	Flint 1	Thick flake fragment	Burnt, unretouched
5	28 F S/C 12	Flint 1	Thin flake	Semi-circular scraper
6	28 H 24	Flint 1	undetermined	Multi-platform small flake core
7	28 G N6 SC 35	Flint 1	Flake fragment	Burnt, unretouched
8	26 G N8 S/C 446	Flint 2	Thick flake	Double burin/bladelet core
9	Caco S/C 648	Flint 2	Thick flake fragment	Unretouched, bipolar
10	SC1	Flint 3	Thick flake	Small flake (bipolar?) core
11	M 23 S/C 1	Flint 3	Thick flake fragment	Burnt, unretouched, bipolar
12	26 G S/C 8	Flint 3	Thick flake?	Burnt, small flake bipolar core

The debitage residue consists of flake fragments and bipolar cores of retouched and unretouched signatures. Evidence of burning was observed in some of the samples.

4.1.4 ITM Rio Maior Flint Samples

The ITM Rio Maior flint samples were identified based on colour, form, and cortex. The photographs displayed in Figure 42 and the description of physical characteristics in Table 3.

Figure 42: ITM Rio Maior Raw Material Flint Samples.

Table 3: Physical Description of Rio Maior Flint Samples

S/N	Rio Maior Sample Name	Physical Characteristics
1	Sample 1	Black, shiny, with conchoidal fracture and conical in shape.
2	Sample 2	Gray/Brown/Red/White-coloured. Tabular in form.
3	Sample 3	Gray/Brown/Milky-coloured. Tabular in form.
4	Sample 4	Gray/Brown/Red/Milky-coloured. Tabular in form and with cortex.
5	Sample 5	Gray/Brown/Milky-coloured. Tabular in form
6	Sample 6	Gray/Brown/Red/Milky-coloured. Compact, with cortex and white patches.
7	Sample 7	Brown/Red/Milky-coloured. Tabular in form and with cortex.
8	Sample 8	Brown/Red/White-coloured. Tabular in form and with cortex.
9	Sample 9	Gray/Brown/Red/Milky-coloured. With folding structures and layering.
10	Sample 10	Gray/Brown/White-coloured. Compact, with cortex, and lateritic.
11	Sample 11	Light gray/White/Milky-coloured. Compact and with cortex,
12	Sample 12	Light-dark gray/White/Milky-coloured. Compact and with cortex,

The Rio Maior flint samples showed a range of colours from monochromatic to polychromatic. Black only appeared in one sample as the dominating colour. Grey appeared in nine samples, brown in nine samples, milky hues appeared in eight samples, white appeared in six samples, and red appeared in six samples.

4.2. Portable X-Ray Fluorescence Spectrometry Results (PXRF).

Portable XRF and EDXRF are non-destructive techniques that offer insights into the geochemical provenance of raw materials used to create the artefacts circulated in prehistoric times (Richards, 2019). The effective provenance techniques for materials like lithics, volcanic glass and obsidian artefacts have been performed using pXRF and EDXRF (Tykot, 2002; Carter and Shackley, 2007; Craig et al., 2007; Summerhayes, 2009; Phillips and Speakman, 2009; Shackley, 2010; Sheppard et al., 2010, 2011; McCoy et al., 2011; Galipaud et al., 2014). The pXRF results are displayed in the Appendix Tables 1-15. Appendix Table 1 is the debitage residues pXRF result; Appendix Table 2 is the concave-based arrowheads pXRF result; Appendix Table 3 is the straight-based arrowheads pXRF result; Appendix Table 4 is the leaf-based arrowheads pXRF result; Appendix Table 5 is the tanged-based arrowheads pXRF result; Appendix Table 6 is the cruciform/side appendages arrowheads pXRF result; Appendix Table 7 is the rhomboid/rombus eye arrowheads pXRF result; Appendix Table 8 is the halberd pXRF result; Appendix Table 9 is the indeterminates arrowheads pXRF result; Appendix Table 10 is the triangles microliths pXRF result; Appendix Table 11 is the trapezoids microliths pXRF result; Appendix Table 12 is the crescents microliths pXRF result; Appendix Table 13 is the indeterminates microliths pXRF result; Appendix Table 14 is the miscellaneous microliths pXRF result; and Appendix Table 15 is the ITM Rio Maior flint samples pXRF result.

4.3 Elemental Composition

The geochemical compositions of rocks or sediments are the result of the complex interplay of several factors that reveal evidence of provenance, history, climate, tectonic settings, weathering of source rocks, and sedimentary processes (Johnsson, 1993). Chemical compositions of terrigenous sediments are the products of source rock nature, chemical weathering, climatic events, transportation, diagenetic factors and erosion (Nesbitt and Young, 1982; Taylor and McLennan,1985; McLennan et al., 1993; Cox and Cullers,1995; Garzanti et al, 2013; Hossain et al., 2017). The natural and anthropic processes are reflected in the elemental composition of geological and archaeological materials. This fact has been employed in this study to determine the source of the raw materials for the VL1 arrowheads and geometric microliths. In this study, the commonest element to all the specimens (artefacts and rock sample) and with the highest values is Si with weight range of (97.15%-36.14%), the highest value is observed in straight based arrowhead (24 G S/C 896) while the least weight was in indeterminates microliths (22H S/C 24) this is followed by the Platinum Group Elements (PGEs) that include Ru with weight range of (6.34%-0.78%), with highest weight in straight-based arrowheads (28 H S/C 11) least weight in ITM Rio Maior Flint (Sample 8), Rh with weight range (5.86%-1.01%) with highest weight in straight-based arrowhead (28 H S/C 11) and least weight in straight-based arrowhead (29 H 24), and Pd with weight range of (4.89%-0.38%) with highest weight in straight-based arrowhead (29 H 24) and least weight in leaf-arrowhead (27 G S/C 74 and 28 F 11). Ni is common to all the specimens but in low numbers, with a weight range of (0.17%-0.01%), with the highest weight in the triangles microliths (28 F S/C 14) and the least weight in the debitage residue and leaf-based arrowhead (24 G S/C 891 and 27 G S/C 74) respectively. Fe is present in some of the specimens with a weight range of (14.16%-0.04%), with the highest weight in the blade (24 H S/C 19), the least weight in debitage residue (23 H S/C 4), straight-based arrowhead (24 G S/C 1024, 28 G S/C 202), leaf-based arrowhead (28 H SC 9), blade (29 H-26), bladelet (Caco S/C 19, Rec Sup S/C 1), trapezoid (27 G SC 184), triangles (24 F S/C 45), miscellaneous microliths (25 G n4 S/C 47a), ITM Rio Maior Flint (Sample 4). The other elements such as (Zr), (Co), (Cu), (Mg), (V), (Al), (Ti), (Ta), (S), (Mn), (As), (Cd), (Hg), (P), (Zn), (Bi), (Pb), (Y), (Mo), and (Pt) are scantly present.

The images of the specimens with the highest and lowest weights of the PGEs are further considered, Ru with the highest weight (6.34%) in straight-based arrowhead (28 H S/C 11), the least weight (0.78%) in ITM Rio Maior flint (Sample 8) Figures 43 and 44, respectively. Rh with the highest weight (5.86%) straight-based arrowhead (28 H S/C 11) Figures 43, and the least weight (1.01%) in a straight-based arrowhead (29 H 24) Figure 45 While Pd with the highest weight of (4.89%) in straight-based arrowhead (29 H 24) Figure 45 and the least weight of (0.38%) in leaf-based arrowheads (27 G S/C 74 and 28 F 11) Figures 46 and 47 respectively.

Figure 43: Straight-based Arrowhead 28 H S/C 11

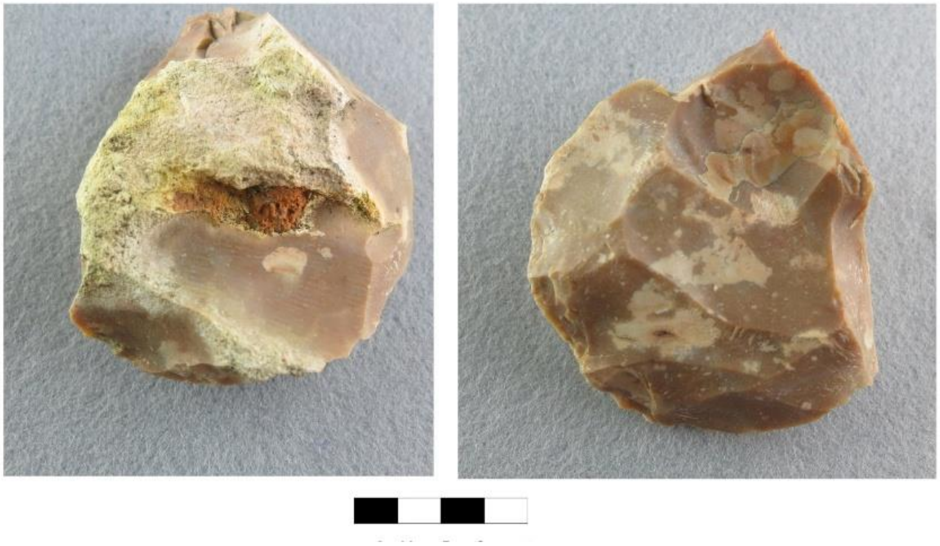


Figure 44: ITM Rio Maior Flint Sample 8

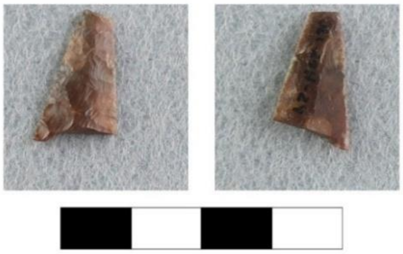


Figure 45: Straight-based Arrowhead 28 H 24

Figure 46: Leaf-based Arrowhead 27 G S/C 74

Figure 47: Leaf-based Arrowhead 28 F 11

4.3.1 Elemental Composition of Geometric Microliths vs ITM Rio Maior Flint Samples.

Comparison is made between the Si vs Ni, and the Pd vs Rh of the geometric microliths and ITM Rio Maior flint. Figures 48 and 49 respectively.

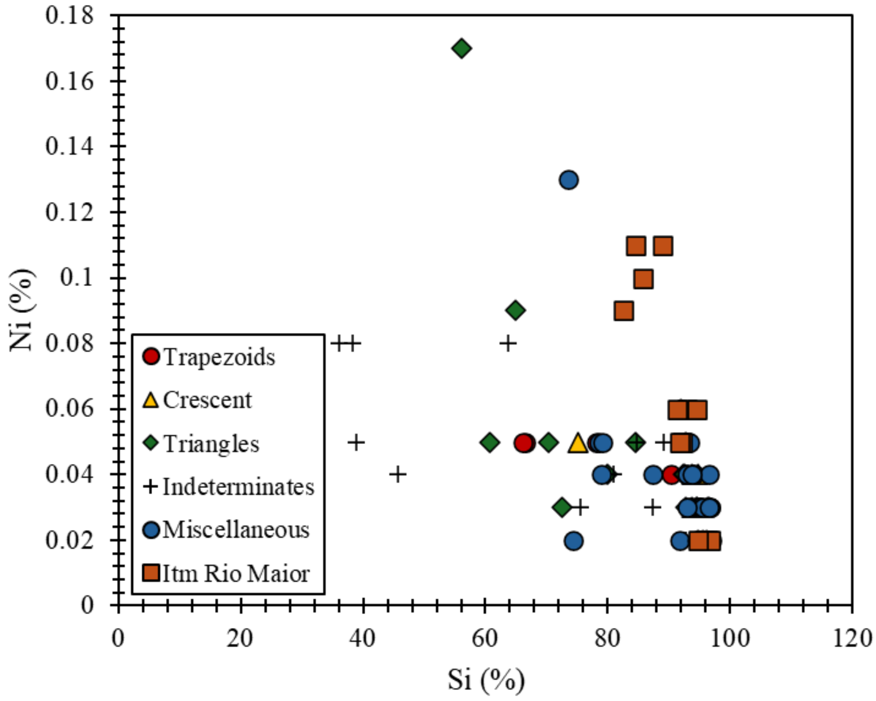


Figure 48: The Plot of Si vs Ni of Geometric Microliths and ITM Rio Maior Flint

Some of the geometric microliths, such as trapezoids, crescents, triangles, indeterminates and miscellaneous cluster around the ITM Rio Maior flint, which suggests that they were probably sourced from Rio Maior flint. However, some triangles, indeterminates, and miscellaneous indicate different sources as they do not cluster with the ITM Rio Maior flint.

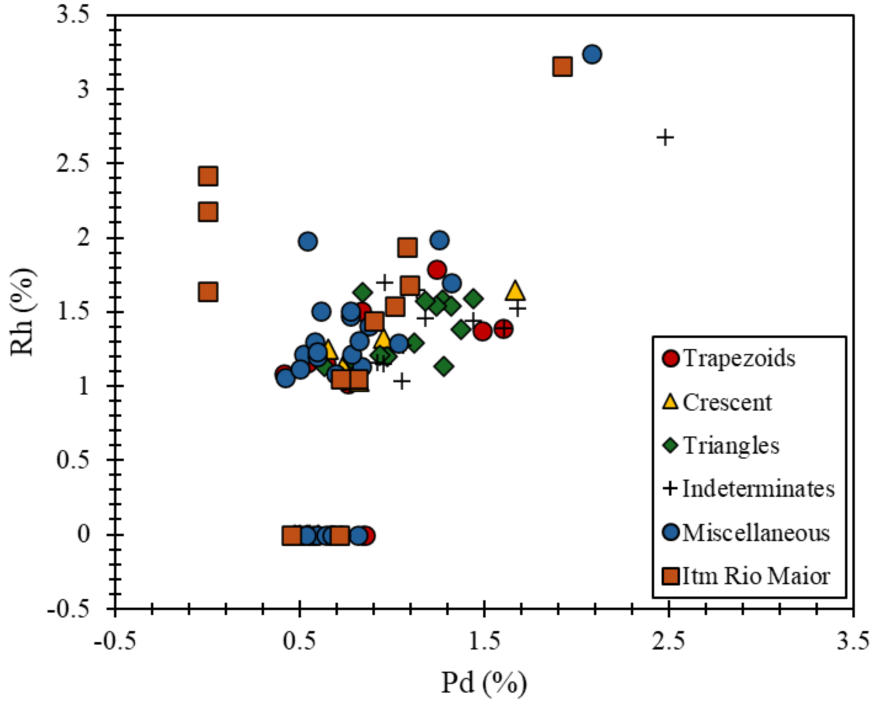


Figure 49: The Plot of Pd vs Rh of Geometric Microliths and ITM Rio Maior Flint

Some of the geometric microliths, such as trapezoids, crescents, triangles, indeterminates, and miscellaneous cluster around ITM Rio Maior flint, probably sourced from Rio Maior flint. However, some crescents, indeterminates and miscellaneous indicate different sources, as they do not cluster with the ITM Rio Maior flint.

4.3.2 Elemental Composition of Debitage Residues vs ITM Rio Maior Flint.

Comparison is made between the Si vs Ni, and the Pd vs Rh of the debitage residues and ITM Rio Maior flint. Figures 50 and 51 respectively.

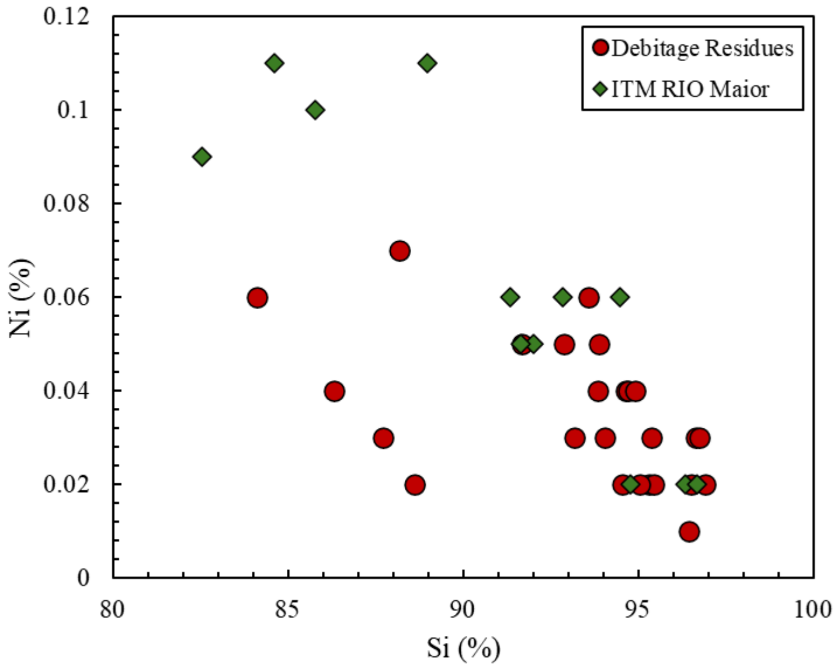


Figure 50: The Plot of Si vs Ni of the Debitage Residues and ITM Rio Maior Flint

Very few of the debitage residues cluster with the ITM Rio Maior flint, which probably indicates the same source. However, the majority of the debitage residues are scattered, which suggests different sources.

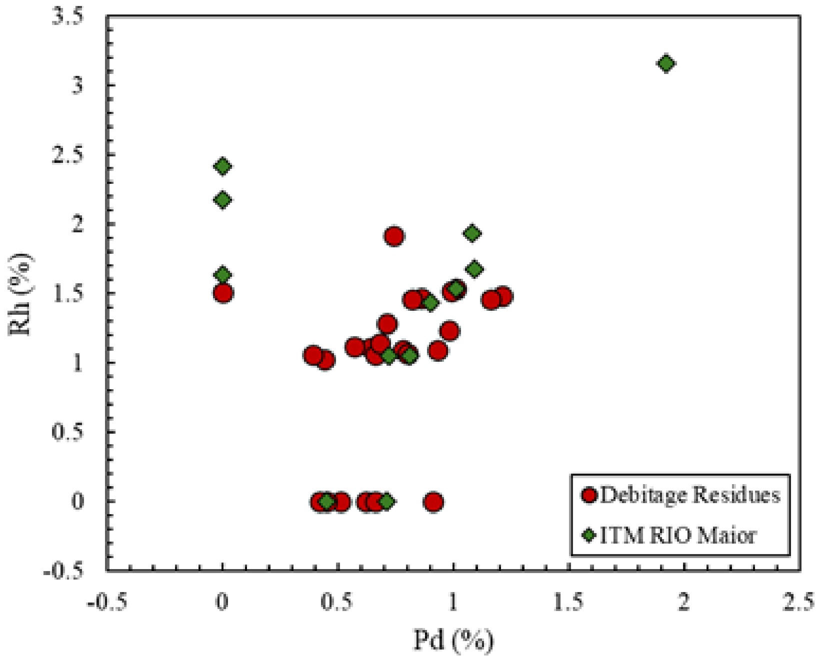


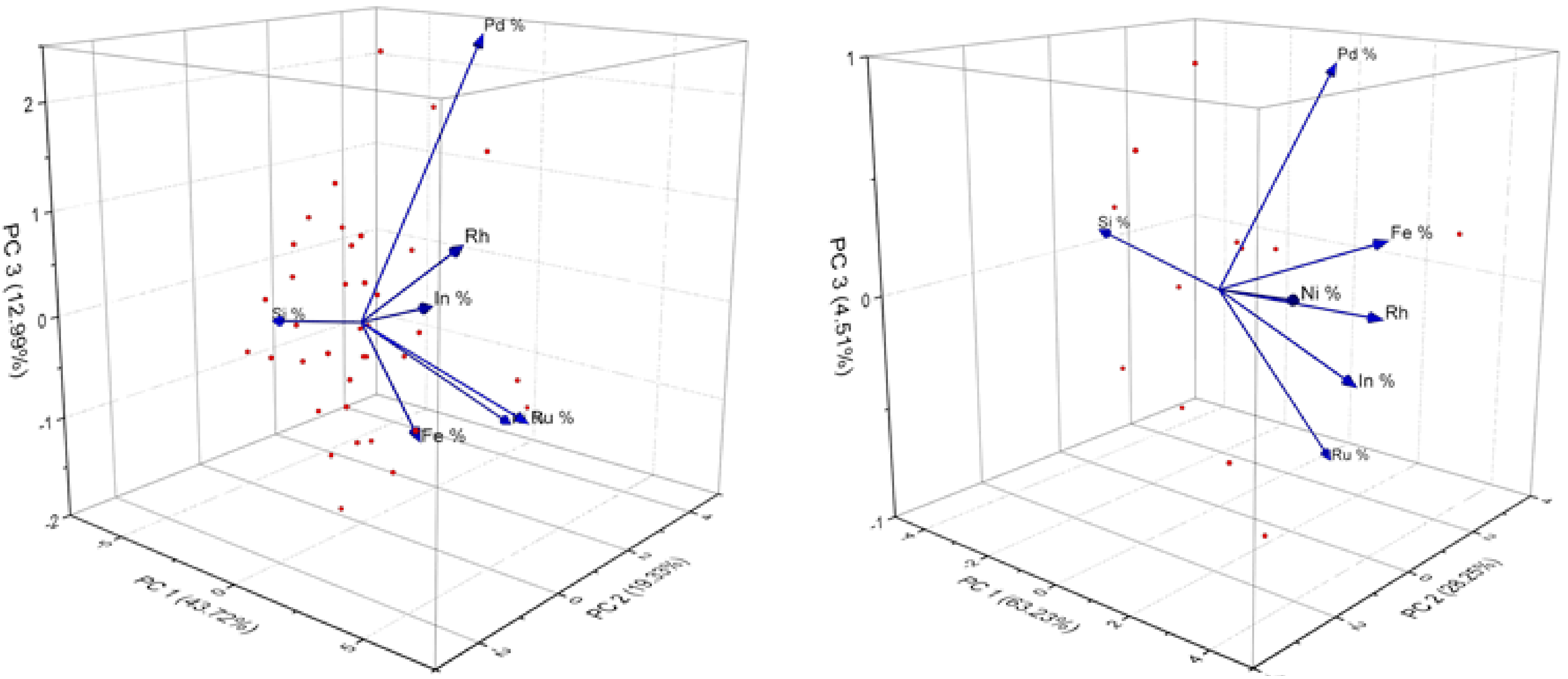
Figure 51: The Plot of Pd vs Rh of the Debitage Residues and ITM Rio Maior Flint

A few of the debitage residues cluster with ITM Rio Maior flint, which suggests the same source. While some of the debitage residues are clustered, which indicates the same source. Some of the ITM Rio Maior flint indicates different sources.

4.3.3 Principal Component Analysis Results (PCA)

In provenance investigations of raw materials, PCA replaces original building components with others that are aesthetically and physicochemically similar. PCA aids in summarising data by highlighting similarities and differences among groups within large datasets. It produces a straightforward, illustrative, and comprehensive graph that uncovers distinct groups within the samples based on their properties. PCA has been employed to examine the characterization, technology, and weathering condition of building materials from historical monuments coupled with cultural heritage monuments. (Moropoulou and Polikreti, 2009). PCA is an orthogonal linear transformation that transforms data into a new coordinate system. Each principal component (PC) is a linear combination of the variables that captures the maximum possible variance in the dataset. PC1 is the dominant axis, PC2 is the secondary axis, and PC3 is the axis with the least variance (Davies, 1986).

PCA was performed on 88 arrowheads, 74 geometric microliths, 12 debitage residues, and 12 ITM Rio Maior flint samples. It illustrates the geochemical variability within the samples, displaying the relationships between major and trace elements (Fe, In, Si, Rh, Ru, Ni, Pd) across the sample population. The analysis reveals distinct geochemical associations and provides insights into the elemental behaviour within these microscale mineral phases. The PCA of each of these artefacts is compared with the ITM Rio Maior flint samples, arrowheads assemblage (Figures 52-56), geometric microliths (Figures 57-61), and debitage residues (Figure 62).



52a: PCA of Concave-based Arrowheads

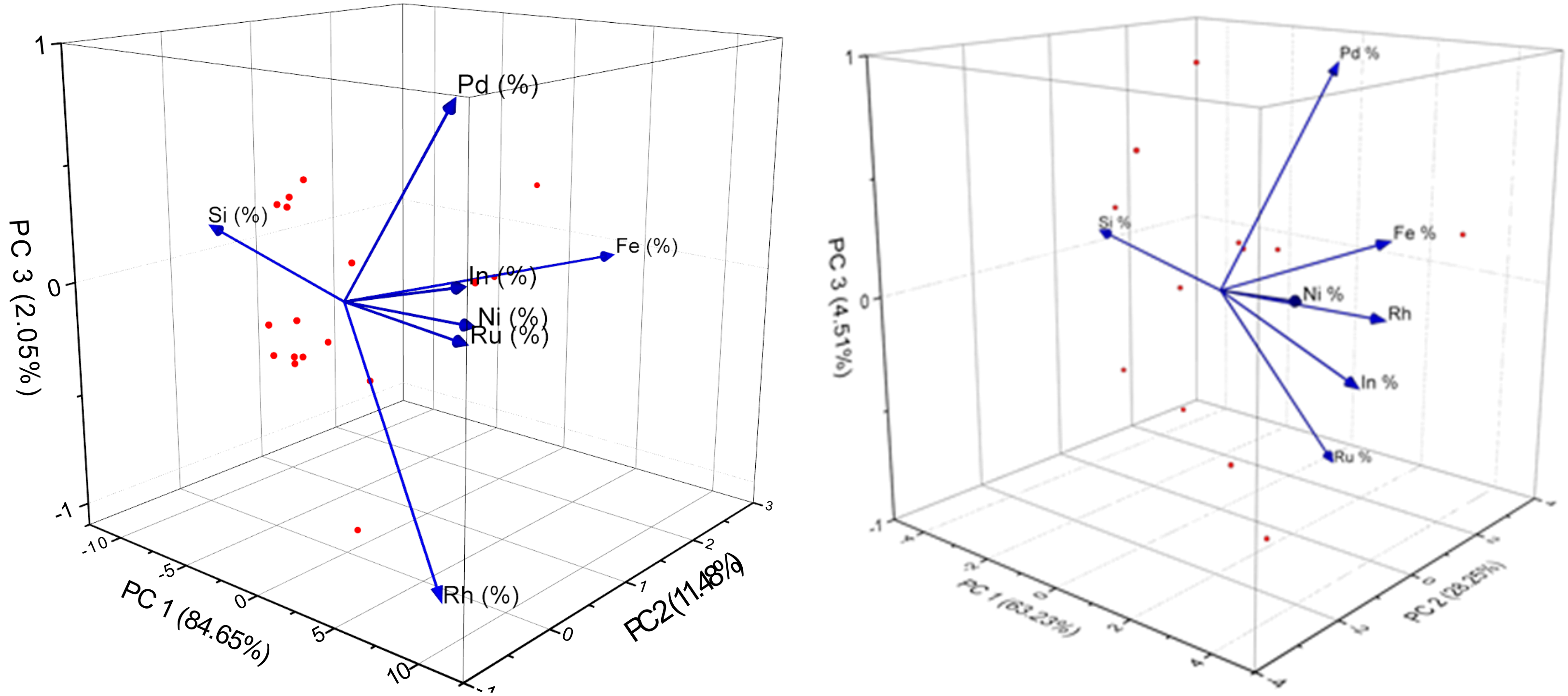
52b: PCA of ITM Rio Maior Flint

Figures 52a-b: PCA of Concave-based Arrowheads and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig.52a: PCA of concave-based arrowheads shows that higher Si values primarily determine PC1 (43.73% of variance). PC2 (19.33%) is influenced by Ru, Rh, Fe, and In, while PC3 (12.99%) shows variations linked to Pd and Ni.

Fig.52b: PCA of ITM Rio Maior flint indicates that PC1 (63.23%) is dominated by Si values. PC2 (28.25%) includes variation related to Ru and Pd, and PC3 (4.51%) is given by In, Rh, Fe, and Ni values.



53a: PCA of Straight-based Arrowheads

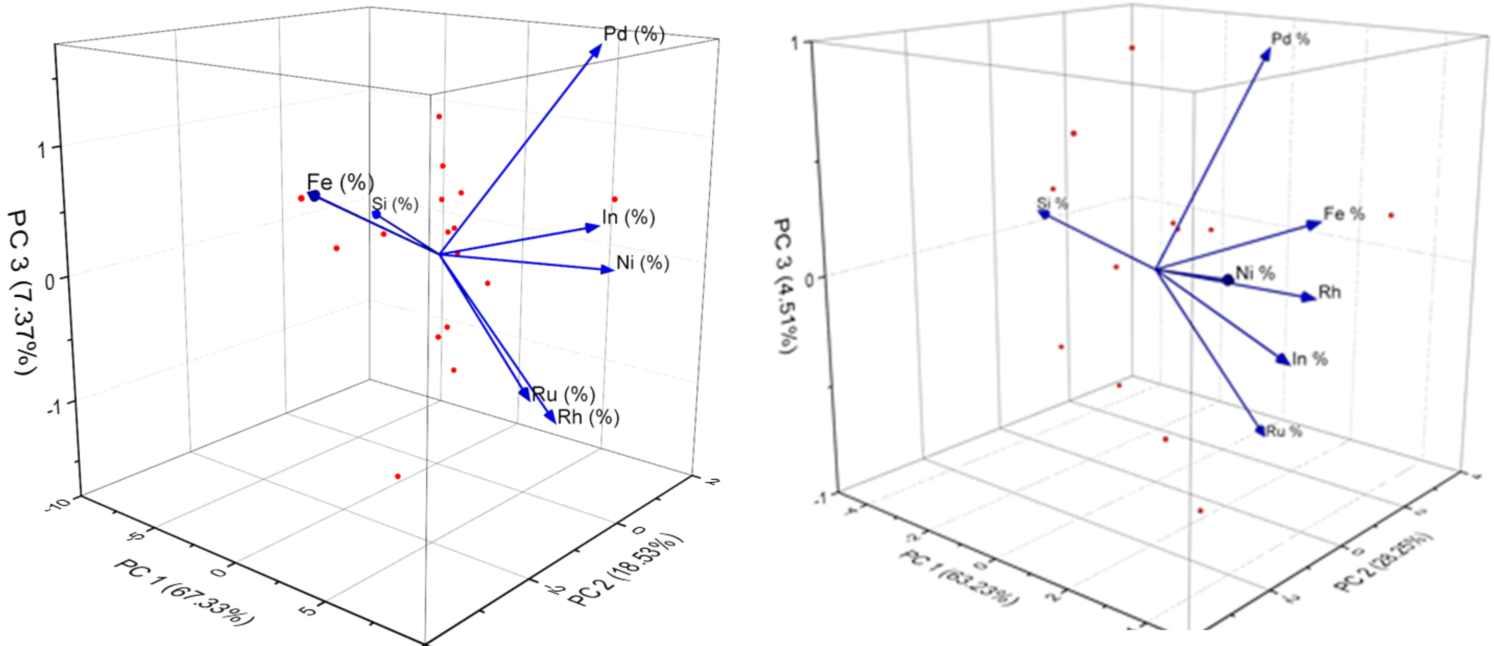
53a: PCA of ITM Rio Maior Flint

Figures 53a-b: PCA of Straight-based Arrowheads and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig.53a: PCA of straight-based arrowheads show that Si values primarily determine PC1 (66.45% of variance). PC2 (11.46%) is influenced by Ru, In, Ni, Pd, and Rh, while PC3 (2.05%) shows variations linked to Fe only.

Fig.53b: PCA of ITM Rio Maior flint. PC1 (63.23%) is dominated by Si, PC2 (28.25%) is given by Ru and Pd, and PC3 (4.51%) by In, Rh, Fe, and Ni values.



54a: PCA of Leaf-based Arrowheads

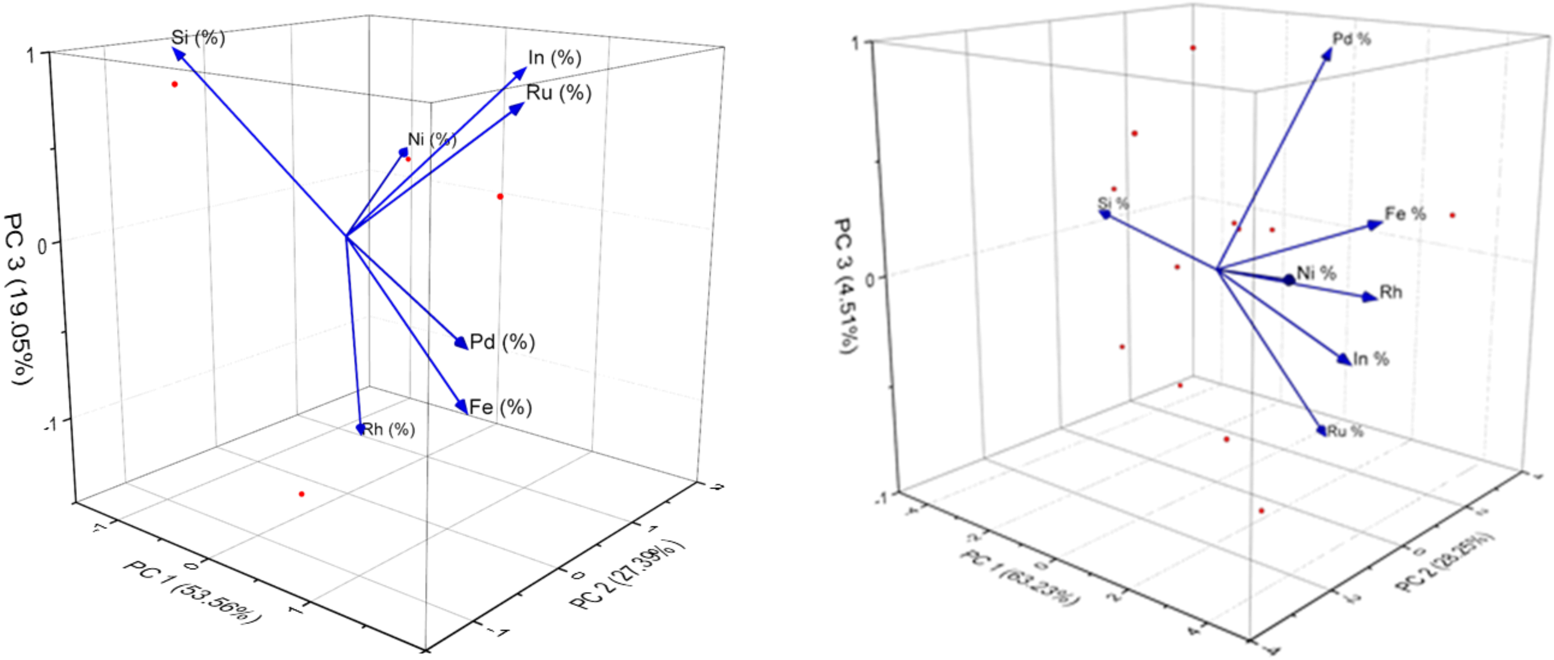
54b: PCA of ITM Rio Maior Flint

Figures 54a-b: PCA of Leaf-based Arrowheads and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig.54a: PCA of leaf-based arrowheads shows that Si values primarily determine PC1 (67.33% of variance). PC2 (18.53%) is influenced by Ru and Rh, while PC3 (7.37%) shows variations linked to In, Ni, Pd, and Fe.

Fig.54b: PCA of ITM Rio Maior flint. PC1 (63.23%) is dominated by Si, PC2 (28.25%) is given by Ru and Pd, and PC3 (4.51%) by In, Rh, Fe, and Ni values.



55a: PCA of Tanged-based Arrowheads

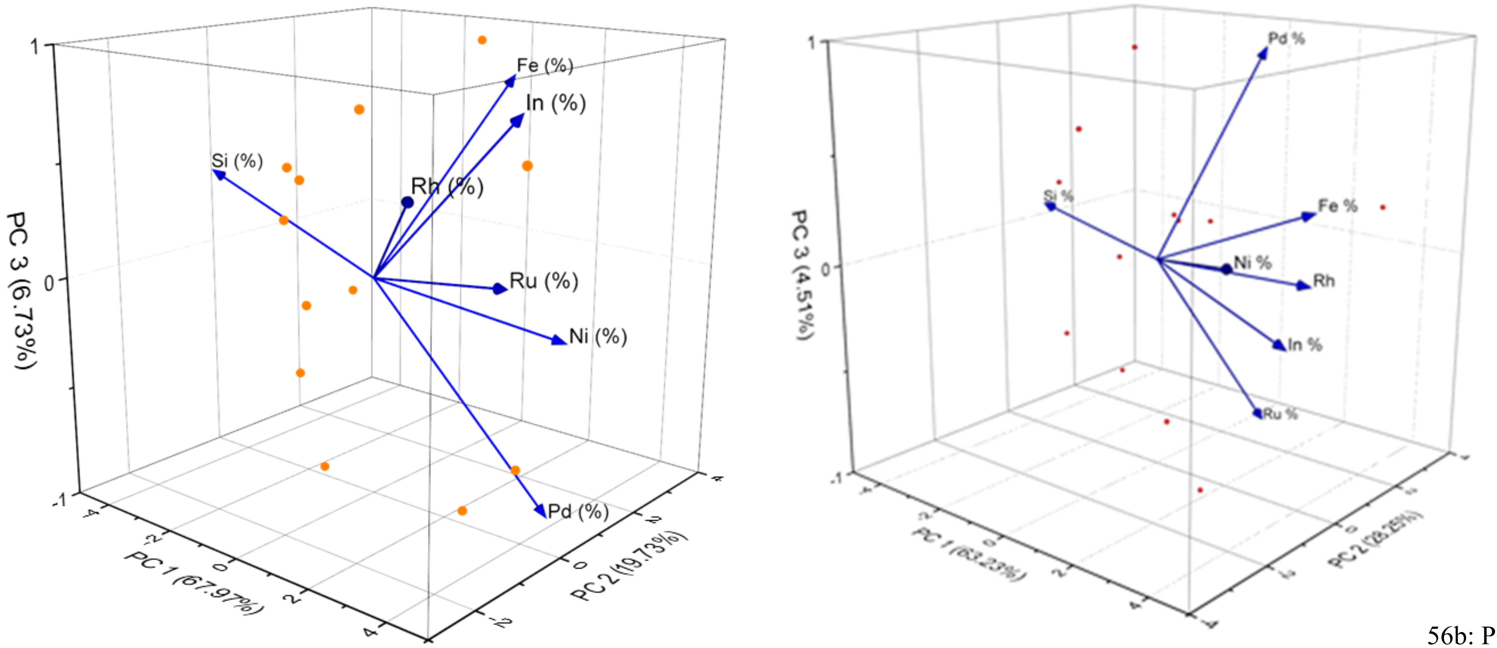
55b: PCA of ITM Rio Maior Flint

Figures 55a-b: PCA of Tanged-based Arrowheads and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig. 55a: PCA of tanged-based arrowheads shows that Si values primarily determine PC1 (53.56% of variance). PC2 (27.39%) is influenced by Rh and Ni, while PC3 (19.05%) shows variations linked to Ru, In, Pd, and Fe.

Fig.55b: PCA of ITM Rio Maior flint. PC1 (63.23%) is dominated by Si, PC2 (28.25%) is given by Ru and Pd, and PC3 (4.51%) by In, Rh, Fe, and Ni values.



56b: PCA of ITM Rio Maior Flint

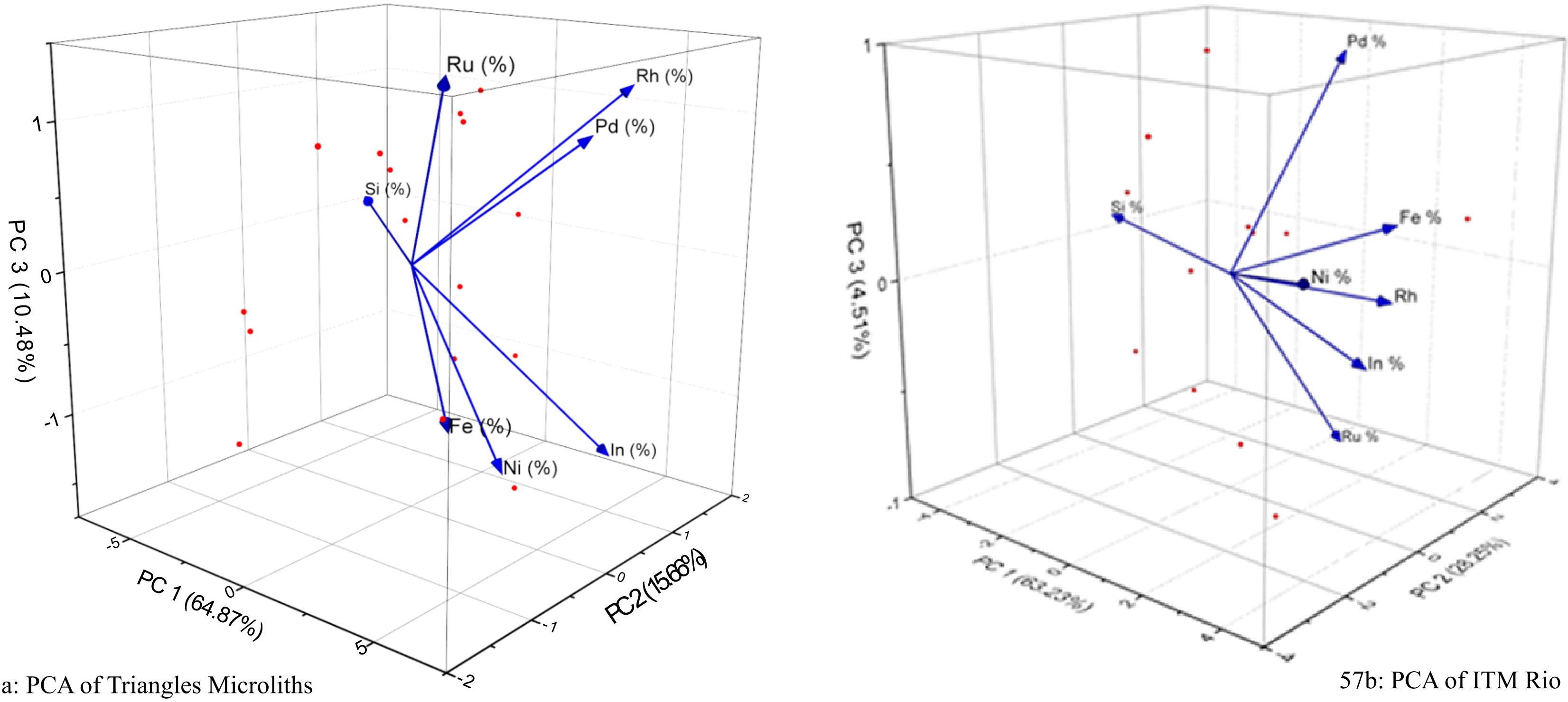
56a: PCA of Indeterminates Arrowheads

Figures 56a-b: PCA of Indeterminates Arrowheads and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig.56a: PCA of Indeterminates arrowheads shows that PC1 (67.97%) is primarily determined by Si values, PC2 (19.73%) is influenced by Ru, In, Rh, and Ni, while PC3 (6.73%) shows variations linked to Fe and Pd.

Fig.56b: PCA of ITM Rio Maior flint. PC1 (63.23%) is dominated by Si, PC2 (28.25%) is given by Ru and Pd, and PC3 (4.51%) by In, Rh, Fe, and Ni values.



57a: PCA of Triangles Microliths

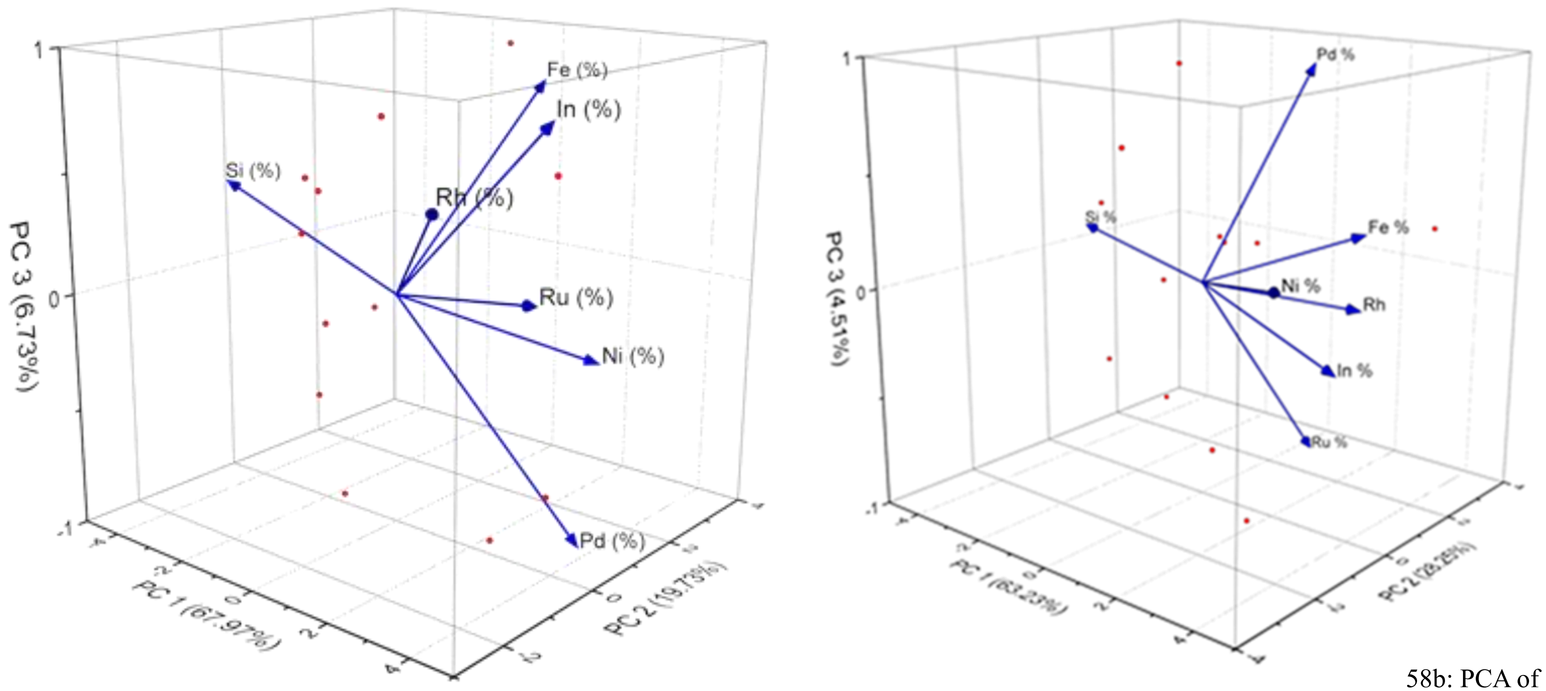
57b: PCA of ITM Rio Maior Flint

Figures 57a-b: PCA of Triangles Microliths and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig.57a: PCA of triangles microliths shows that PC1 (64.87%) is primarily determined by Si values, PC2 (15.66%) is influenced by Ru, Fe, Ni, while PC3 (10.48%) shows variations linked to In, Rh, and Pd values.

Fig.57b: PCA of ITM Rio Maior flint. PC1 (63.23%) is dominated by Si, PC2 (28.25%) is given by Ru and Pd, and PC3 (4.51%) by In, Rh, Fe, and Ni values.



58b: PCA of ITM Rio Maior Flint

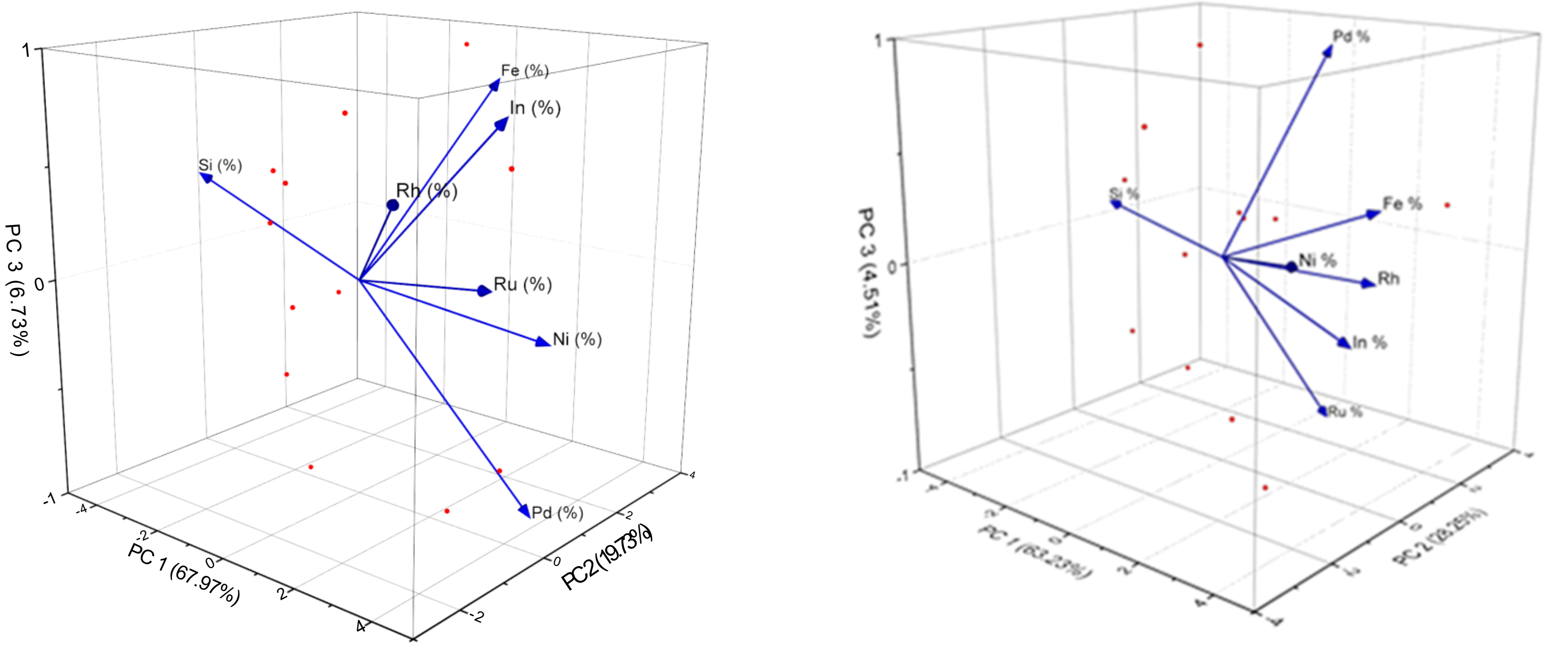
58a: PCA of Trapezoids Microliths

Figures 58a-b: PCA of Trapezoids Microliths and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig.58a: PCA of trapezoid microliths shows that PC1 (67.97%) is primarily determined by Si values, PC2 (19.73%) is influenced by Ru, In, Pd, Ni, Rh, while PC3 (6.73%) shows variations linked to Fe only.

Fig.58b: PCA of ITM Rio Maior flint. PC1 (63.23%) is dominated by Si, PC2 (28.25%) is given by Ru and Pd, and PC3 (4.51%) by In, Rh, Fe, and Ni values.



59a: PCA of Crescent Microliths

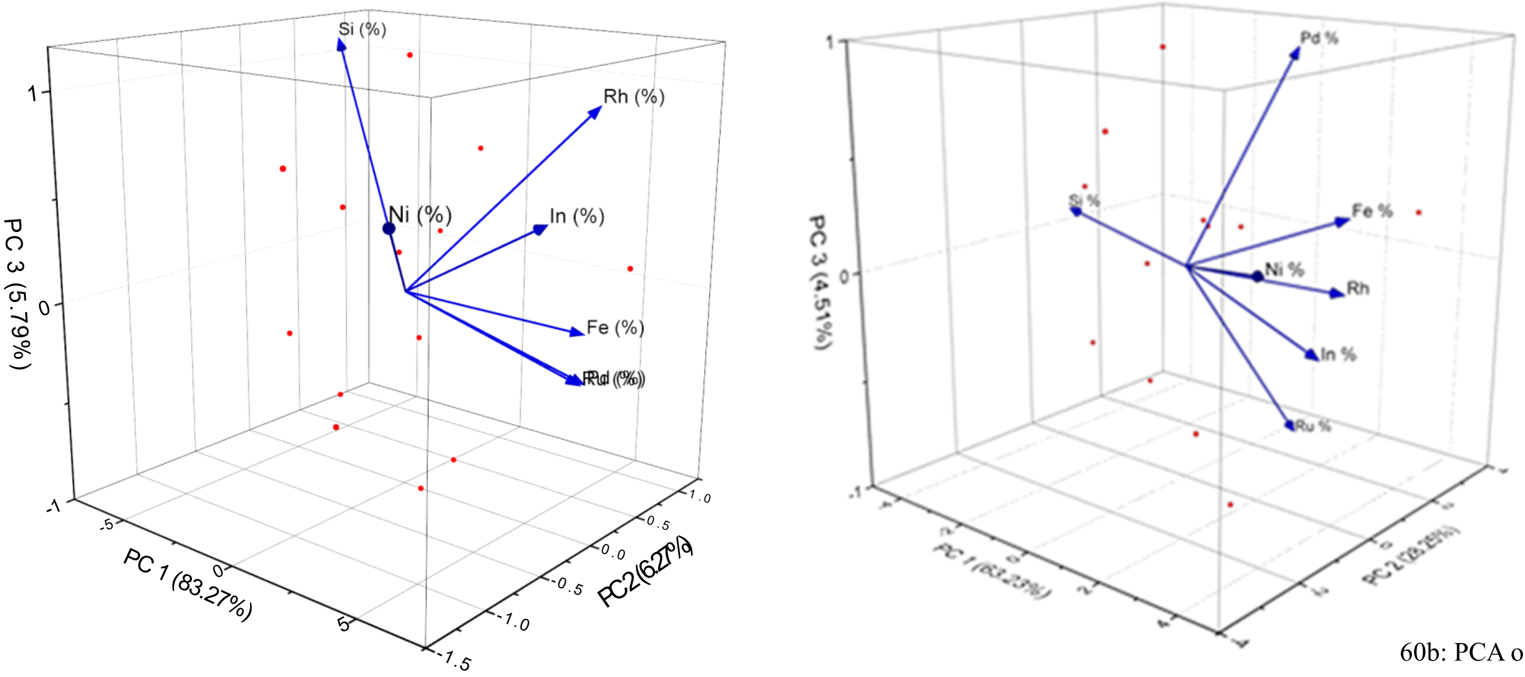
59b: PCA of ITM Rio Maior Flint

Figures 59a-b: PCA of Crescent Microliths and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig.59a: PCA of crescent microliths shows that PC1 (67.97%) is primarily determined by Si values, PC2 (19.73%) is influenced by Ru, while PC3 (6.73%) shows variations linked to In, Ni, Rh, Pd, and Fe.

Fig.59b: PCA of ITM Rio Maior flint. PC1 (63.23%) is dominated by Si, PC2 (28.25%) is given by Ru and Pd, and PC3 (4.51%) by In, Rh, Fe, and Ni values.



60a: PCA of Indeterminates Microliths

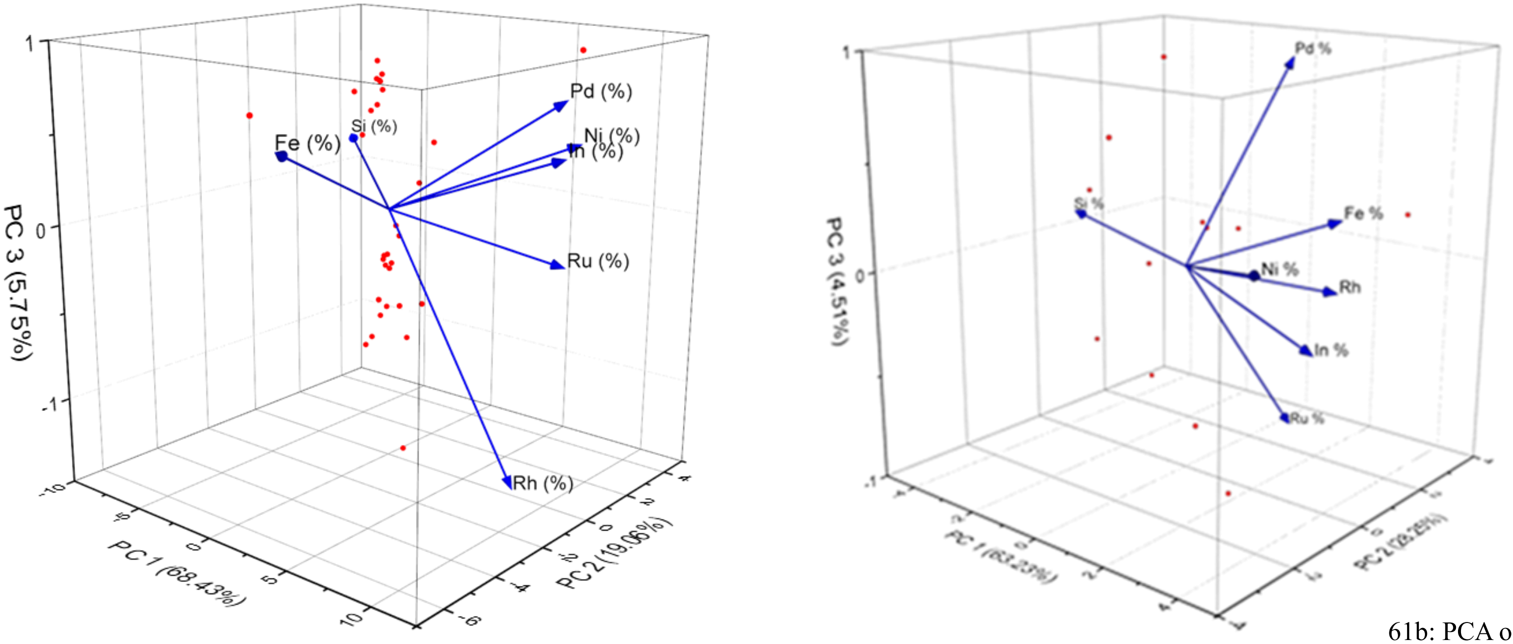
60b: PCA of ITM Rio Maior Flint

Figures 60a-b: PCA of Indeterminates Microliths and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig.60a: PCA of indeterminates microliths shows that PC1 (83.27%) is primarily determined by Si values, PC2 (6.27%) is influenced by Ru, Pd, Fe, In, Rh, while PC3 (5.79%) shows variations linked to Ni only.

Fig.60b: PCA of ITM Rio Maior flint. PC1 (63.23%) is dominated by Si, PC2 (28.25%) is given by Ru and Pd, and PC3 (4.51%) by In, Rh, Fe, and Ni values.



61b: PCA of ITM Rio Maior Flint

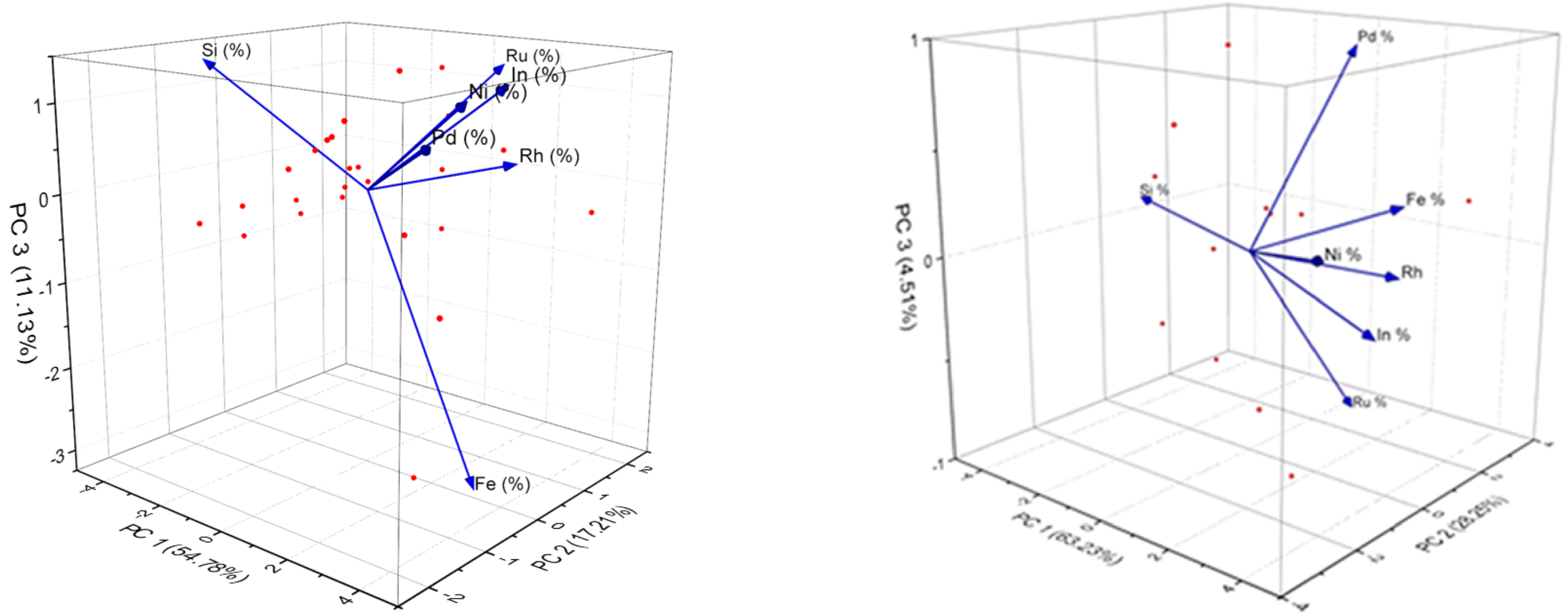
61a: PCA of Miscellaneous Microliths

Figures 61a-b: PCA of Miscellaneous Microliths and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig.61a: PCA of miscellaneous microliths shows that PC1 (63.43%) is primarily determined by Si values, PC2 (19.06%) is influenced by Ru, In, Ni, Pd, Rh, while PC3 (5.75%) shows variations linked to Fe only.

Fig.61b: PCA of ITM Rio Maior flint. PC1 (63.23%) is dominated by Si, PC2 (28.25%) is given by Ru and Pd, and PC3 (4.51%) by In, Rh, Fe, and Ni values.



62b: PCA of ITM Rio Maior Flint

62a: PCA of Debitage Residues

Figures 62a-b: PCA of Debitage Residues and ITM Rio Maior Flint

The points (red dots) represent samples, and the blue arrows indicate variable loadings, with the longer arrows denoting greater influence. Samples that appear close to each other on the plot have similar elemental composition.

Fig.62a: PCA of debitage residues shows that PC1 (54.78%) is primarily determined by Si values, PC2(17.21%) is influenced by Ru, while PC3 (11.13%) shows variations linked to In, Ni, Pd, Rh, and Fe.

Fig.62b: PCA of ITM Rio Maior flint. PC1 (63.23%) is dominated by Si, PC2 (28.25%) is given by Ru and Pd, and PC3 (4.51%) by In, Rh, Fe, and Ni values.

4.4. Comparative Analysis: Anta 1 de Vale da Laje Arrowheads vs Rio Maior Flint Samples

Sources of raw material for stone tools act as indicators of past foragers' behaviour, based on the location and transfer distance (Brantingham, 2006). This is determined through geochemical and geological methods that characterise flint microfacies and identify the geographical origins of the sources (Masson, 1981; Demars, 1985; Geneste, 1985; Tarriño, 2001; Mangado Liach, 2005; Fernandes and Raynal, 2006; Delvignes, 2016; Sánchez de la Torre et al., 2017). Studies on lithic raw material sourcing reveal shifts in resource exploitation strategies over time, highlighting forager economic and seasonal patterns (Féblot-Augustins, 1993, 1999). Reconstructing past foragers' mobility and land-use strategies involves analysing changes in travel distances, raw material quality, and quantities at sites, and documenting various lithic reduction phases that lead to artefact abandonment within the *chaîne opératoire* framework (Leroi-Gourhan, 1964; Geneste, 1985; Soressi and Geneste, 2011). This context is applied to VL1 to determine if the raw materials for arrowheads were sourced from Rio Maior flint and other local/distant sources. Figures 63-66.

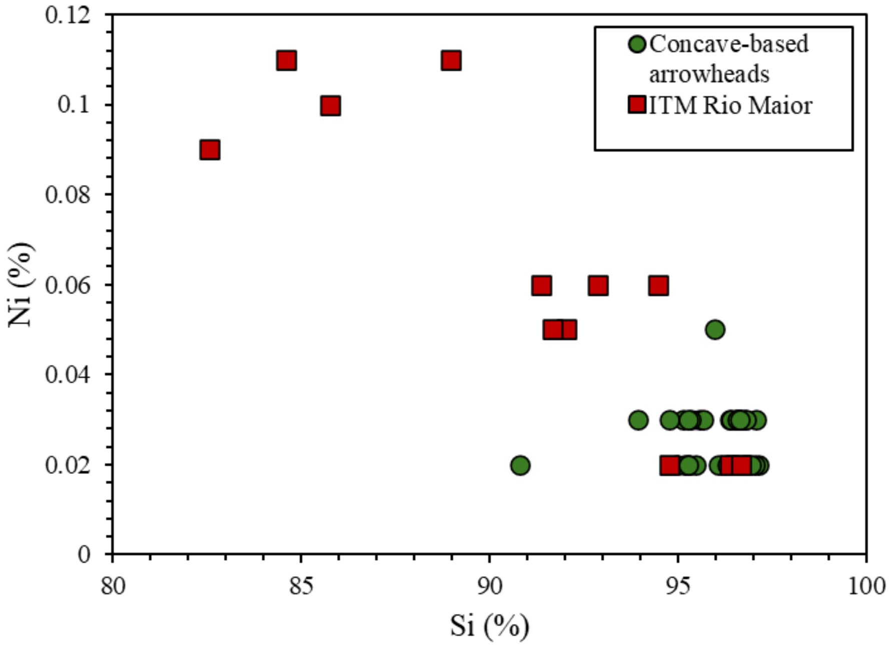


Figure 63: Plot of Si vs Ni of Concave-based Arrowheads and ITM Rio Maior Flint

The majority of the concave-based arrowheads do not cluster with the ITM Rio Maior flint, which suggests different sources. However, a rare concave-based arrowhead cluster with the ITM Rio Maior flint, which suggests the same source.

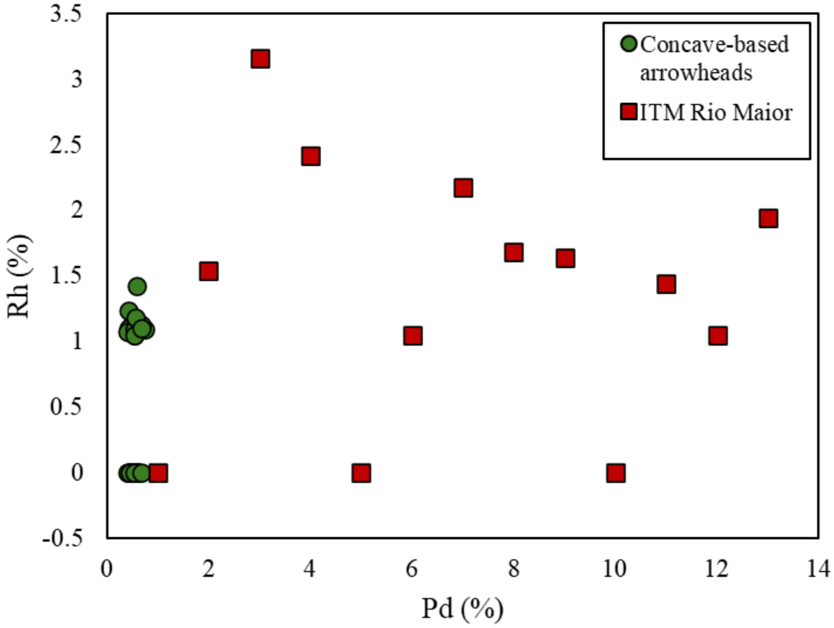


Figure 64: Plot of Pd vs Rh of Concave-based and ITM Rio Maior Flint

The majority of the concave-based arrowheads do not cluster with the ITM Rio Maior flint, which probably suggests different source(s). However, very rare of the concave-based arrowheads are associated with the ITM Rio Maior flint.

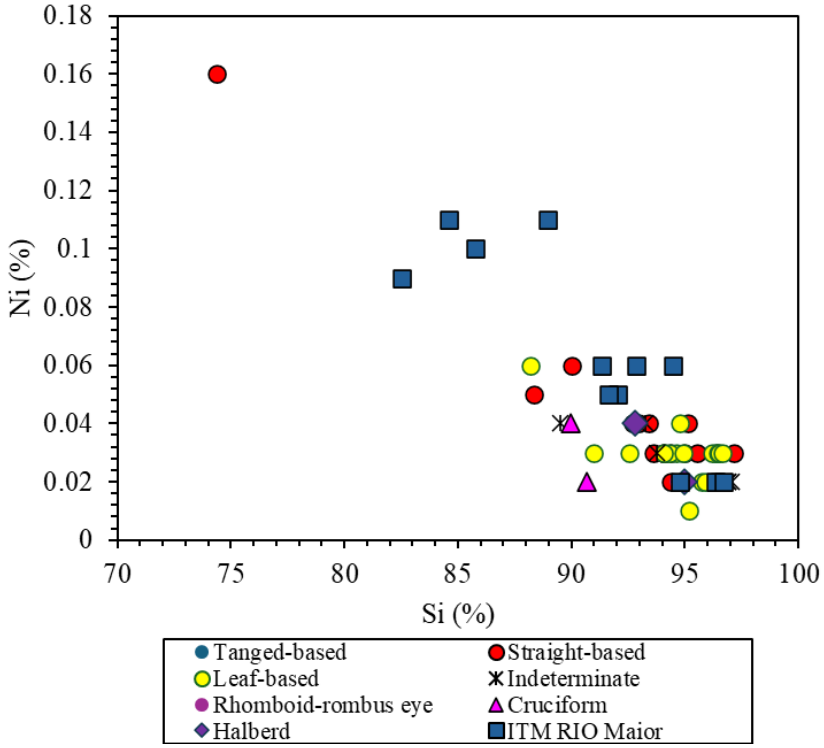


Figure 65: Plot of Si vs Ni of Other Arrowheads Typology and ITM Rio Maior Flint

Some straight-based and leaf-based arrowheads suggest distant sources, while others, such as straight-based, leaf-based, cruciform, halberd, and indeterminate arrowheads, are associated with Maior flint, which probably indicates the same sources.

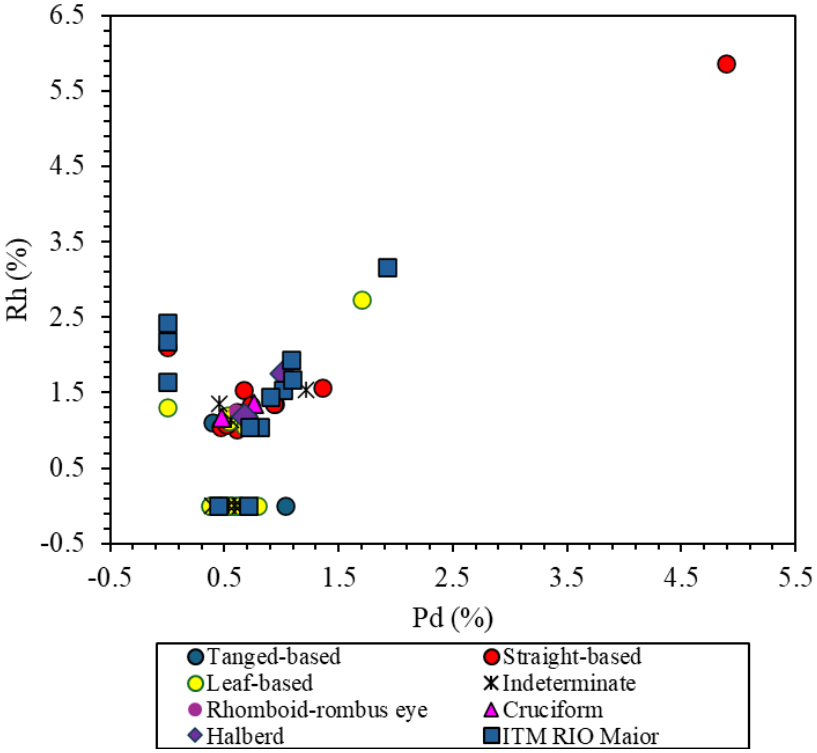


Figure 66: Plot of Pd vs Rh of Other Arrowheads Typology and ITM Rio Maior Flint

Some of the straight-based, leaf-based, tanged-based, cruciform, rhomboid, halberd, and indeterminate arrowheads are associated with ITM Rio Maior flint. Some straight-based and leaf-based arrowheads indicate distant sources.

4.5. Scanning Electron Microscope Results (SEM)

Aubry (1991) used SEM to study lithic raw material sources and understand prehistoric behaviours. Fernandes et al. (2007) applied SEM to distinguish surface changes in archaeological artefacts and their geological source rocks. They also used SEM to confirm the' evolutionary chains" characteristic of different neo-cortical families. Similarly, in this study, SEM was used to analyse debitage residues, revealing the presence of flint and silcrete in the samples. Two samples revealed silcretes.

Group 1 Flints

Flint 1

Light brown to grey opaque lacustrine flint with algae fragments (Characea), freshwater gastropods, ostracods and peloids. It is a silicified bioclastic calcarenite, bioclasts are quite well recognizable and mostly white, while peloids are dark. The silica matrix is light brown to grey, with large white patches less silicified (more calcareous).

Flint 2

Light brown vitreous and fine-crystalline flint with large white (less silicified and more calcareous) patches. It contains many thin algae fragments or tubules (Characea) and ostracods. Probably a variant of flint 1 deposited in a deeper and quieter lacustrine/lagoonal environment because of its fine texture and less taxonomic diversity; no gastropods were detected.

Flint 3

Reddish to grey, with a few white patches. Very fine-crystalline, translucent and vitreous flint. It contains thin algae tubules and fragments, probably ostracods and maybe other colonial organisms not well identified. It is probably of a marine or lacustrine depositional environment.

Group 2 Silcretes

This group is identified as red/brown to subordinate grey/green alluvial sediments (alternating mudstones, siltstones and sandstones horizons in a millimetric/centimetric scale). It is encrusted by white to pink silica mineralization. The muddy sediments are mostly red while the alluvial grains are grey and are constituted mainly by quartz, also micas, feldspars and iron oxides were detected. Secondary porosities or fissures are filled by a precipitated white to pink silica mineral (likely opal), somewhere zoned. Figures 67a-d and Figures 68a-d respectively.

Silcrete 1 - M 23 S/C 3

It is a large and thick laminar flake detached with a hard hammerstone. The dorsal part is corticated (red clay), surface shows some bidirectional lamellar negatives. On the ventral surface, there is a discontinuous lateral simple/denticulate retouch. It’s a lateral scraper. The raw material is an encrusted sediment, silicified by silica. Alternating muddy and sandy horizons are detected, among the coarser minerals, detrital quartz is the most frequent, also feldspars and clay minerals are recognizable. There is sparse Fe oxide mineralizations or concretions. The silica cement precipitated in the voids corresponds to probable opal (needs further confirmation with Raman analysis). Figures 67a-d.

Fig.67a: M 23 S/C 3 Artefact

Fig.67b: M 23 S/C 3 Microscopic Dorsal View

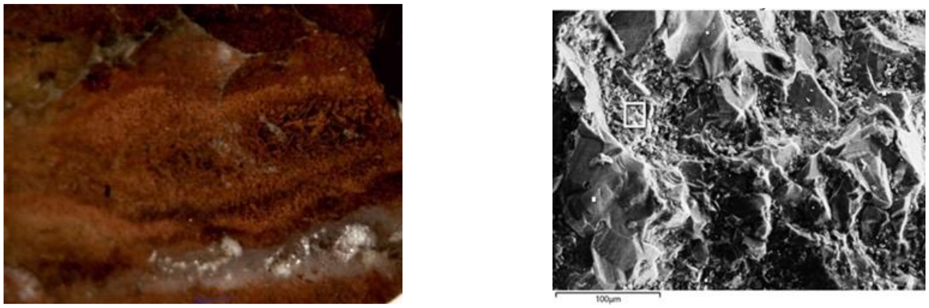


Fig.67c: M 23 S/C 3 Microscopic Ventral View

Fig.67d: M 23 S/C 3 SEM Image

Figures 67a-d: Debitage Residue M 23 S/C 3

Fig.67a is the debitage residue of the M 23 S/C 3 artefact. Fig.67b is the dorsal view of M 23 S/C 3. Fig.67c is the ventral view of M 23 S/C 3. Fig.67d is the SEM image displaying the microstructural features of the fibrous pattern.

Silcrete 2 – REC SUP SC 2

It is a cortical flake (green clay), it is similar to sample M23 SC3 as an encrusted sediment, silicified by silica. Alternating muddy and sandy horizons are detected, with the coarser minerals being the most frequent. Among these, detrital quartz is the most prevalent, followed by feldspars and clay minerals. However, the sample is less silicified. The green-grey horizon is constituted mostly by detrital quartz (thin sandstone), whilst the brown horizon is mostly encrusted mud.   Figures 68a-d.

Fig.68a: REC SUP SC 2 Artefact

Fig.68b: REC SUP SC 2 Microscopic Dorsal

Fig.68c: REC SUP SC 2 Microscopic Ventral

Fig.68d: REC SUP SC 2 SEM Image

Figures 68a-d: Debitage Residue REC SUP SC 2

Fig.68a is the debitage residue of the REC SUP SC 2 artefact. Fig.68b is the dorsal view of REC SUP SC 2. Fig.68c is the ventral view of the REC SUP SC 2 artefact. Fig.68d is the SEM displaying the microstructural features of the moderately fibrous pattern.

4.6. Spatial and Statistical Patterns

Spatial analysis is a vital tool that allows the viewing of regions from broad landscapes to specific sites (McCoy and Ladefoged, 2009; Gillings, 2012; Rennell, 2012). Similarly, lithic tools (arrowheads and geometric microliths) recovered from the VL1 site are employed to offer insights about prehistoric people's behaviour. According to Olszewski (2001), "tool types are often assigned meaning, that is, they are used as the basis for interpretations of technology, activity, mobility, group identity, spatial and chronological distribution." This is applied to the VL1 site, where lithics were distributed across stratigraphic levels, revealing significant variations based on the assemblage of arrowhead typology and geometric microliths analyzed. The arrowheads assemblage identified in VL1 is eight (8), comprising concave-based, straight-based, leaf-based, tanged-based, rhomboid, cruciform, halberd, and indeterminates. The geometric microliths include triangles, trapezoids, crescents, miscellaneous, and indeterminates. Tables 4 and 5 (Figures 69 and 70) respectively.

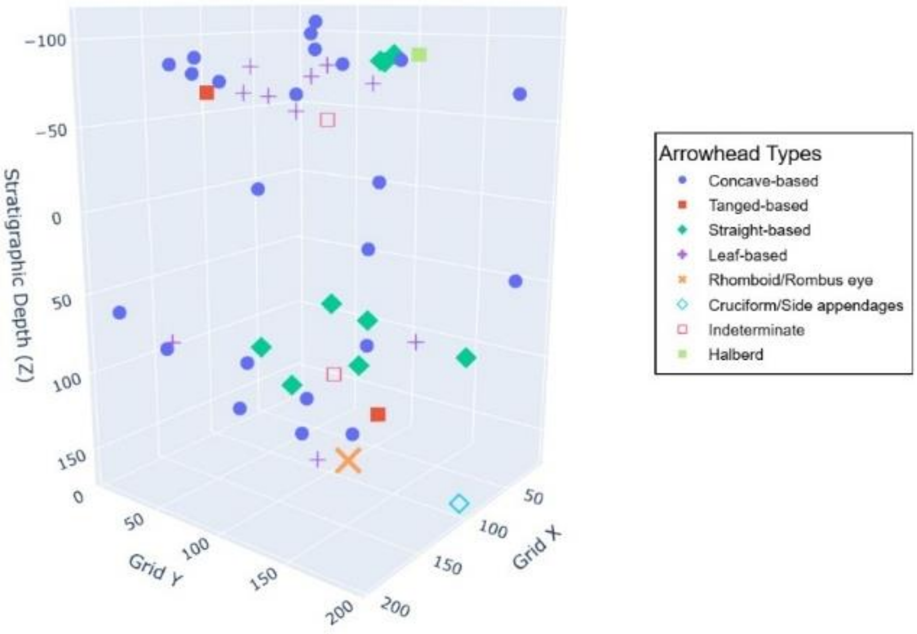


Figure 69: 3D Spatial Distribution of Arrowheads Typology by Excavation Grid Coordinates and Stratigraphic Depths

The diagram illustrates the spatial distribution and stratigraphic positions of various arrowhead types within an archaeological excavation grid. The three axes represent horizontal spatial coordinates (Grids X and Y) and vertical stratigraphic depth (Z), allowing for the analysis of arrowhead typology distribution and their chronological changes.

Table 4: Arrowheads Assemblage

Arrowheads Typology	**Frequency**
Concave-based	23
Straight-based	9
Leaf-based	10
Tanged-based	2
Cruciform/Side appendages	1
Rhomboid/Rombus eye	1
Halberd	1
Indeterminates	2
TOTAL	**49**

The table shows that the arrowhead assemblage contains: concave-based typology (23), straight-based typology (9), leaf-based typology (10), tanged-based typology (2), rhomboid/rombus eye (1), cruciform/side appendages (1), halberd (1), and indeterminates (2).

4.6.1 Arrowheads Typology

In this section, the arrowhead typology is analysed based on appearance across the stratigraphic layers.

Concave-based Arrowheads

It occurs between levels -100 and -50, associated with tanged-based, leaf-based, and straight-based arrowheads, spreading across the stratigraphic layer. It is absent from -50 to 0, then appears alone at 0 to 50, spreading across that layer. At 50 to 100, it occurs with leaf-based arrowheads or alone. From 100 to 150, it appears alone and evenly distributed across the sterile layer.

Straight-based Arrowheads

It occurs at level (-100 to -50) in association with itself and co-exists with a concave-based arrowhead. It is absent at levels (-50 to 50). It occurs alone at the level (50 to 100) and level (100 to 150) and spreads across the stratigraphic layer. It also exists alone on the sterile layer.

Leaf-based Arrowheads

It occurs at level (-100 to -50) in association with a concave-based arrowhead, spread across the stratigraphic layer. It occurs along the boundary line (-50) in association with a concave-based arrowhead. It is absent at the level (0-50). It occurs along the boundary line at level (-100) in association with a concave-based arrowhead and exists alone at level (100 to 150). It exists alone on the sterile layer.

Tanged-based Arrowheads

It occurs at level (-100 to -50) in association with a concave-based arrowhead. It is absent at levels (-50 to 150). It exists alone on the sterile layer.

Rhomboid/Rombus eye Arrowheads

It is absent from all levels (-100 to 150). It exists alone on the sterile layer.

Cruciform/Side appendages Arrowheads

It is absent from all levels (-100 to 150). It exists alone on the sterile layer.

Indeterminates Arrowheads

It exists alone at level (-50 to 0). It is absent at level (-100 to -50). It is absent at levels (-50 to 100). It occurs along the boundary line at level (150).

Halberd

It occurs only at level (-100 to -50). It is absent from levels (-50 to 150) and the sterile.

In summary, at the horizontal level, concave-based arrowheads are widely distributed across the stratigraphic layers/levels. In contrast, leaf-based arrowheads are more clustered together towards higher X and Y coordinates. This probably suggests a spatial distribution in technology, tool usage, group mobility, and/or symbolic practices.

Stratigraphically, tanged-based and straight-based arrowheads occur both near the surface and at deeper levels, which probably suggests longer temporary usage. Rhomboid, cruciform, and indeterminate only occur at deeper levels, while halberd occurs only at the surface level. This probably indicates a mobile group that may have passed through the site on seasonal mobility once/twice, and/or through interaction with other local or distant neighbouring sites.

Based on Technology

This is according to Olszewski (2001), "tool types are often assigned meaning, that is, they are used as the basis for interpretations of technology…" The arrowheads are interpreted based on each typology as representing a new technology along each stratigraphic layer, as detailed below:

Level (-100 to -50): There exist five levels of technology, as indicated by the arrowheads typology of concave-based, tanged-based, leaf-based, straight-based, and halberd.

Level (-50 to 0): There exist two levels of technology as indicated by the arrowheads typology of leaf-based and indeterminate.

Level (0 to 50): There exist only one level of technology as indicated by concave-based arrowheads.

Level (50 to 100): There exist three levels of technology as indicated by the arrowheads typology of leaf-based, concave-based, and straight-based.

Level (100 to 150): There exist three levels of technology as indicated by the arrowheads typology of leaf-based, concave-based, and straight-based.

On the sterile layer: There exist seven levels of technology as indicated by the arrowheads typology of concave-based, straight-based, tanged-based, leaf-based, cruciform/side appendages, rhomboid/rombus eye, and indeterminates.

In summary, the clustering of specific arrowhead types across different stratigraphic layers supports the idea of multiple occupation phases, each with evolving tool traditions. Alternatively, these distribution patterns might indicate specialized activity zones dedicated to hunting, craftwork, or symbolic practices such as burial.

4.6.2 Geometric Microliths

Tools reveal a complex set of social behaviours, and their study reveals the social dynamics of past cultures (Semenov, 1964; Keely, 1980; Mansur-Franchomme, 1983; Álvarez, 2003). In this section, the geometric microliths are analysed based on how they appear across the stratigraphic layers. These include trapezoids, triangles, crescents, miscellaneous, and indeterminates, Table 5.

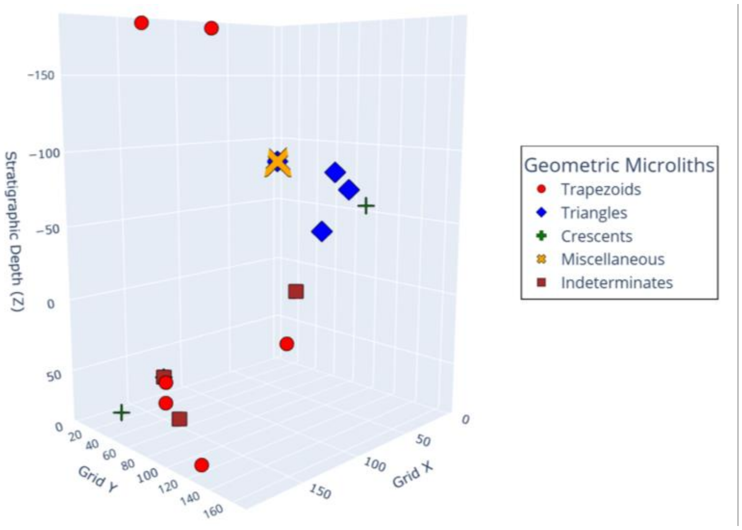


Figure 70: 3D Spatial Distribution of Arrowheads Typology by Excavation Grid Coordinates and Stratigraphic Depths

The diagram illustrates the spatial distribution and stratigraphic positions of various geometric microliths within an archaeological excavation grid. The three axes represent horizontal spatial coordinates (Grids X and Y) and vertical stratigraphic depth (Z), allowing for the analysis of geometric microliths distribution and their chronological changes.

Table 5: Geometric Microliths Assemblage

Geometric Microliths	Frequency
Triangles	4
Trapezoids	6
Crescents	2
Miscellaneous	1
Indeterminates	3
TOTAL	**16**

This table shows that the geometric microliths are triangles (4), trapezoids (6), crescents (2), miscellaneous (1), and indeterminates (3).

Triangles Microliths

It is absent at levels (-150 to -100). It occurs at level (-100 to -50) associated with miscellaneous. It occurs alone at level (-50 to 0). It is absent at levels (0-50) and on the sterile layer.

Trapezoids Microliths

It occurs at the level (-150 to superficial level). It is absent at levels (-150 to 50). It occurs on the sterile layer associated with indeterminates. It exists alone on the sterile layer.

Crescents Microliths

It is absent at levels (-150 to -100. It exists alone at level (-100 to -50). It is absent at levels (-50 to 50). It exists alone on the sterile layer.

Miscellaneous Microliths

It is absent at levels (-150 to -100). It occurs alone at level (-100 to -50) associated with triangle. But absent at level (-50 to 50) and on the sterile layer.

Indeterminates Microliths

It is absent at levels (-150 to 0), it exists alone at level (0-50). It exists on the sterile layer associated with crescent and trapezoid. It exists alone on the sterile layer.

Phases of Occupation

At level (-150 to superficial layer), there exists one phase of occupation based on the presence of a trapezoid.

At level (-150 to 100), no tool is present.

At level (-100 to -50), there exist three phases of occupation based on the presence of triangle, miscellaneous and crescent.

At level (-50 to 0), there exists one phase of occupation based on the presence of a triangle.

At level (0 to 50), there exists one phase of occupation based on the presence of indeterminates.

On the sterile layer, there exist three phases of occupation based on the presence of trapezoid, crescent, and indeterminate.

In summary, the clustering of specific geometric microliths across different stratigraphic layers supports the idea of multiple subsistence economies, each with evolving tool traditions. Alternatively, these distribution patterns may indicate adaptive behaviour to climate change, whereby small animals were being hunted and other farming practices.

Although this spatial distribution analysis was done based on the available coordinate data.

This chapter presents the techno-typological, SEM, and analytical findings on the elemental composition of PCA for the VL 1 artefacts and Rio Maior flint and spatial analysis. Techno-typology identified different arrowhead types and geometric microliths, while SEM confirmed silcrete as the raw material. The clustering plots (PCA) illustrate the degree of association and spatial distribution of data that shed light on the technology, activities, and mobility at VL1. These techniques establish a framework for geochemical fingerprinting of lithic tool sources, which is explored in the next chapter.

CHAPTER FIVE

5.0 DISCUSSION

5.1 Overview

This chapter provides a comprehensive overview of the lithic analysis conducted at the Anta 1 de Vale da Laje (VL1) funerary site, combining typological, geochemical, petrographic (SEM) and spatial analyses as previously stated. Typologically, it reveals the dominance of unbroken arrowhead points. The geochemical results highlight the findings of Platinum Group Elements (PGEs) - specifically ruthenium (Ru), rhodium (Rh), and palladium (Pd) - found in various lithic artefacts, including arrowheads, microliths, debitage residues, and ITM Rio Maior flint raw material samples. These trace elements were detected using portable X-Ray Fluorescence (pXRF), and the presence of flint and silcrete in some debitage residues was confirmed through Scanning Electron Microscope (SEM) analysis. The analysis employs Principal Component Analysis (PCA), and Spatial Analysis to interpret similarities and differences in composition among artefact types and the known potential raw material source- ITM Rio Maior flint. The Spatial Analysis offers insights into both short-term and long-term site usage, as dominated by the distribution of concave-based arrowheads and various geometric microliths, which suggests different subsistence economies practised across the stratigraphical layers. Overall, these findings deepen our understanding of the geochemical signatures of various raw material sources exploited during the Neolithic and their significance in studying prehistoric resource sourcing, exchange networks, and social organization.

5.2 The Range of Resources

The emergence of cohesive communities with strong family or group ties characterizes the Neolithic period. Funerary rituals and practices played a crucial role in these complex, socially structured societies (Gibson, 2016). These practices often included various cultural materials at burial sites, offering insight into social hierarchies and regional interactions (Geber et al., 2017). Social mechanisms are involved in sourcing raw materials for artefacts (Hughes, 2011). Different procurement strategies have been used by scholars, which include, Guilbeau et al. (2019) classified the range of resources based on nearness to the source as ‘local’ raw materials sources within a range of 1-2 hours walking distance to the site; two days' journey to the site as ‘regional’, while more than a few days to the site is ‘exogenous’ coupled with other variety of raw materials. Moreover, Dolan et al. (2019) identified three acquisition modes: first, through direct collection from primary sites or secondary geological outcrops/ along river gravels of a few kilometres from the source (Binford, 1979; Gould and Saggers, 1985; Frahm, 2012); secondly, via the embedded method, where lithic materials are gathered incidentally while performing other activities such as food gathering or visiting allies (Binford, 1979); and third, through trade and exchange with other groups, leading to raw materials or finished tools being obtained indirectly (Hughes, 2011). These models illustrate how lithic procurement was both an economic and a social activity during the Neolithic period.

In this study, techno-typological analysis revealed that the assemblage was dominated by flint arrowheads, especially concave-based (42%) and straight-based (23%) arrowheads (Figure 25). This corresponds to the findings of Cruz et al. (2013) at Gruta do Morgado Superior, Tomar, Portugal, where flint arrowheads dominated the lithic assemblage, dated from the cardial Early Neolithic to the Late Bronze Age based on stratigraphical analysis. Also, Cruz and Berruti (2015) documented that the arrowheads (38) from Gruta do Morgado Superior had impact fractures of 47%, which indicated prior usage of the arrowheads. Similarly, the result of Andrade (2015), from the megalithic communities in Ribeira da Seda (North Alentejo, Portugal), dated Neolithic to Chalcolithic, based on petrographic analysis, revealed a wide variety of flint arrowheads sourced from a wide range; these arrowheads are parallel to those identified in this study. Moreover, the abundance of concave-based arrowheads in VL1 aligns with Caninas et al. (2011), who documented the prevalence of concave-based arrowheads in the Tumulus at Charneca das Vinhas, Rodão, Portugal, during the Chalcolithic period. Similarly, Jordão (2017) discovered that both concave- and straight-based arrowheads were common at the Zambujal settlement during the Chalcolithic period. However, VL1 Dolmen spans from the Middle Neolithic to the Bronze Age, while Charneca das Vinhas and Zambujal are dated Chalcolithic period. Also, Forenbaher (1999) stated that arrowheads were very common within the flint-bearing zone of less than 30 km and outside the flint-bearing zone in Portugal. These results imply that VL1 functioned as a funerary site where arrowheads were deposited as burial offerings and possibly served as short-term camps for hunting and habitation from the Middle Neolithic to the Bronze Age, in which they could have sourced the raw material from a wide range of resources of flint outcrops, both from local and distant areas. This is further confirmed by Cruz et al. (2014) that VL1 served as a funerary space, noted that the high mobility was linked to gathering and hunting activities. This is evident in this study (Figures 52-56) - the wider scatter of samples across PCA space indicates that arrowheads were made from multiple flint sources with different geochemical signatures. This diversity suggests that communities were not reliant on a single quarry but instead had access to a broad range of lithic resources. This variability can be interpreted as evidence of high mobility, as prehistoric peoples moved across landscapes to different flint outcrops or through exchange networks. Similarly, Sousa (2011) added that the silicified schist raw material used for these arrowheads was not from Extremadura, indicating a trans-regional exchange network among Neolithic communities. Furthermore, Cruz et al. (2013) noted that arrowheads were chronological markers that indicated phases of occupation in the Alto Ribatejo region and the Iberian Peninsula.

In the same vein, the techno-typological result revealed the presence of geometric microliths, dominated by triangular microliths (27%) and trapezoids (21%) (Figure 40). This aligns with the findings of Nukushina (2015), who accounted for the presence of geometric microliths from the Sado shell middens of Amoreiras (Alcácer do Sal, Portugal) obtained from a nearby source, but with a lower number of triangles (19.72%). Similarly, it corresponds to the findings of Cruz et al. (2013), who documented the presence of geometric microliths (number not mentioned) from the Gruta do Morgado Superior. Also, Andrade (2015) documented the presence of flint geometric armatures (number not mentioned) from the megalithic communities of Ribeira da Seda obtained from a wide range of sources. In this study, a wide scatter of PCA is observed in (Figures 57-61), which probably revealed that a wide range of flint sources were used to produce geometric microliths. The presence of diverse geometric microliths at VL1 could be linked to the site's function during the transition from the Mesolithic hunter-gatherer to the Neolithic agricultural practices. This is because geometric microliths were markers of cultural transformations between groups of Mesolithic hunter-gatherers and/or Neolithic farmers (Cortell-Nicolau et al., 2025). In central and southwestern Portugal, geometric microliths spanned from the Upper Paleolithic to the Late Neolithic with their dominance in tools mostly evident during the Late Mesolithic (Araújo 1995/1997; Vierra, 2004). VL1 is further supported to have functioned as an aggregation site (Cortell-Nicolau et al., 2023) and maintained through social networks (Barrera-Cruz et al., 2024). The diverse techno-typological assemblage of arrowheads and geometric microliths at the VL1 site suggests that the prehistoric peoples continued their ancestral practices, Akkermans (2004).

The wider scatter of PCA of the debitage residues in (Figure 62) indicated a wide range of flint sources. This could be attributed to erosional activities, as findings correspond to Pereira and Carvalho (2017) that geochemical variations existed in flint nodules, primary and secondary sources.

Therefore, the probable sources of VL1 could be linked to various mechanisms such as down-the-line exchange (Renfrew, 1975), direct and embedded procurement (Binford, 1979), territoriality (Bradley and Edmonds, 1993), symbolic exchange (Edmonds, 1995), and long-distance raw material trade (Hughes, 2011). Collectively, these models suggest that Neolithic communities utilized a wide range of resources through different forms of interaction and procurement strategies.

5.3 Implications for Exchange and Mobility

Residentially, agricultural societies are stable and harness the resources within and beyond their landscapes. They are characterised by informal lithic technology and acquire lithic materials using direct, embedded, and indirect methods (Binford, 1979; Parry and Kelley, 1987; Andrefsky, 1994). Several factors influence the technological organization of mobile and sedentary groups and hence cannot be classified as formal versus informal chipped stone technology (Tomka, 2001; Doelman, 2005). This is reflected in VL1.

5.3.1 Direct Procurement vs Exchange

In nature, there is a limited and uneven distribution of natural resources across the landscape; hence, prehistoric people were mobile in search of raw materials (Aubry et al., 2012). In this study, it was observed that the source of materials was direct procurement based on the pXRF results obtained, in which the highest and lowest values of PGEs (Ru, Rh, and Pd) were observed in VL1 artefacts and the ITM Rio Maior flint samples. The highest weight of Ru (6.34%) was observed in straight-based arrowhead 28 H S/C 11 (Figure 43), and the lowest weight of Ru (0.78%) was in ITM Rio Maior flint sample 8 (Figure 44). The highest weight of Rh (5.86%) was observed in a straight-based arrowhead 28 H S/C 11 (Figure 43), and the lowest weight of Rh (1.01%) was in straight-based arrowhead 29 H 24 (Figure 45). Also, the highest weight of Pd (4.89%) was observed in a straight-based arrowhead 29 H 24 (Figure

45), and the lowest weight (0.38%) was observed in leaf-based arrowheads 27 G S/C 74 and 28 F 11 (Figures 46 and 47), respectively. This aligns with Monteiro et al. (2018), who reported PGEs in the industrialized area of the Tagus estuary and noted higher concentrations in deeper sediments; hence, this suggests that the flint raw material used to produce the lithic tools is probably within the Tagus Basin. Even though PGEs are rare in flint, they are documented in ophiolites and chloride-rich environments (Saad, 2024). Trace concentrations of PGEs are associated with flint or PGE-bearing geological settings, such as black shales or volcaniclastic sediments (Frontiers in Earth Science, 2021). Additionally, PGE (Pt, Rh, and Os) have been detected in localised areas of the estuary and coastal sediments due to anthropogenic activities (Cobelo-García et al., 2011; Almécija et al., 2015, 2016a, b). Moreover, Berezhnaya and Dubinin (2023) reported that PGEs transformation occurs in the river-sea mixing zone. They also identified light PGEs as Ru, Rh, and Pd based on atomic weight. However, Pereira and Carvalho (2017) documented the presence (lower values) of Rh and Pd in the 19 archaeological specimens analysed from the Cerradinho do Ginete campsite using portable X-ray fluorescence (pXRF). Notwithstanding, the identification of PGEs from VL1 needs additional investigation because, during the Neolithic period, flint was heated to create tools (Karsten, 1994; Malmer, 2003), which may have affected the artefacts' composition as observed in debitage residues (Table 2). This aligns with Borradaile et al. (1998), who discussed heat modification impacting the magnetic susceptibility of cherts. Other processes that could alter artefact composition include chemical weathering processes—such as pore fluid exchange (Bush and Sieveking, 1986) and carbonate leaching from cherts (Hurst and Kelly, 1961) could also alter geochemical signals critical for sourcing cherts archaeologically. According to Howard's hypothesis (1999), weathering processes like solution and leaching significantly influence mineralogy, with chemical dissolution being the main mechanism behind river patina formation on cherts. Leaching was reported by Adewumi (2019; 2025) on the VL1 site, which could have affected the artefacts' composition. Pereira and Carvalho (2017) further noted that there exist significant geochemical variations within individual flint nodules, and that possibly greater variations from primary and secondary flint sources due to erosional activities based on pXRF.

Furthermore, based on elemental composition analysis, few of the geometric microliths were probably sourced from the ITM Rio Maior flint source, as displayed in (Figure 48) plot of (Si vs Ni), where some triangles, trapezoids, crescents, indeterminates, and miscellaneous are associated with the ITM Rio Maior source; similarly in (Figure 49) plot of (Pd vs Rh), some of the triangles, trapezoids, crescents, indeterminates, and miscellaneous are associated with the ITM Rio Maior source. But the plot of Si versus Ni for the debitage residues and ITM Rio Maior flint (Figure 50) only a very few of the debitage residues cluster with the ITM Rio Maior flint source, which suggests the same source. Also, the plot of (Pd vs Rh) of debitage residues and ITM Rio Maior flint, a few of both samples cluster together, while some of the ITM Rio Maior flint indicate distant sources.

In Figure 63, the plot of (Si vs Ni) of the concave-based arrowheads and ITM Rio Maior flint, rare samples of the concave-based arrowheads cluster with the ITM Rio Maior flint, which suggests the same source. Also, in (Figure 64), the plot of (Pd vs Rh) majority of the concave-based arrowheads and ITM Rio Maior flint did not cluster together, suggesting different sources entirely, while very rare concave-based arrowheads are associated with the ITM Rio Maior flint. The clustering togetherness of some and a few artefacts, as indicated above, implies that prehistoric people could have obtained the samples from the Rio Maior flint source through embeddedness.

Moreover, in terms of direct procurement, the petrographic and SEM analyses of debitage residues revealed silcrete in M 23 S/C 3 (Figures 67a-d) and REC SUP SC 2 (Figures 68a-d). This aligns with Aubry et al. (2022), who noted that Upper Paleolithic lithic assemblages from Côa Valley sites were obtained from flint and silcrete originating in the Lusitanian, Tagus, and Douro Basins in the Northern and Southern Meseta, about 100 to 250 km away (Mangado Llach, 2005; Aubry et al., 2012, 2016). Similarly, Andrade (2015) stated the use of flint and silcrete to produce bifacial points at the megalithic communities of the Ribeira da Seda, sourced from the Estremadura Limestone Massif (Cenomanian flint). However, Pereira and Carvalho (2017) indicated that most of the archaeological specimens from Cerradinho do Ginete were linked with geological samples from the southern part of the Rio Maior basin, while rare were linked with geological samples from the middle Tagus River Basin based on pXRF analysis.

Additionally, silcrete procurement and usage have been linked to specific techno-complexes, increased mobility, and technological innovations such as heat treatment, backing, bifacial, and pressure flaking, along with symbolic behaviours (Singer and Wymer, 1982; Ambrose and Lorenz, 1990; Wurz, 1999; Henshilwood et al., 2001; McCall, 2007; Mourre et al., 2010;

Lombard et al., 2012; Brown et al., 2012; Schmidt et al., 2013; Will and Mackay, 2017). Hence, the search for this raw material by the prehistoric people within the local and other sources could be linked to its characteristics. As noted by Binford (1980) that opportunistic short-distance resource gathering occurs especially during seasonal mobility periods (Binford, 1980). Similarly, Oosterbeek (2025) stated that Neolithic agropastoralists exhibited diverse mobility patterns. Hence, it could be suggested that other local sources within the plains of the Tagus Basin were utilized; this is consistent with the procurement strategy embedded in daily subsistence activities (Binford 1979). This is supported by Marks et al. (1991) that in Portugal, mobility and the procurement of raw materials took place within the siliciclastic formations of Upper Cenomanian flints in the sedimentary Basin of the River Tagus (TSB). Silcrete formation in the Tagus Basin could be due to several formation processes, some of which include the geological structural faults that enabled the weathering of parent materials, thereby leading to diverse soil formation (Biondino et al., 2018). Others include hydrothermal and weathering activities, thereby leading to the alteration of rock mineralogy (McClay, 1990; Minguely et al., 2010), and the diagenesis and lithification of the weathered bedrock of the Estremenho Limestone Massif (Adewumi, 2025), among others.

Largely, this suggests that both direct procurement and exchange sources provided resources to the VL1 site. This is supported by Moleiro (2015) that the materials used in the construction of the VL1 Dolmen were sourced just 100 m from the monument based on archaeopetrographic analysis. Thorpe and Williams-Thorpe (1991) stated that the sourcing of raw materials for short distances typically ranges from 1 to 2 km in European regions. Moreover, Jorge (2014) reported that the lithic tools recovered from settlements and burial sites on the Mondego Plateau in Central-Northern Portugal during the Late Neolithic were sourced within a range of 1 to 15 km, based on landscape-scale and comparative artefact studies. Pereira et al. (2015) observed that the lithic assemblage from the Epipaleolithic Pena d'Água Rockshelter was sourced approximately 14 km away from the secondary sources of the chert outcrop, as indicated by pXRF, VP-SEM-EDS, and m-XRD analyses. Additionally, flint could have been obtained either directly during seasonal mobility or through exchange, as indicated by Stojanovski et al. (2020), who opined that pastoralist groups moved seasonally to the VL1 monument during the Neolithic, given by evidence of milk residues in pottery. Oosterbeek (2025) noted that prehistorical agropastoralists in Portugal exhibited diverse mobility patterns during the exploitation of marine and terrestrial resources along the

Tagus tributaries and Alentejo plain. Furthermore, through exchange (Gambe, 1999), due to social networks (Whallon, 2006), could also have facilitated resource transfer.

5.3.2 Neolithic Mobility

Mobility constitutes a fundamental characteristic of both ancient hunter–gatherer societies and contemporary communities, due to the limited and uneven distribution of natural resources across the landscape (Aubry et al., 2012). Binford (1982) proposed that hunter-gatherers sourced their raw materials along river sediments about ten km from their habitation sites during daily foraging. This is further emphasized by Andrade and Matias (2011) that during the Neolithic period in the Lisbon Peninsula, occasional exploitation sites were used by the seasonal groups for flint procurement strategies and lithic artefact production during mobility. Moreover, Gopher (1994) noted that arrowheads are chronological markers due to their different morphologies that have changed over different periods, especially during the Neolithic period. Torrence (1989) accounted that lithic technology and procurement methods are dependent on the degree of mobility and sedentism. Highly mobile groups are characterised by formal lithic technology that requires high energy for sourcing raw materials to produce quality tools. Mobile hunter-gatherers and early farmers used a direct or embedded procurement strategy in the replacement of their tools with local materials during their mobility range in upland and lowland environments (Shackley, 1990; Vierra, 1990; Roth, 2000). This is observed at the VL1 site based on the assemblage of the techno-typological of arrowheads and geometric microliths recovered. This suggests that during the foraging, mobile groups probably obtained lithics directly from ITM Rio Maior sources through embeddedness to carry out their activities at VL1, as reflected in (Figure 65) plot of (Si vs Ni), in which some straight-based, leaf-based, halberd, cruciform, rhombus, tanged-based, and indeterminates arrowheads are associated with the ITM Rio Maior flint. Similarly, in (Figure 66) plot of (Pd vs Rh), some straight-based, leaf-based, tanged-based, rhomboid, cruciform, halberd, and indeterminate arrowheads are associated with the ITM Rio Maior flint. In the same vein, but in contrast, in (Figure 65) plot of (Si vs Ni), rare straight-based arrowheads show distant sources. Similarly, in (Figure 66) (plot of Pd vs Rh), rare straight-based arrowheads indicate distant sources, as noted by (Sousa, 2011). Thus, it confirms the high mobility of the agropastoralist groups. Since VL1 served as a funerary site, prehistoric peoples could have brought the arrowhead from distant source(s) as offerings for the dead. This aligns with Forenbaher (1999) that arrowheads were obtained far beyond the flint-bearing zone for burials. This implies that prehistoric people were engaged in high mobility patterns and exploited a wide range of resources during the Neolithic period.

In summary, the prevailing local and regional catchment of raw materials, which characterized the hunter-gatherer societies in Western Iberia, this feature is kept dominant in the Neolithic megalithic context of VL1, despite the cultural differences and the expected increase of distance interaction.

5.4 Social and Symbolic Implications

Forenbaher (1999) observed that arrowheads were very common at burial sites, often found in large quantities during the Late Neolithic to Chalcolithic period. He identified two types of Portuguese arrowheads (concave and convex) that are chronologically significant. He emphasised that arrowheads found in burials are usually complete, while those from settlements tend to be broken. Based on the assemblage of arrowheads recovered from VL1, it can be concluded that the site primarily served both a short-term subsistence economy and long-term social and symbolic practices, such as funerary rites. This pattern is seen in this study, where most arrowheads have points. As shown in (Figure 26), 69 out of 88 arrowheads have points, while 19 do not. This aligns with Forenbaher (1999), who suggests that more complete and unused arrowheads were found at burial sites, often offered as burial goods, while settlement deposits contained fewer incomplete and used arrowheads. Similarly, this confirms the report of (Palomo and Gibaja, 2002; Gibaja and Palomo, 2003) that complete arrowheads accompanied the burials of warriors' bodies about the Costa de Can Martorell in Spain. This pattern is similar to Zambujal settlement arrowheads, where 39% of the tips or wings were broken (Jordão, 2017). This further supports the idea that VL1 arrowheads were likely offered as burial goods to indicate social status and symbolic values, as discussed by Torrence (1986) and Pétrequin et al. (2012). Additionally, evidence from Oosterbeek et al. (1992) and Cruz et al. (2014) suggests that VL1 functioned more as a funerary space for the community. This is supported by data in this study (Table 4, Figure 69), where concave-based arrowheads are evenly distributed across stratigraphic layers, followed by leaf-based and straight-based arrowheads also spread throughout some layers. Clark et al. (1974) noted that projectile design reflects the creativity of their makers to serve various purposes, indicating these arrowheads were likely used for burial and other symbolic practices at VL1. Moreover, according to Table 5 and Figure 70, the geometric microliths reinforce VL1's role as a short-term camp for agropastoralists, consistent with a subsistence economy (Stojavnoski et al., 2020). Cortell-Nicolau et al. (2025) have highlighted that shape variations of European geometric microliths serve as cultural markers distinguishing groups of Mesolithic hunter-gatherers and Neolithic farmers. This suggests an evolving role for VL1 as the oldest site in the Alentejo region. The recovered geometric microliths further emphasize, as Cortell-Nicolau et al. (2020) highlight, that both the last hunter-gatherers and the first farmers used these tools, making them a reliable proxy for understanding the process of Neolithisation. The presence of both arrowheads and geometric microliths at VL1 provides archaeological evidence of the site playing a role in the Neolithic diffusion through direct contact and cultural exchange between agriculturalists and hunter-gatherers, facilitating the transfer of technology and cultural modifications (Cortell-Nicolau et al., 2020). Therefore, this further substantiates the evidence of VL1 as a notable site within the Iberian Peninsula, according to the Dual Model (Bernabeu 1996; Bernabeu et al. 1993).

In summary, geometric microliths are a specific type of lithic tool considered that is considered part of composite arrowheads (García-Puchol et al., 2014).

5.5 Comparison with Other Regional Neolithic Sites

Comparison is done with Neolithic sites from Portugal as discussed below:

5.5.1 The Menhirs of Alto da Cruz (Mora, Portugal)

Alto da Cruz Menhirs are megalithic monuments situated in the Mora Municipality, Évora District, Alentejo Region (South Portugal). These menhirs consist of a first menhir and a cruciform menhir, and they are of chrono-cultural significance in a funerary megalithic monument. The site is located very close to the contact zone of the Hesperian Massif and the Tagus Tertiary Basin (3 km north and 1 km to the northeast). The basement rocks are made up of porphyritic granites and outcropped with pegmatites. These menhirs, cruciform (designated as sector 1) and a single menhir (designated as sector 2), were investigated to understand their original standing positions or displacement, their functionality for funerary and ceremonial purposes, and chrono-cultural significance. The methodological assumptions proposed by Edward C. Harris were adopted for the stratigraphic excavations of the layers (Leonor, 2021).

The archaeological finds consist of handmade and wheel-made ceramics, various quartz flake types, and three Roman coins (evidence of reuse). The research confirmed that these menhirs have fallen from their original locations, except for two. Other discoveries include the use of cupules. Furthermore, the limited pottery and lithics recovered from the site suggest dates to the Early Neolithic (Leonor, 2021).

In comparison with VL1, the Alto da Cruz Menhirs appear to be older than VL1 based on the scarcity of pottery and lithic finds dated Early Neolithic, while VL1 is Middle to Late Neolithic. Also, cupules were present in the Alto da Cruz Menhirs Monuments but are completely absent in VL1.

However, both sites show similarities as related to reuse and the sites' function for funeral and ceremonial purposes.

5.5.2 Flint Sources and Mobility at the Chalcolithic (3500–2200 BCE) Settlement of Zambujal (Portugal)

The Zambujal Chalcolithic walled settlement is located on a hilltop about 120 m above sea level, on the right margin of the Pedrulhos River, which is a tributary of the Sizandro River. This settlement is found in the southwest of Torres Vedras in the Portuguese Estremadura. It was established at the start of the 3rd millennium and later deserted in the middle of the 2nd millennium BC. It has experienced more than five phases of construction (Kunst and Lütz, 2008). The Zambujal site is composed of both local and regional raw source materials. The local raw materials are Paleogene silcretes and Cenomanian flints, both in primary and secondary forms. The regional flint sources include the Alenquer flint source (a Paleogene deposit) that belongs to the Benfica Formation, composed of marly and calcaretes, and of Alcanede Limestones between Ota, Alenquer and Casais, coupled with silicites outcropping from the northern part of the Alenquer. Its silcrete is of pedogenic origin. It consists of palustrine and lacustrine as primary facies, and the Runa silcretes are secondary facies. They are characterized by microcrystalline to macrocrystalline matrices with chalcedony. Other regional flint sources are Sintra and Lisboa of Cretaceous origin. They are composed of Cenomanian limestones that belong to the Bica Formation. The Lisboa Formations consist of nodules and interlayered flints of conglomerate clasts (secondary sources). Lisboa facies are *wackestone* to *packstone* containing (*Gastropoda*, *Bivalvia*, *Ostracoda*, and *Foraminifera*). Sintra microfacies are mudstones with bioclasts containing (*sponge spines* and *Bivalvia segments*). Generally, Cenomanian flint consists of cryptocrystalline to micro quartz (Jordão and Pimentel, 2022).

Jordão and Pimentel (2022) investigated fifty-two geological samples to determine their local and regional sources based on petrographic analysis coupled with 1,200 lithic tools. The techno-typological analysis of the lithic tools consists of flake and bladelet cores, cortical flakes, core preparation material and debris, and non-cortical flakes and bladelets. Macroscopic analysis was used for the classification into groups and informal microfacies. The results indicated silcrete beds (breccia facies) and silcrete nodules (chalcedonic facies), which belong to the Paleogene. The Cenomanian flints consist of nodules to bedded forms formed by diagenetic replacement. It contains micro quartz, partially replaced carbonates and micrite aggregates. The bioclasts contain foraminifera, gastropods, ostracods, and sponge spines. The analysed samples were composed of flint, silcrete and chalcedony and classified into eight microfacies.

In comparison with VL1, the M10 (chalcedony and silcrete) of Zambujal settlement corresponds texturally to the M 23 S/C 3 of the VL1 site. Likewise, M1 of the Zambujal settlement corresponds texturally to REC SUP SC 2 of VL1. In terms of cortex, VL1 silcretes (M 23 S/C 3 and REC SUP SC 2) also contain cortex, which corresponds to the Zambujal settlement Paleogene silcrete. Moreover, the presence of opal in the Paleogene silcrete was observed in the M 23 S/C 3 of the VL1 site (needs further confirmation with Raman analysis).

In conclusion, the flint and silcrete raw materials proved to be sourced from both local and regional sources. Hence, this suggests that the prehistoric groups were highly mobile for the exploitation of local and regional sources to produce lithic tools.

CHAPTER SIX

6.0 CONCLUSION

6.1 Summary of Key Findings

Geochemical fingerprinting analysis has become a potent approach in archaeology for investigating the mobility of people, goods, and raw materials across prehistoric landscapes. This is achieved through the investigation of the elemental composition of artefacts such as lithics and geological raw material samples. This helps archaeologists to reconstruct the procurement strategy to gain insights into the prehistoric socio-economic behaviour during the Neolithic period.

The results from Anta 1 de Vale da Laje are vital to the wider debate about the Neolithisation process in the Iberian Peninsula because they show that communities were engaged in extensive social networks. The site functioned as a centre for various activities, including funerary, social, and economic. It facilitated both local gatherings and long-distance involvement in material and social exchange systems that extended beyond the immediate landscape. This enhances our understanding of connectivity, mobility, and cultural relationships during the Neolithic period.

The typological assemblages from VL1 offer insights into the diverse technological industries, seasonal and site-consistent use. The variations within the assemblage display functional, cultural, and symbolic dimensions of activity. The overall diversity in the lithic assemblages supports the idea of long-term and occasional use of the site, which probably signifies multiple phases of activity. The changes in the typological (qualitative) classification and quantitative (relative frequency of types) of the arrowhead assemblages are dependent on chronology and a reflection of the prehistoric people group's technology and local adaptation. The dominance of arrowheads with points suggests that they were deposited as burial goods and/or for other symbolic practices. The assemblage of arrowheads and geometric microliths at the VL1 site further establishes the significance of the site in the Neolithisation process in this region and as an important archeological site in the Iberian Peninsula, as it confirms the Dual Model theory of Neolithic diffusion.

The geochemical analysis results support the presence of Platinum Group Elements in the Tagus estuary. However, further analysis of high resolution is needed to confirm this result. Based on this result, it further reveals that prehistoric people utilized their local resources to produce lithic tools. This suggests that the people adapted to the local resources in their immediate landscape and revealed their level of cognition to understand the characteristics of these resources. This is further revealed in the silcrete obtained in this study, indicating that the people employed the resource management skills for the quality of material and knapping characteristics.

The spatial analysis of the lithic distribution across stratigraphic layers displays a non-random pattern, which provides evidence of changes in technology, phases of occupation, long-term use of the site, and the confirmation of the evolving role of the VL1 site from the Mesolithic hunter-gatherers and/or Neolithic agropastoralists

The discovery of non-local lithics suggests that VL1 was part of a larger Neolithic exchange network. Interaction could have occurred via down-the-line exchange, seasonal migrations, or social ties supported by megalithic monument traditions. These patterns imply that the mobility of materials served not only practical purposes but also helped maintain wider social bonds.

Lithic mobility held both functional and symbolic importance. Although local materials met practical needs, the presence of both local and non-local lithics in funerary settings suggests intentional choices that extend beyond mere utility. Placing these materials within the monument turned the site into a centre for social activities, identity, and interaction. Thus, the mobility of raw materials revealed how material culture played a role in shaping social and ritual aspects of Neolithic life.

Additionally, the identification of local, regional and distant sources indicates the VL1 involvement in local, regional, and wider social networks. Ultimately, this research underscores the significance of material culture in fostering social interaction, thereby enriching our understanding of exchange, connectivity, and cultural expression in Neolithic Portugal.

6.2 Contributions to Knowledge

This research contributes to prehistoric lithic technology, raw material sourcing, and behavioural patterns of the prehistoric people at the VL1 site by integrating typological, geochemical, and spatial analyses. The key contributions are outlined below:

The study presents an innovative approach that combines several different analytical methods to investigate the origin of lithics. This approach integrates typological classification, portable X-ray fluorescence (pXRF), scanning electron microscopy (SEM), and multivariate statistical analyses (PCA) with spatial analysis. This integrated approach provides a comprehensive understanding of the type and geochemical composition of lithic artefacts. This multi-method strategy is a relatively novel application to a single site in southwestern Iberia, and it serves as a background for future studies in similar archaeological contexts.

The classification and groupings of arrowheads into eight typologies and microliths into five types offer insights into the various groups that visited the VL1 site, since it is known that each group of agropastoralists is unique in its technology. It also confirms that different subsistence economies/occupations and other activities were performed on the site. The dominance of unbroken/unused arrowheads highlights the VL1 site's function as a funerary site. This study's typological groupings provide a reference for comparative studies within the region and other regions.

Comparing the elemental composition of VL1 lithic artefacts with the ITM Rio Maior flint source, this study revealed that some of the arrowheads typology and geometric microliths were probably sourced from the Rio Maior flint source, which indicates that the prehistoric people utilized resources beyond their immediate landscape probably for quality materials, social networks, and other practices. However, some arrowheads typology and debitage residues indicate different other local/distant sources, exchange or trade. This investigation contributes to the provenance study, prehistoric mobility patterns, landscape strategy, and exchange networks in the Iberian Neolithic period.

The SEM analysis revealed the presence of silcrete in some of the artefacts. This is novel in the study of the lithic assemblage of the VL1 site. This further confirms the prehistoric peoples' cognitive and technological knowledge in their preferences for raw materials from local and distant sources to make specific tools for specific purposes, such as funerary rites. These findings highlight VL1 site function and symbolic practices of prehistoric cultures.

Spatial distribution of diverse lithic tool types across stratigraphic layers at VL1 revealed changes in the groups of people, technology, phases of occupation, and other symbolic practices over time at the site. The presence of diverse arrowheads across multiple layers indicates long-term or repeated occupation, while the changes in the geometric microliths across the stratigraphic layers display a preference for tool usage and subsistence economy. These spatial patterns contribute to a wider interpretation of subsistence strategy and land-use practices,

Overall, this study contributes a comprehensive analysis of the understanding of lithic technology, raw material sourcing, and prehistoric behaviour at VL1 site. The results offer insights into knowledge on procurement strategies, technology, mobility, and other symbolic practices during the Neolithic period of the Iberian Peninsula.

6.3 Limitations of Methodological Research

The limitations of the pXRF method used include the following:

i. Inability of pXRF to detect Rare Earth Elements (REE), which are best suited for provenance studies.

ii. It cannot determine the geographical origins of diverse materials.

iii. Since pXRF is not sensitive to contamination, if artefacts were dirty, patinated, and weathered, results may reflect surface contamination rather than the true internal composition.

iv. pXRF results vary based on the detection time of the instrument, the type and water content of the sample.

v. The difference in the precision and range of pXRF.

vi. Inability to perform destructive analysis on some of the geological samples for thin sectioning to study the mineralogical composition of the samples.

vii. Inability to perform petrographic and other non-destructive analysis on the archaeological samples.

Others include the inability to get samples from other sources within short and long-range distances to the VL1 site.

6.4 Recommendations for Future Research

Since no single methodological approach is sufficient to determine geochemical provenance. To comprehensively confirm these findings, the researcher hereby recommends the following:

- Inductively Coupled Plasma Mass Spectrometry (ICP-MS) for detecting ultra-trace elements and enhancing provenance resolution.
- Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) for precise multi-element quantification.
- Energy-Dispersive X-Ray Fluorescence (EDXRF) to determine the elemental composition of artefacts.
- Paired with Energy-Dispersive X-ray Spectroscopy (EDS/EDX) for wider elemental coverage and verification of chemical composition.
- Laser Ablation ICP-MS for minimally destructive, micro-scale compositional analysis of samples.
- Strontium ($^{87}Sr/^{86}Sr$) Isotope analysis to assess geographic origin and mobility patterns.
- Use Wear Analysis to determine the specific functions of the different tool types.
- Other raw material sources within short- and long-range distances to VL1 must be sampled for the provenance study.
- The exposure time of the pXRF machine on the samples should be increased to more than 30 seconds to observe if the artefacts and the raw materials will give different elemental composition results.

APPENDICES

Appendix Table 1: Debitage Residues pXRF Result

DEBITAGE RESIDUES																
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Zr %**	**Ni %**	**Co %**	**Cu %**	**Mg %**	**V %**	**Al %**	**Ti %**	**Ta %**	**S %**
26 G S/C 8	94.05	2.49	2.12		0.91	0.29	0.08	0.03	0.02	0.01						
M 23 S/C 3	88.17	1.65	2.53	1.49	1.21	2.53		0.07			2.34					
M 23 S/C 1	94.53	1.87	1.51	1.11	0.64	0.32		0.02								
24 G S/C 812	94.63	2.19	1.48	1.51		0.08	0.06	0.04								
24 G S/C 893	96.91	1.63	0.69		0.62	0.12		0.02								
21 M SC 1	96.65	1.38	1.27		0.66			0.03	0.01							
24 G S/C 1022	95.38	1.82	1.09	1.03	0.44	0.2		0.03								
SC 1	96.74	1.57	1.15		0.51			0.03								
24 F-S/C 10	93.59	1.68	2.19	1.09	0.93	0.41		0.06				0.04				
27 G S/C 30	93.18	2.13	1.79	1.29	0.71	0.87		0.03								
28 G 15	91.68	1.75	2.23	1.46	1.16	1.65		0.05								
24 G S/C 894	94.71	1.49	1.76	1.09	0.78	0.08		0.04								
26 G S/C 446	95.3	1.89	1.07	1.06	0.66			0.02								
27 F Nivel 5 S.C. 31	95.46	1.79	1.14	1.06	0.39	0.14		0.02								
28 G N6 SC 35	88.6	1.96	1.46	1.47	0.86	0.65		0.02		0.01			4.98			
REC SUP SC 2	87.72	1.57	1.74	1.14	0.68	3.16	2.77	0.03						1.1		
24 G S/C 891	96.44	1.83	1.11		0.45	0.16		0.01								
28 F S/C 12	94.91	1.68	1.42	1.07	0.8			0.04							0.08	
27 G 71	86.32	2.15	1.96	1.54	1.01	0.46		0.04					6.18			0.3
27 G S/C 203	93.83	1.52	2.24	1.46	0.82	0.09		0.04								
Caco S/C 648	92.88	1.81	2.58	1.52	0.99	0.17		0.05								
24 F 10	84.12	2.26	2.45	1.92	0.74	1.77		0.06					6.43	0.24		
24 F SC 4	93.89	1.65	1.89	1.24	0.98	0.27		0.05								
28 H 24	96.5	1.81	1.13		0.42	0.11		0.02								
23 H S/C 4	95.06	1.87	1.31	1.12	0.57	0.04		0.02								

Appendix Table 2: Concave-based Arrowheads pXRF Result

CONCAVE-BASED ARROWHEADS																	
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Co %**	**V %**	**Al %**	**Ti %**	**Ta %**	**S %**	**Mn %**	**As %**	**Cd %**	**Hg %**
28 E 1	90.8	2.03	1.3	1.24	0.43	0.26	0.02			3.9				0.02			
26 G-50	96.69	1.69	0.94		0.64		0.02	0.01									
28 E 20	95.24	1.89	1.03	1.12	0.46	0.15	0.02					0.06		0.02			
29 H-2	96.63	1.73	1.08		0.53		0.03										
23 L S/C 1	95.56	1.73	0.91	1.07	0.43	0.12	0.03		0.02		0.1						
27 F 13	95.29	1.91	1.13	1.1	0.43	0.13	0.02										
28 F 21	94.98	1.72	1.32	1.05	0.55	0.15	0.02								0.21		
24 K S/C 1	95.14	1.87	1.36	1.07	0.41	0.14	0.03										
25 G 40	96.76	1.56	1.09		0.55		0.02										
29 G-9	96.86	1.58	0.98		0.47	0.08	0.02										
Caco 30	96.52	1.86	1		0.49	0.08	0.03	0.01									
28 E 9	93.93	2.2	1.33	1.42	0.59	0.36	0.03				0.1						
27 G S/C 22	95.18	1.74	1.06	1.08	0.57	0.34	0.02										
26 G 52	96.36	1.72	1.23		0.6	0.06	0.02										
23 H NIV. 9 V.E. 29	96.36	1.69	1.14		0.42		0.03						0.36				
27 E S/C 32	97.13	1.51	0.87		0.45		0.02										
26 G 127	97.07	1.62	0.8		0.47		0.03										
27 F 33	96.58	1.78	1.1		0.41	0.1	0.03										
24 G 22	96.75	1.54	1.08		0.57		0.03										
Sara 3	96.39	1.68	1.33		0.57		0.03										
28 H-23	96.48	1.59	1.08		0.58		0.02									0.25	
24 G S/C 897	97.04	1.52	0.91		0.5		0.02										
27 G 49	96.57	1.66	1.2		0.54		0.03										
27 G SC 73	96.68	1.76	0.98		0.54		0.03										
26 G 37	95.67	1.73	0.74	1.09	0.74		0.03										

Appendix Table 2 Continued

Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Co %**	**V %**	**Al %**	**Ti %**	**Ta %**	**S %**	**Mn %**	**As %**	**Cd %**	**Hg %**
Caco S/C 303	96.79	1.97	0.92		0.58		0.03										
21 LS/C 1	94.77	1.95	1.43	1.13	0.69		0.03										
26 G 134	96.79	1.69	0.99		0.5		0.02										
27 E S/C 29	95.32	1.92	1.09	1.09	0.53		0.03										0.02
23 I SC 1	96.06	1.67	1.38		0.53	0.33	0.02										
24 G 15	96.3	1.72	1.38		0.45		0.02				0.02						
25 G 1	96.64	1.73	0.9		0.57		0.03				0.12			0.01			
24 G 1	96.92	1.62	0.89		0.54		0.02										
27 G-9	95.26	1.75	1.21	1.18	0.57		0.03										
26 G 149	95.96	1.97	1.17		0.67	0.18	0.05										
28 F 23	95.46	1.75	1.07	1.05	0.55	0.1	0.02										
26 G S/C 73	95.25	1.65	1.32	1.1	0.67		0.02										

Appendix Table 3: Straight-based Arrowheads pXRF Result

STRAIGHT-BASED ARROWHEADS															
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Co %**	**V %**	**Al %**	**Ti %**	**Ta %**	**Cu %**	**Mn %**	**S %**
29 H 24	92.93	2	1.5	1.01	0.61	1.89	0.04						0.01		
28 G SC-20	94.99	1.98	1.09	1.17	0.53	0.2	0.03								
24 G S/C 1024	95.53	1.76	1.13	1.04	0.47	0.04	0.03								
Caco S/C 650	92.68	2.73	1.69	2.11		0.69	0.04		0.04						
27 G S.C 187	94.06	1.92	1.57	1.21	0.59	0.46	0.03				0.14				
26 G 101	94.38	1.76	1.03		0.41	2.29	0.02				0.11				
28 E 24	88.36	1.76	2.04	1.53	0.67	0.74	0.05			4.6	0.23				
28 E-76	94.05	2.25	1.35	1.34	0.73	0.21	0.03							0.04	
27 E SC 3	93.4	2.21	1.62	1.35	0.94	0.24	0.04				0.18			0.02	
28 E 3	90.02	3.09	2.67	1.57	1.36	0.67	0.06								0.55
Caco 18	96.32	1.86	1.11		0.51	0.12	0.02					0.07			
28 E 2	96.6	1.75	1.04		0.55		0.02	0.01							
28 H S/C 11	74.36	6.34	5.73	5.86	4.89	1.75	0.16								0.41
24 M 1	96.38	1.47	1.21		0.62	0.25	0.03						0.01	0.02	
24 G S/C 896	97.15	1.53	0.85		0.44		0.03								
Caco 23	96.19	1.88	1.2		0.56	0.14	0.02								
29 G S/C 4	96.51	1.58	1.35		0.47	0.05	0.02							0.02	
28 G SC 19	95.15	1.93	1.13	1.08	0.52		0.04				0.15				
Caco S/C 649	93.63	2.05	1.48	1.24	0.61	0.26	0.03				0.22		0.01		0.46
28 G S/C 202	94.37	1.72	1.55	1.35	0.93	0.04	0.03								

Appendix Table 4: Leaf-based Arrowheads pXRF Result

LEAF-BASED ARROWHEADS															
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Mn %**	**Ni %**	**S %**	**Cu %**	**Ta %**	**Ti %**	**Mg %**	**V %**	**Co %**
28 E S/C 29	96	1.51	1.39		0.57	0.05		0.03			0.01				
27 F 23	95	1.96	1.29	1.2	0.57	0.33		0.03							
23 K S/C 2	91	2.08	1.35	1.2	0.67	3.13		0.03	0.35			0.23			
E 28 134	96	1.73	1.03		0.64	0.36		0.03		0.01					
27 E 3	96	1.77	0.96		0.45	0.85		0.02				0.2			
27 G S/C 74	95	1.94	1.16		0.38	1.3	0.02	0.01							
28 F 11	96	1.47	1.39		0.38	0.13		0.03				0.14			
29 G-5	93	1.95	1.26	1.1	0.54	0.37		0.03				0.21	1.97		
28 H SC 9	94	2.1	1.2	1.2	0.53	0.04		0.03	0.58						
27 H-1	97	1.4	1.1		0.6	0.38		0.03							
27 G S/C 182	94	2.13	1.57	1.3		0.85		0.03						0.03	
28 F 25	96	1.46	1.38		0.79	0.33		0.02				0.13			
28 H 34	95	1.71	1.23	1.1	0.72	0.28		0.03							
28 E-37	95	1.87	1.44	1.04	0.68	0.1		0.04		0.01					0.01
27 F 30	88	2.92	3.61	2.7	1.7	0.71		0.06							
26 G S/C 439	97	1.64	1.11		0.53			0.03						0.04	

Appendix Table 5: Tanged-based Arrowhead pXRF Result

TANGED-BASED ARROWHEADS										
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Ti %**	**Mn %**	**P %**
28 G 11	94.42	2.07	1.52		1.03	0.49	0.03	0.21		0.23
Sara 4	95.07	1.94	1.44	1.1	0.4		0.04		0.02	
28 F S/C 8	96.4	1.89	1.2		0.47		0.03			
28 E 136	94.78	1.87	1.15	1.15	0.71	0.31	0.03			

Appendix Table 6: Rhomboid/Rombus eye pXRF Result

RHOMBOID/ROMBUS EYE ARROWHEADS						
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Ni %**
28 E 53	94.61	2.1	1.4	1.26	0.6	0.02

Appendix Table 7: Cruciform/Side Appendages pXRF Result

CRUCIFORM/SIDE APPENDAGES ARROWHEADS									
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Mg %**	**Al %**
29 G-3	90.7	1.97	1.19	1.17	0.48	0.08	0.02	4.39	
24 G S/C 1164	89.98	2.35	1.39	1.36	0.76	0.36	0.04		3.76

Appendix Table 8: Halberd pXRF Result

HALBERD								
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Mn %**
26 G 143 (outside)	94.97	1.59	1.37	1.18	0.68	0.17	0.02	0.02
(Inside)	92.8	2.17	1.95	1.75	1.01	0.25	0.04	0.02

Appendix Table 9: Indeterminates Arrowheads pXRF Result

INDETERMINATES ARROWHEADS									
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Mn %**	**Ni %**	**Al %**
28 H SC 19	93.26	1.85	1.87	1.54	1.22	0.18	0.03	0.04	
29 G 4	97.04	1.56	0.83		0.54			0.02	
28 E S/C 3	96.89	1.66	0.84		0.59			0.02	
28 H S/C 21	93.74	2.25	2.07	1.36	0.46	0.08		0.03	
29 HS/C 6	96.72	1.68	0.92		0.64			0.02	
28 E-135	96.88	1.64	1.05		0.4		0.01	0.02	
27 F S/C 2	89.52	2.17	1.26	1.14	0.56	0.74		0.04	4.57

Appendix Table 10: Triangles Microliths pXRF Result

TRIANGLES MICROLITHS																					
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Ti %**	**Zn %**	**Al %**	**Mn %**	**V %**	**Cu %**	**S %**	**Zr %**	**Bi %**	**Ta %**	**Pb %**	**Y %**	**P %**	**Mg %**
25 G SC 65	92.75	1.51	2.62	1.2	0.97	0.09	0.05	0.81													
28 F S/C 14	56.1	3.17	2.43	1.59	1.44	1.29	0.17	11.68	0.18	21.68		0.33	0.03								
28 G n6 S/C 35b	64.91	2.03	3.01	1.38	1.37	1.23	0.09	9.71	0.16	15.48		0.31		0.32							
28 F S/C 3	94.56	1.92	1.39	1.18	0.78	0.12	0.03														
27 G S/C 39	92.74	1.51	1.28		0.47	0.54	0.03	1.18							2	0.17					
24 G S/C 895	96.34	1.75	1.16		0.54	0.07	0.03					0.05			0.06						
27 F 6	91.95	1.97	2.2	1.58	1.27	0.31	0.06	0.6				0.06									
24 F S/C 45	96.57	1.61	0.95		0.6	0.04	0.03	0.2													
28 G n6 S/C 35a	79.95	2.25	1.8	1.54	1.32	0.46	0.04	6.33	0.08	5.99		0.23									
28 E 4	84.43	1.72	2.24	1.21	0.93	0.57	0.05	2.99	0.05	5.43		0.13	0.01								
27 G 64	60.69	2.84	1.48	1.13	1.28	0.98	0.05	12.29	0.2	18.65		0.39	0.01								
28 G nivel 4 S.C. 25	92.33	2.03	1.58	1.54	1.24	0.37	0.04	0.85													
25 E n6 S/C 8a	92.55	2.25	1.81	1.57	1.18	0.09	0.04	0.35			0.02						0.08				
24 G S/C 1025	72.54	2.58	1.73	1.29	1.12	0.8	0.03	8.52	0.1	11.01	0.03	0.25									
26 G S/C 137	94.71	1.73	1.62	1.13	0.63		0.02	0.16													
22 H S/C 17	70.26	1.71	1.96	1.63	0.84	0.3	0.05	0.55		21.78					0.17		0.1	0.48	0.17		
Caco S/C 318	95.43	1.8	1.17	1.06	0.4	0.1	0.02														
27 G 60	78.95	2.1	1.99	1.47	0.77	2.13	0.04	0.45		11.66			0.01							0.39	
Caco S/C 653	91.86	1.95	2.44	1.7	1.32	0.65	0.06														
28 F S/C 13	73.45	5.48	7.2	3.24	2.08	2.09	0.13			3.98			0.04								2.31

Appendix Table 11: Trapezoids Microliths pXRF Result

TRAPEZOIDS MICROLITHS													
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Cu %**	**Ti %**	**Zn %**	**Al %**	**Mn %**	**V %**
29 H 6	94.82	2.02	1.23	1.08	0.41	0.16	0.03		0.21				0.03
F 28 13	95.34	1.65	1.27	1.16	0.54		0.03						
28 E 82	92.86	2.12	1.93	1.51	0.83	0.16	0.04		0.54				
27 G SC 184	96.77	1.46	1.11		0.57	0.04	0.03						
28 H 40	94.8	1.87	1.07		0.84	0.06	0.03		1.3				
29 H S/C 9	66.48	3.43	2.32	1.79	1.24	0.85	0.05	0.01	7.24	0.12	16.3		0.21
27 G 59	90.3	1.67	1.48		0.85	3.91	0.04		1.62			0.03	
28 G S/C 1a	95.01	1.83	1.24	1.02	0.76	0.1	0.03						
28 E 97	94.65	1.56	1.06		0.68	1.44	0.03		0.57				
29 G-10	94.82	1.77	1.54	1.16	0.64		0.03						0.04
Caco S/C 376	66.04	2.39	1.9	1.39	1.6	0.99	0.05	0.02	10.1	0.15	15		0.32
28 H S/C 19	78.16	2.5	1.58	1.37	1.49	0.49	0.05		5.81	0.09	8.23		0.24

Appendix Table 12: Crescents Microliths pXRF Result

CRESCENTS MICROLITHS														
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Ti %**	**Zn %**	**Al %**	**Mn %**	**V %**	**As %**	**Co %**
29 H-23	93.73	1.85	1.77	1.32	0.95	0.06	0.04	0.26			0.02			
Caco S/C 18	94.63	1.63	1.8	1.25	0.65		0.04							
21 G S/C 1	96.22	1.26	1.43		0.5	0.57	0.02							
29 H 26	95.56	1.68	1.16		0.55		0.02							
27 G 45	94.46	1.4	1.6	1.03	0.82	0.5	0.03	0.16						
28 E n6 S/C 43	93.67	2.2	1.62	1.13	0.73	0.17	0.03	0.43						
27 G 194	75.19	2.12	2.45	1.65	1.67	2.09	0.05	3.84	0.04	10.71		0.2		
28 H 25	92.82	1.78	1.52	1.31	0.82	1.23	0.03				0.02		0.24	
28 H S/C 18	74.3	1.66	1.84	1.2	0.59	13.18	0.02			6.7				0.15
28 F S/C 8	96.44	1.71	1.11		0.72		0.03							
28 H S/C 32	96.37	1.67	1.4		0.52		0.03							
23 H S/C 45	96.74	1.38	1.17		0.49		0.03	0.05						

Appendix Table 13: Indeterminates Microliths pXRF Result

INDETERMINATES MICROLITHS																			
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Ti %**	**Zn %**	**Al %**	**Mn %**	**V %**	**Cu %**	**S %**	**Zn %**	**Mg %**	**Co %**	**P %**	**Cd %**
22 H S/C 24	36.14	4.97	3.55	2.68	2.48	5.19	0.08	12.32	0.31	31.2		0.45	0.02	0.4		< LOD		0.25	
27 G S/C 5	89.05	2.53	2.11	1.7	0.96	0.5	0.05	2.36						0.7			0.02		
28 E 19	87.36	2.65	2.05	1.65	1.13	0.43	0.03	0.18		4.46	0.04								
28 F N7 S/C 18/19	80.93	2.33	1.48	1.27	1.06	0.44	0.04	3.89		8.39		0.11			0.06				
29 H 26	96.36	1.45	1.32		0.54		0.03												0.3
27-G Nivel 9 S.C. 189	63.64	2.65	2.55	1.46	1.18	1.09	0.08	11.36	0.11	15.7		0.22							
24 S/C 61	45.63	3.11	2.12	1.39	1.61	1.26	0.04	14.19	0.24	29.5		0.45						0.11	
24 E S/C 9	84.76	2.05	1.45	1.16	0.92	0.47	0.05	3.54	0.04	5.51			0.02						
24 F Nivel 6 S.C. 1	94.1	1.93	1.24	1.15	0.95	0.12	0.02	0.44											
27 G 22	38.35	3.5	2.35	1.52	1.68	1.44	0.08	15.62	0.3	34.3		0.39	0.03	0.3		< LOD		0.12	
27 F Nivel 5 S.C. 30b	75.52	2.45	1.6	1.03	1.05	0.63	0.03	8.12	0.13	9.21		0.2	0.02						
24 G S/C N 900	92.67	1.81	1.23		0.59	0.16	0.02	3.33	0.06			0.13							

Appendix Table 14: Miscellaneous Microliths pXRF Result

MISCELLANEOUS MICROLITHS																			
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Ti %**	**Zr %**	**Al %**	**Mn %**	**V %**	**Cu %**	**Mg %**	**Co %**	**P %**	**Mo %**	**Pt %**	**As %**
23 H S/C 10	91.66	1.89	1.08	1.22	0.52	1.13	0.02	1.29	1.11						0.02		0.05		
24 F SC 2	92.99	2.73	1.94	1.51	0.61	0.17	0.04												
28 E Nivel 3 S.C. 14	94.82	1.55	1.02		0.59	1.77	0.02	0.22											
25 G n4 S/C 47a	96.89	1.28	1.04		0.48	0.04	0.02	0.22			0.01								
23 H S/C 3	93.45	1.82	1.85	1.3	0.58	0.28	0.03	0.6											
Caco SC 20	94.95	1.68	1.5	1.04	0.74	0.05	0.02				0.02								
27 G S/C 36	78.48	2.72	2.18	1.99	1.25	0.88	0.05	1.85		10.5		0.14							
22 H S/C 5	94.05	1.52	1.46		0.64	0.15	0.03	2.06				0.08							
Rec Sup S/C 17	94.66	1.71	1.54	1.08	0.69	0.13	0.03											0.15	
28 F SC 47	93.85	1.73	1.31	1.13	0.83	0.11	0.04	0.96							0.01				
21 H S/C 13	93.34	1.71	1.71	1.41	0.87	0.65	0.03	0.28											
27 H S/C 2	87.38	2.87	1.97	1.98	0.54	0.31	0.04	0.19		4.69	0.02								
22 G 62	96.35	1.7	1.17		0.67	0.06	0.03				0.01								
24 F 182	93.24	2.24	1.9	1.29	1.03	0.23	0.05												
27 G 61	93.74	1.74	1.76	1.23	0.59	0.5	0.04	0.3											

Appendix Table 15: ITM Rio Maior Flint pXRF Result

ITM RIO MAIOR FLINT														
Artefact Number	**Si %**	**Ru %**	**In %**	**Rh %**	**Pd %**	**Fe %**	**Ni %**	**Ti %**	**Mn %**	**Cu %**	**Mg %**	**Co %**	**S %**	**Y %**
Sample 1	92.86	1.62	2.59	1.54	1.01	0.28	0.06			0.02				
Sample 2	82.55	2.03	6.4	3.16	1.92	0.62	0.09				2.61	0.05	0.41	
Sample 3	85.78	1.51	5.47	2.42		0.42	0.1		0.1		3.72		0.46	
Sample 4	96.35	1.29	1.58		0.71	0.04	0.02							
Sample 5	94.48	1.55	1.96	1.05	0.81	0.09	0.06							
Sample 6	84.6	1.27	5.61	2.18		0.3	0.11				5.19		0.59	0.13
Sample 7	92.02	1.74	2.76	1.68	1.09	0.44	0.05	0.21						
Sample 8	88.97	0.78	4.02	1.64		0.26	0.11				3.63		0.53	
Sample 9	96.68	1.53	1.25		0.45	0.05	0.02							
Sample 10	91.35	1.17	2.57	1.44	0.9	0.27	0.06				2.24			
Sample 11	94.77	1.74	1.61	1.05	0.72	0.06	0.02							
Sample 12	91.65	1.55	3.42	1.94	1.08	0.19	0.05		0.08			0.03		

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