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Regional provenance of dolerite prehistoric objects through mineral analysis

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ABSTRACT

A methodology based on the mineral analysis determination has been developed to identify the origin of dolerite stone outcrops collected to fabricate lithic objects during the Prehistoric period. The method is based on the use of inductively coupled plasma mass spectrometry (ICP-MS) to analyse rare earth elements (REE) and trace elements. Additionally a no destructive geochemical analysis based on X-ray fluorescence (XRF) was employed for major elements analysis. The aforementioned methodologies were applied to samples from different archaeological fields or natural outcrops located in the Mediterranean area of Spain, between Valencia and Alicante. Principal component analysis (PCA) was employed to interpret the dolerite geological provenance. These preliminary results show that statistical analysis permits to distinguish stone sample origins according to their REE profile at regional level and that Ti/Fe major element relation perhaps shows just coarse differences between samples collected on the extreme north and south of the studied region. The proposed method could be useful to discriminate the regional origin of lithic objects belonging to dolerite rocks and to interpret the primary material transport and exchange of lithic materials in Valencian Prehistory.

1. Introduction

Geochemical studies of lithic artefacts try to find a relationship between the archaeological materials and the primary source present in the nature using sample chemical profile. In the last decades the determination of the geologic source and provenance of lithic objects and west flakes has become a standard practice in archaeological research to evaluate prehistoric migrations and fluxes. Provenance information results are useful to rebuilt settlement patterns, and investigate stone tool technologies, exchange systems and territoriality [1–10].

Technological advances have recently improved the ability to determine the source of archaeological materials. Archaeological lithic provenance studies have typically focused on ground stone tools, flints and obsidian. Obsidian is a volcanic glass widely used for stone tools; it is an ideal material for reconstructing exchange systems since it occurs geologically in a limited number of location, and is frequently found in archaeological sites even far from a source and may be chemically fingerprinted, allowing source attribution of artefacts based on it which has been designed a multi-method exploratory approach to chemically characterise geological obsidian samples from Sardinia [9]. For the full chemical characterisation of geological samples a combination of electron microprobe analysis, X-ray fluorescence (XRF), neutron activation analyses (NAA) and inductively coupled plasma mass spectrometry (ICP-MS) was employed. The results provided detailed view of the distribution of obsidian from the central Mediterranean Island sources. Some studies have been centred on polished stone axes that are important components in Neolithic manufacturing techniques being made with the petrological characterisation of Neolithic polished stone axes found in several archaeological sites in central Sardinia [1]. A petrographical description was given for all encountered lithologies, and bulk rock chemical analyses of minerals of 12 representative samples were given, with the aim of determining the origin of the raw materials. The mineralogical and petrological characterisation showed useful information to distinguish between exotic ("nephrite" and glaucophane schist) and local (phonolite) Sardinia polished stone axis raw material origins.

Williams-Thorpe et al. [10] studied one of the best known petrological groups of polished stone implements found in England and Wales which comprise axes and related artefacts made of fine to medium-grained quartz dolerite. These implements of Neolithic and Bronze age date are termed Group XVIII. The aim of this work was to establish geochemical and magnetic characteristics of Group XVIII implements using non-destructive methods X-ray fluorescence analysis (XRF) and magnetic susceptibility measurement, to compare the chemical and magnetic characteristics with those of potential sources including the Whin Sill, consisting on outcrops or secondary deposits which crops out extensively in northern England, additionally than other igneous outcrops in northern Britain. In this way it was tested the validity of source(s) of Group XVIII and produced a geochemical and magnetic

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description for the Group which can help in non-destructive provenancing of dolerite artefacts. However the aforementioned instruments proved to be very effective in testing the proposed Group XVIII source implements, but did not always provided enough information to identify specific alternative source for non-Group XVIII implements. The methods were considered by the authors not suitable for very weathered artefacts which have not fresh surfaces available for measurements.

Geochemical analysis were also applied in a world heritage site as Stonehenge [2,3] to explore the correlation between the rhyolitic and dacitic lithologies from the site and the two main volcanic sequences of Ordovician age belonging to the Fishguard Volcanic Group and the Sealyham Volcanic Formation exposed across north Pembrokeshire . The work of Bevins et al. [2] was innovative in the use of zirconium (Zr) chemistry using laser-ablation ICP-MS analysis in archaeopetrological provenancing and in a second work Bevins et al. [3] concluded that the only dacitic or rhyolitic lithology which can be matched with any degree of confidence between the Stonehenge landscape and a specific source area is the so-called 'rhyolite with fabric' lithology, which matches with foliated rhyolitic rocks exposed in the Pont Saeson area of north Pembrokeshire, and Craig Rhos-y-felin. This is correlated with some confidence in terms of petrography, whole rock geochemistry and mineral chemistry.

In this preliminary study we have tried to identify outcrop raw materials collected for manufacturing lithic objects during the Prehistoric period employing ICP-MS to analyse rare earth elements (REE) and trace elements. No destructive geochemical analysis consisted on the use of XRF was also employed for determination of major elements. The ultimate goal was to develop a method to complete the reconstruction of transportation networks of prehistoric tools already developed in a comprehensive study of thin film analysis with polarizing microscope [11]. Oxides, major elements, trace elements and rare earth elements (REE) were determined in a total of 16 samples including 13 natural and 3 archaeological stone samples. All of them collected in archaeological fields or natural outcrops located in the Mediterranean area of Spain, between Valencia and Alicante region. The multi-element capability of ICP-MS was employed to identify changes in trace elements and REE between samples [12,13]. XRF is a technique that permit direct, fast, cheap and safe analyses for archaeological sample compared with mass spectrometry one. We employed XRF to analyse specimens and elements trying to identify markers that permit us to observe differences between samples and identify archaeological objects raw material provenance avoiding the use of destructive ultra-trace (REE) analysis.

Principal component analysis (PCA) was used to identify archaeological dolerite samples of raw material provenance employing REE as variables. Ti/Fe relations were pin-point as a coarse provenance marker for dolerite objects belonging to the studied region.

2. Materials and methods

2.1. The studied samples

An amount of 16 dolerite samples were analysed; 13 from natural outcrops were exploited during the antiquity as quarries and 3 prehistoric utensil fragments (Fig. 1) from archaeological work-field samples were collected from different areas of Valencia Community (Fig. 1). Sample numbers and geographical origin are summarized in Table 1. Samples S1, S2, and S3, were collected in Finestrat (Orxeta, Alicante) outcrop employed as an ancient quarry, composed of a relativity fresh igneous rock defined as dolerite labrador and piroxene. Samples S4, S5, S6, S7, and S8 are from Pinós, Xinorlet-Font d' Almorquí y Sax (Alicante) outcrops, following a certain structural tectonic alignment by the intersection of various failures. The basic rock outcrop of Pinos is located on the eastern slope of the saline dome of Cabezo de la Sal; materials here have two textures: one of the outcrop edges and one (ortophyric to subtrachytic) and the rock mass (monocrystalline hypidiomorphic dibasic). At NW of Sax into the Vinalopó (Santa Eulalia) depression, several hints of dolerite are found and some of them are difficult to locate, because they are small in size and are located at ground level. The larger quarry (CG) is dead today. These materials have been classified as trachyandesites and microgabbro with coarse andesina.

Sample S9 comes from Sierra Orihuela, precisely from the hint call "el Tunel" and intense exploitation. The rock in this outcrop is relatively fresh, very compact, dark greenish colour and size of fine and homogeneous

Fig. 1. Collected sample locations. Dolerite polished stone. "Arenal de la Costa" object raw material provenance (brown colour). "Bancal de Satorre" object raw material provenance (blue colour). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)