



UMEÅ UNIVERSITY

CARVED MOUNTAINS AND MOVING STONES

Applications of Near Infrared Spectroscopy for Mineral
Characterisation in Provenance Studies

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“In stones the beauty common to all the kingdoms of nature seems vague, even diffuse, to man, a being himself lacking in density, the last comer into the world, intelligent, active, ambitious, driven by an enormous presumption. He does not suspect that his most subtle researches are but an exemplification within a given field of criteria that are ineluctable, though capable of endless variation. Nonetheless, even though he neglects, scorns, or ignores the general or fundamental beauty which has emanated since the very beginning from the architecture of the universe and from which all other beauties derive, he still cannot help being affected by something basic and indestructible in the mineral kingdom: something we might describe as lapidary that fills him with wonder and desire”.

Roger Cailliois,

The Writing of Stones. University of Virginia Press 1985, p. 3.

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List of papers

- I. Linderholm, Johan, Paul Geladi, and Claudia Sciuto. 2015. 'Field-Based near Infrared Spectroscopy for Analysis of Scandinavian Stone Age Rock Paintings'. *Journal of Near Infrared Spectroscopy* 23 (4): 227–36.
- II. Sciuto, Claudia, Johan Linderholm, and Paul Geladi. 2017. 'Near Infrared Imaging Spectroscopy for Raw Materials Characterization: The Example of a Mesolithic Dwelling Site in Northern Sweden'. In Pereira Telmo, Xavier Terradas and Nuno Bicho (eds.) *The exploitation of Raw Materials in Prehistory. Sourcing, Processing and Distribution*, Cambridge Scholars Publishing. 113-120.
- III. Sciuto, Claudia, Lorenza La Rosa, Paul Geladi, Johan Linderholm and Mikael Thyrel 2018 'Hyperspectral Imaging for Characterization of Lithic Raw Materials. The Case of a Mesolithic Dwelling in Northern Sweden', *Journal of Lithic Technology*, <https://doi.org/10.1080/01977261.2018.1543105> (published online).
- IV. Sciuto, Claudia, Dominique Allios, Ryad Bendoula, Antoine Cocoual, Marie-Elise Gardel, Paul Geladi, Alexia Gobrecht, Natalie Gorretta, Nominoë Guermeur, Silvain Jay, Johan Linderholm and Mikael Thyrel 2019 'Characterization of Building Materials by Means of Spectral Remote Sensing: The example of Carcassonne's Defensive Wall (Aude, France)', *Journal of Archaeological Science Reports* 23: 396-405, <https://doi.org/10.1016/j.jasrep.2018.10.030> (published online).
- V. Sciuto Claudia and Dimitris Roubis 'Combining Archaeological Survey and Provenance of Raw Materials by Means of Portable Near Infrared Spectroscopy. The Example of Montescaglioso (Basilicata, Italy)'. *Unsubmitted manuscript*

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Author's contribution

Introductory text

All photos and images have been created by the author except where explicitly mentioned.

Papers

Paper I

JL and PG designed the study. JL PG and CS collected and processed data. JL wrote the manuscript with contributions from all co-authors.

Paper II

CS designed the study, collected the data and wrote the manuscript with comments from all co-authors.

Paper III

CS and JL designed the study. CS, PG and MT statistically modelled the dataset. LLR and CS built and implemented the 3D GIS platform. CS wrote the manuscript with comments from all co-authors.

Paper IV

CS, DA and JL designed the study. DA, CS, NG and AC collected the archaeological/architectural data. NG, AG, RB and SJ collected and pre-processed the hyperspectral images. CS, PG, JL and MT statistically modelled the dataset. CS wrote the manuscript with comments from all co-authors.

Paper V

CS and DR designed the study and collected data. CS wrote the manuscript with contributions from DR.

Abstract

The study of stone artefacts is a combination of anthropological archaeology and geology, rooted in analytical techniques for determining the materials' composition, typological stylistic classification and interpretation of cultural patterns. In this thesis, the archaeology of materials is considered in the context of sites- and landscape transformation, economic history and development of techniques. Focus has been on applications of near infrared spectroscopy (NIR) for characterising minerals in different case studies. Interdisciplinary protocols are implemented in order to account for the various aspects of stone artefacts, merging geochemical investigation and digital documentation.

This thesis consists of two parts: an introductory text and five research publications. In the first paper, a NIR portable probe is tested to measure iron oxide-based pigments in rock paintings in Flatruet (Sweden). The study demonstrates that the probe is useful for characterising different sections of paint *in-situ* and pinpointing similarities and dissimilarities in the pigments used for the figures. The second and third papers are aimed at studying the use of raw materials for tool production in a Mesolithic settlement in Northern Sweden. In the second paper is shown that hyperspectral imaging helps characterise the mineral composition of a selected group of tools and the spectral signature of quartz, quartzite, and flint are examined. In the third paper, hyperspectral imaging-based classification is applied to the entire dataset of lithic tools and flakes collected during excavation of the site. The objects are divided into categories of raw materials according to their spectral features and the distribution is visualised on a 3D GIS platform. The fourth paper deals with the application of hyperspectral imaging, a field probe (MicroNIR) and portable Energy-Dispersive X-Ray Fluorescence (ED-XRF), for in-situ characterisation of building materials on the inner wall of the fortified citadel of Carcassonne (France). The research shows how the combination of these analytical methods in conjunction with a stratigraphic study of the architecture helps to understand the use and re-use of materials in different construction phases. The last paper shows how an in-field NIR-probe may be used in landscape surveys for instant characterisations of different stone types. This study was carried out in the district of Montescaglioso, Southern Italy, to highlight patterns of use and distribution of artefacts made of local calcarenite (limestone) in the period between the 6th and 3rd century BC.

Sammanfattning (Summary in Swedish)

Studier av stenartefakter utgörs av en kombination av ämnen som arkeologi, geologi och kulturanthropologi, ofta genom analystekniker som analyserar materialsammansättning, typologi, stilistisk klassificering och tolkning av kulturmönster. I denna avhandling betraktas stenmaterialens arkeologi i sammanhang som landskapsförändring, ekonomisk historia och teknikutveckling. Ett tvärvetenskapligt arbetssätt används för att belysa olika aspekter av stenartefakter, genom att kombinera geokemiska analyser med digital dokumentation. Tillämpningen av, främst fältbaserad, nära infrarödspektroskopi (NIR) för karakterisering av sten och mineral, testas och utvärderas i olika fallstudier.

Denna avhandling består av två delar: en kappa och fem vetenskapliga artiklar. I den första artikeln undersöks hur en fältbaserad NIR-teknik kan analysera järnoxidbaserade pigment och hållmålningar vid Flatruet (Sverige). Studien visar att tekniken är användbar för att *in situ* karakterisera olika slag av färg och identifiera de olika motivens olikheter i pigmentering. I artikel två och tre studeras användningen av råmaterial för verktygsproduktion i en mesolitisk bosättning i norra Sverige. I artikel två används hyperspektral bildbehandling för att karakterisera mineralsammansättningen i ett urval av stenverktyg och NIR-spektra för kvarts, kvartsit, skiffer och flinta undersöks. I den tredje artikeln tillämpas hyperspektral bildbaserad klassificering på hela datasetet av avslag och stenverktyg som samlats in i samband med utgrävningen av bopplatsen. Föremålen är uppdelade i råmaterialskategorier efter deras spektrala egenskaper och deras rumsliga fördelning över bopplatsen visualiseras på en 3D GIS-plattform. Den fjärde artikeln behandlar tillämpning av hyperspektral bildbehandling, fältbaserade NIR- och röntgenfluorescens (ED-XRF) tekniker, för *in situ* karaktärisering av byggmaterial på den inre ringmuren av Carcassonnes befästning (Frankrike). Undersökningen visar hur kombinationen av dessa analysmetoder tillsammans med en stratigrafisk studie av arkitekturen bidrar till att förstå användningen och återanvändningen av byggnadsmaterial under olika konstruktionsfaser. I den femte artikeln används av en bärbar NIR-instrumentering för att i fält undersöka olika typer av stenmaterial och försöka karakterisera dem. Studien utfördes i distriktet Montescaglioso i södra Italien för att belysa användningsmönster och distribution av artefakter gjorda av lokal calcarenit (kalksten) under perioden mellan sjätte och tredje århundradet f.Kr.

1 Introduction

*'Look upon our condition, crumbling castle.
You would not believe where I'm from...'*¹

In her talk² at the WORLD.MINDS annual symposium in 2017, Prof. Nicola Spaldin, a pioneer in research on multiferroics, argued that the development of civilizations is linked to progress in materials science. According to her, after passing through the Stone and Metal Ages we are now living in societies dominated by silicon, which forms the core of all the digital devices we use to communicate and transfer information.

Her argument is certainly appealing and raises a few questions about materials and their role in the development of human societies. Silicon is so important for small components in digital devices and it happens to be the second most abundant element in the earth's crust, after oxygen. With 90% of the earth's rocks and soils made of silicate minerals, silicon in its various forms has always dominated our environment and our civilizations. Humans never got beyond the Stone Age.

The deep interaction between human societies and geological resources is quite evident when we examine our daily environments. We live in buildings made of stone, travel to work on roads made of stone and even brush our teeth with toothpaste that contains ground stone³. The transformation of materials from the earth's crust, with quarrying and transportation of large volumes of rock, contributed, over the centuries, to the development of new landscapes characterised by carved spaces of extraction and new skylines of architecture. The process of modelling geological deposits started a long time ago and can be traced back to the dawn of civilization. The long alliance with stone, instituted through the creation of prehistoric tools, appears to be a characteristic of our entire history (Foley and Lahr 2015; Waelkens, Herz, and Moens 1992). Rocks are and have always been a constant

¹ King Gizzard and the Lizard Wizard. Crumbling castle. *Polygondwanaland*, 2017.

²

https://www.youtube.com/watch?v=YdKO3CZ_0VM&feature=youtu.be&fbclid=IwAR0E9lNcwqUoNsqeEZMBgJPR_ZPzlAUXdcdzLZcSWznDXWuPcLF7hqxW92g

³ Ground calcium carbonate is used as an abrasive in many brands of toothpaste.

presence in humans' existence, as a part of our lives, in varying degrees: from the personal sphere (such as jewellery) to architecture and landscapes, they are "*linked to power, wealth and both local and global inequality*" (Boivin and Owoc 2004, 1). Traces of this long interrelation can be documented in artefacts of all types and sizes, from all chronologies throughout the globe: from knapped tools to Roman funerary monuments, from rock-cut settlements to Gothic cathedrals.

Archaeological records are extremely rich in their evidence of stoneworking products, as many of the other signs of past lives (such as organic materials or oral traditions) decay and are lost. A close look at these artefacts, identifying materials, techniques of manufacture and transformation of rocks over time, could help trace the modes of interaction between human communities and geological resources. This approach would help understand both the role humans played in shaping bedrocks and the function of geological materials in fostering cultural processes.

An accurate reconstruction of the part that stones played in humans' history requires a change of mind-set, an approach to material culture that focuses simultaneously on the materials' physical qualities, trade/availability and technical knowledge required for production. Since the Industrial Revolution and with the advent of capitalism and consumerism, materials are seen as consumable resources to be exploited for large-scale production, resulting in objects being used and then thrown away. In order to understand the importance of mineral materials in the development of our civilizations it is necessary to rethink the idea of materials themselves (Smith and Wobst 2004).

Artefacts made of stone constitute the intersection between geology and civilization, matters in which the dualism culture/nature is overthrown by a new type of subject. The study of stone artefacts entails the methodological and theoretical challenges of bridging branches of information that are apparently distant, as well as cultural patterns and scientific knowledge. Interactions of human and non-human actors can be traced on different chronological and spatial scales using intertwined theoretical bodies and data-collection methods.

In this thesis the words *object* and *artefact* are used to indicate geological bodies that have been altered, directly or indirectly, through their encounter with human beings. Thus, the terms are applied in reference to things that are the result of the co-occurrence of different factors, things that are constructed by entangled circumstances in shape and material. Social practices, in conjunction with material and physical agents, are intertwined in a stone artefact the moment we observe it (Pétursdóttir 2013).



Figure 1. Carcassonne, view of the courtine wall and tower number 24. Dominique Allios during a data collection campaign in spring 2018.

Materials carry traces that testify to interactions. Some of these attributes can be visible to the naked eye, for example, an object's colour or shape.

A large monument, such as the citadel of Carcassonne (Aude, France), comprises different pieces of masonry resulting from processes of construction and destruction over time (Fig. 1). Different sections of masonry are constructed using distinct materials and techniques that can be documented, to a certain extent, through visual observation. Intangible attributes, such as the cultural role that the artefact assumed in a certain historical period, can be inferred from the context. The uppermost portion of the tower in Fig. 1 was reconstructed in the XIX-XX century, in accordance with Viollet-le-Duc's project, imitating the ancient pieces of masonry that were still visible on the monument. In this instance, the materials can be optically recognised according to their style, distinguishing them from the masonry below. However, only by knowing Viollet-le-Duc's theory of architecture can we comprehend that the restoration was carried out to suit a clear conceptual plan of the designer (Hearn 1990; Viollet-le-Duc 1881).

This traditional dualism between the tangible and intangible qualities of material culture appears to omit a third aspect: certain attributes of artefacts are not visible but are pertinent to the materials' characteristics, such as molecular structure or elemental composition. The geochemical definition of the stone type is intangible and yet is fundamental to defining the object and its qualities. Analytical tools could be beneficial in deciphering some of this evidence and shedding light on information otherwise unseen (Sciuto, forthcoming). The stone types used for Viollet-le-Duc's restoration have a mineral composition that clearly distinguishes them from the other materials that can be found in the structure.

The archaeology of stones should be established as interdisciplinary practice, mapping the interactions between raw materials, manufacturers, communities and environments, in which artefacts are to be considered products of social relations, symbolization, physical interactions with the environment and subsistence. This theoretical framework supports the formulation of research questions that are comprehensive of different issues and require the application of diversified methods of data collection. Epistemological issues influence the choice of tools for the collection of empirical data and sampling strategies, proving how archaeological research cannot be separated from interdisciplinary practice.

1.1 Research questions

This research has been carried out as part of the Mobima project (Mobile Imaging in Archaeology). Mobima is a two-year project conducted at the MAL - Environmental Archaeology Laboratory at the University of Umeå, funded by the

Marcus and Amalia Wallenberg Foundation (grant MAW 2012.0136) and led by Johan Linderholm and Paul Geladi. The project's goal was the testing and developing of the application of near infrared spectroscopy (NIR) on archaeological materials. The aim was to develop a protocol for the application of fast non-destructive image-based techniques on a large scale to field studies and laboratory screening analysis. Within the framework of Mobima, this thesis narrows down the research scope to the investigation of potentialities in the application of NIR-based analysis for provenance studies of geological materials. The study includes various types of artefacts such as painted cliffs, lithic tools, buildings, coffins, millstones and quarries, as well as different kinds of materials such as iron oxide, quartz, quartzite, limestone and sandstone. The reason for such variety is the desire to include a large range of different contexts and related methodological issues in order to evaluate the application of this analytical technique in distinct scenarios.

Looking more closely at the materials it was apparent that an interdisciplinary approach to the study of stone artefacts could help understand the dynamics of exploitation of natural resources and the technical development of ancient societies. It was therefore decided to conduct the study on two parallel and complementary research axes. As technical details of the analytical techniques were elucidated it became evident that the systematic acquisition of spectral data was linked to the evaluation of the role of material studies within archaeology and archaeological science. This work is intended to be an attempt to overcome the idea that lithic materials are simply symbols or representations of human society or that materials science is merely to be considered the chemical classification of artefacts and geological sources. In order to achieve this, a combination of methods on the cutting edge of natural science and humanities has been explored, arguing that the application of a NIR-based analytical instrument could represent an interesting novelty for archaeometric research. The aim of this methodological endeavour is to find effective tools for recording analytical data in different contexts: from a controlled laboratory environment to large standing structures and surveys of exposed outcrops. The development of these methods is directed towards testing protocols for collecting information on materials that can be fully integrated into archaeological practice rather than being considered an accessory study. A more efficient assimilation of materials science into archaeological research will help procure increasingly more information about the agency of stone in shaping our past and our future.

The study of materials in this respect is approached from an environmental-archaeological perspective, attempting to look at stone artefacts as ecological agents.

Stone artefacts are used as a proxy to understanding transformations of landscapes and human societies, adaptation and the use of geological resources. Moreover, the artefacts are always considered in context, looking at the objects' interactions and exchanges with their environment. The relationship between humans and rocks is analysed in this context from a diachronic perspective, as a mutual influence, evolving over time and geography.

Finally, a new definition of materials science and a new protocol for documenting material evidence is discussed relative to its epistemological implications. The study of stone artefacts is examined by referring to new materialistic positions and the general theoretical debate on the separation between cultural archaeology and archaeological sciences (Boivin 2005; Hodder 2012; Jones 2004; Pétursdóttir 2013; Ruggles and Silverman 2009; Tilley 2004).

1.2 Structure and choice of case studies

The choice of case studies is the result of a selection based on several criteria. The first scope is undoubtedly the development and testing of the application of NIR spectroscopy in a range of different case studies to evaluate the potential of this technique in disparate contexts. The sites chosen for the study present quite different environmental setups, being situated in landscapes characterised by distinct geological and geomorphological features. The chronology and cultural context are also non-homogeneous, ranging from isolated prehistoric dwellings to protohistoric production sites and medieval citadels (chronologies, contexts and types of material are summarised in Table 1). All this variability should not be regarded as a lack of organic research planning, but rather as a diversification of the frameworks that aim to include a large spectrum of materials and contexts. In compiling a diverse set of case studies in which NIR spectroscopy served as a tool for characterising stone artefacts, the effectiveness of the analytical technique was demonstrated, highlighting its advantages and limitations. Moreover, in all of the examined cases, the archaeological study of stone artefacts was achieved by integrating different methods of recording and visualising material properties. The characterisation of the mineral configuration of materials was associated with the typological classification of the items and was discussed according to the context. The scale of the case studies also plays an important role in the organisation of the work. The papers have been organised according to the extent of the investigated areas in order to compare the performance of NIR spectroscopy with different levels of detail and different scales of datasets. A single panel rock painting has been analysed in the first paper while an entire dwelling site is the object of the second and third papers. A citadel has been studied in the fourth paper concluding with the survey of a small district in the fifth case study. Finally, the choice has also been based on practical and strategic reasons

regarding collaboration and ongoing projects involving Umeå University and other institutions. The first three papers are based on Swedish research projects, while the fourth paper is the result of a long-term collaboration with French institutions: the University of Rennes 2, the IRSTEA (National Research Institute of Science and Technology for Environment and Agriculture) in Montpellier and the Amicale Laïque de Carcassonne. The fifth and final paper is part of the CHORA project (CHORus of Resources for Archaeology) hosted by the Graduate School in Archaeology in Matera (Italy). Fig. 2 shows the geographic location of the case studies, in *italic* are the other sites mentioned as examples in the text.

Table 1. Sites, chronology and context of the case studies included in this thesis.

Site/Paper	Location	Chronology	Context	Type of materials
FLATRUET Paper I	Härjedalen, Sweden. Rock cliff, approx. 800 m.a.s.l.	2000 BC, Neolithic	Animal and human figures painted in red on a cliff. Exact chronology and phases of the painting are still unknown.	Earth-based pigment on rock surface. Iron oxides and metamorphic sandstone
LILLSJÖN (RAÄ 260) Papers II and III	Ångermanland Sweden. Lake/river bank.	7200-5800 BP, Mid-Late Mesolithic	Semi-subterranean dwelling. An early settlements in Northern Sweden established after deglaciation.	Lithic tools, quartz and quartzite
CARCASSONNE Paper IV	Aude, France. Hill, alluvial plain.	1 st century BC- 19 th century	Fortified citadel inhabited since the 1 st century BC. The defensive walls and the urban configuration underwent several phases of reconstruction	Building materials. Sandstone masonry.
MONTE-SCAGLIOSO Paper V	Basilicata, Italy. Riverbank/hill tops.	6 th -3 rd century BC	Indigenous settlements and workshops for production of calcarenite artefacts. Manufacture and circulation of objects.	Coffins, building materials and objects used for food production. Calcarenite (limestone)



Figure 2. Location of the case studies. Flatruet and Lillsjön (RAÄ 260) in Sweden are the subjects of the first, second and third paper; Carcassonne (Aude, France) is the case study discussed in the fourth paper while the research described in the fifth paper was carried out in the territory of Montescaglioso (Basilicata, Italy).

In italic are indicated the other sites mentioned in the text.

2 Archaeology and stones

The use of bedrock to produce artefacts is a practice that has been widespread throughout the entire history of humanity. Different materials have been processed using various techniques in all contexts of human settlements. Over time, rocky surfaces have been used in multiple ways and transformed on several occasions. As a consequence, the archaeological evidence of interaction between humans and stones takes many shapes. From obsidian nodules exploited for prehistoric tools to white marble used for renaissance sculptures, many geological facies have crossed the human path.

A general archaeological approach to this multitude of evidence naturally presents some theoretical and methodological challenges. The archaeology of stones is scattered among a distinct tradition of studies, characterised by geography, chronology and cultural context. Typological knowledge of stone artefacts has achieved a high degree of accuracy due to the extensive history of studies within the various branches of archaeology. The details of how typological studies of stone artefact have developed is outside the scope of this thesis. The aim of this work is to transcend the boundaries within the discipline and investigate common practices in the study of materials. The accent here has been moved from the morphology to the substance of artefacts, considering how information regarding materials is gathered and used in archaeological contexts. The goal is broad and a doctoral thesis is not sufficient to investigate the problem in detail. Rather, I wish to direct the reader's attention to materials studies, considering the theoretical and methodological issues involved.

All sorts of stone types are accessible these days and quite easy to find in Western countries. Our economic system and technical developments enable the extraction and transportation of materials from virtually every location on the planet. The choice of a specific material is mainly determined by design trends and economical status: the rich owner of a villa will be likely to choose marble tiles for garden paving while a landlord of more modest means will have his home pavement made of concrete slabs. Patterns of supply and the use of materials in the past can also be traced, taking into account a number of environmental and cultural factors.

The availability of materials and the possibility of transporting them is a determinant aspect for understanding their use in the past. For the decorations in the Palace of Versailles, Louis XIV could afford to transport red marble columns to Ile-de-France. These columns had been quarried in the Aude region (Southern France). In this instance, the presence of a good road network and the economic power of the French monarchy enabled the long-distance transportation of prestigious materials,

the marbles of Caune-Minervois (Fig. 2), to be used in the spectacular architectural project (Bonnet 2012). Provenience studies on stones across the Mediterranean basin from the Bronze Age to Late Antiquity have been conducted on a large scale focusing on status-related products such as marble and demonstrating how luxury materials can travel huge distances (Gorgoni et al. 2002; Lazzarini 2004; Waelkens, Herz, and Moens 1992).

Different circumstances, outlined in Paper III, show how the Mid and Late Mesolithic period in Northern Sweden was characterised by the adoption of local materials, such as quartz and quartzite, for tool making, while flint is an allochthonous material and was primarily used during the Early Mesolithic phase, corresponding to the early colonization of Northern Scandinavia after deglaciation (Forsberg and Knutsson 1995; Olofsson and Rodushkin 2011). In Mesolithic Sweden, materials with a good degree of workability were not available and had to be carried from remote southern and eastern regions. Only a small number of flint tools have been found in dwelling contexts and the number of artefacts decreases in subsequent chronological phases. As a consequence of adaptation, the lithic technology in quartz and quartzite was developed as an alternative to flint.

Technical knowledge and the technological progress of communities are also important factors to consider when studying methods of quarrying and manufacturing stone artefacts. For example, in the Romanesque Church of Saint-Nectaire in the Puy-de-Dôme (Auvergne, France; Fig. 2), the external portion of the double-sided walls and the foundations are built using trachyte and trachybasalt, volcanic rocks that are quite hard to cut and which are dense and heavy. The vault masonry and the rubble filling inside the walls are made of rhyolitic pumice, which is porous, light, easy to cut and transport. This architectural stratagem resulted in a substantial lightening of the structure, which could be supported in the nave by columns instead of pillars (common in Romanesque churches, Allios 2016). Medieval builders reduced the effort of working the harder and heavier materials to a minimum, preferring a rational use of the available rocks and an accurate calculation of weight and volumes, allowing them to create a structure unique in its genre, in which the pillars recall the interior of St. Peter's Basilica in Rome.

Cultural preference is another important variable to take into account when studying materials. Certain stones are precious or regarded as having magical properties and are therefore considered to possess great value, for example, Alpine jades in Neolithic Europe, transported thousands of kilometres from their original outcrops (in the Western Italian Alps; Fig. 2). Bright green polished axes can be found all over Central Europe, dated from around the second half of the 5th and beginning of the

4th millennium BC and have often been interpreted as individual status markers (Pétrequin et al. 2012, 2017). All these different factors are involved in the choice of materials in various contexts. Thus, rock types and their stories can help to shed light on the socio-economical context in which the artefact was produced. The study of materials is conducted through the characterisation of their geological facies, chemical/mineralogical composition and eventual alteration of the original matter. Such information is usually gathered by observing the microscopic features of the rock or using chemical analytical procedures. Each method provides a different set of information and requires a distinctive sampling strategy (for an extensive discussion of analytical methods and sampling strategies, see Chapter 4). A satisfactory set of information about materials and their use cannot be gathered without good knowledge of the informative potential embedded in the substances of artefacts. Considerable differences exist in the way that the study of stones has been undertaken within different traditions of study. Scientific practice and cultural interpretations have not always been free of problems and/or the risk of making deterministic assumptions. In the following paragraphs, I illustrate how the characterisation of rocky materials is approached in distinct archaeological sub-disciplines.

2.1 Quarries

A study of materials that encompasses stages of manufacture, distribution and use should take into account extraction and production sites. Quarries are where the first stages of bedrock transformation occur, where the stone is extracted and shaped according to the characteristics of the material and production requirements. Quarries are where empirical human knowledge of the geological substratum is perfected and labour techniques are adapted to the rock's characteristics. Techniques for extracting blocks and managing production waste are adjusted to the geology of bedrocks and the degree of professionalism of the workers. Different extraction techniques are adopted according to the morphology of the outcrop, the resistance of the materials and the transport infrastructures. Several quarrying strategies have been documented at archaeological sites and in ethnoarchaeological comparisons. Outdoor exploitation includes quarries on ledges or shelves, linear quarries, vertical quarries and quarries in recesses (or funnels). Subterranean quarrying can be carried out in wells, caves, “stone-walling and back-filling” of tunnels, or with pillars left to support the quarry ceiling. These types of extraction methods leave macroscopic traces that are still recognisable today – persistent markers in the landscape (Fig. 3).

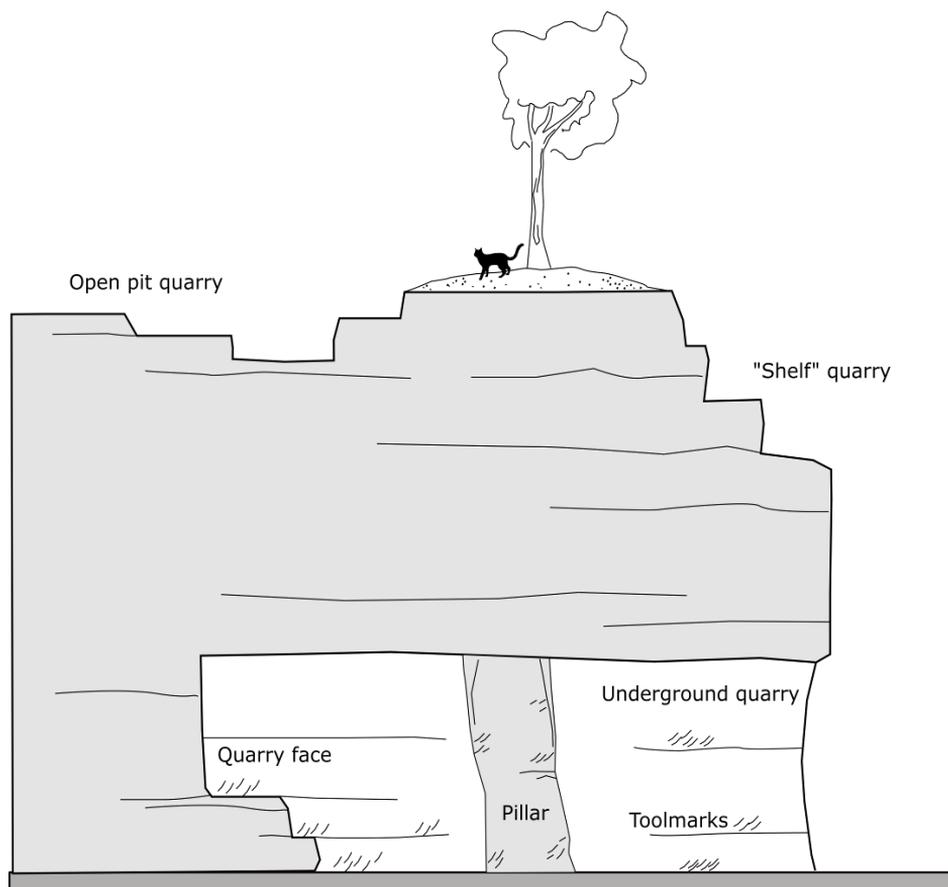


Figure 3. Different types of quarry. Modified from Adam 1988.

Archaeological interest in material extraction techniques started at the end of the 19th century, triggered by the number of ancient inscriptions that could be found at a number of production sites (Bruzza 1884; Hirt 2010). The archaeology of quarry sites, involving stratigraphic investigation of archaeological records, has been developed over the last 60 years because of a renewed interest in production sites (Dworakowska 1983; Waelkens, Herz, and Moens 1992; Ward-Perkins 1972). We are now able to depend on a good reference record of excavated and published quarry sites all over Europe, especially in France. In a way, French scholars have been pioneers in the study of quarry sites with the work of Jean-Claude Bessac (Bessac 1986, 1996, 1991, 2007; Massih and Bessac 2009) and specific research centres dedicated to the study of stoneworking techniques (for example, a section of the LAMOP- Laboratoire de médiévisitique occidentale de Paris, at the University Paris 1 Pantheon-Sorbonne).

Large volumes of stone for various purposes might not originate in large extraction sites, like the ones described above, but rather from more a sporadic exploitation scattered over a wide area, such as in the case of the trachyte and trachybasalt used for the construction of the already-mentioned Romanesque Church of Saint-Nectaire (Allios 2016; see map Fig. 2), where traces of extractions have been observed over a large section of erratic blocks of trachyte and trachybasalt (Fig. 4). The characteristics of this type of extraction are such that the exploitation has been carried out on several boulders or outcrops simultaneously, gathering a relatively small quantity of material from each intervention. This type of supply system requires a high level of geological expertise and only highly-skilled workers were able to survey and recognise good raw material.



Figure 4. Traces of extractions on erratic block in Treizanches, Puy-de-Dôme (Auvergne, France). The stone quarried has been used for building the Church of Saint Nectaire.

A similar process can be observed in Mesolithic Northern Scandinavia, where sources of raw materials for tools, quartz and quartzite, could be found scattered in the landscape, as pebbles in moraine deposits or nodules in bedrocks (see Paper III). The phenomenon of lithic procurement in large areas has been investigated in a Scandinavian context and shows how the practice of gathering raw materials might have shaped social structures (Nyland 2017a, 2017b).

Finally, in several cases we are able to recognise an artefact's stone type even though the quarry remains unknown. Such circumstances are recurrent due to the large-scale transformation of landscapes, particularly over the last couple of centuries, as a consequence of industrialization. Most of the rocky materials considered valuable in the ancient world have subsequently been recognised as profitable. The most famous example is the marble quarry basin in the Apuan Alps near Carrara (Fig. 2). Quarried since the Roman Period, the fine white marble has been heavily exploited over the centuries to the point that the configuration of the mountains themselves has completely changed. As a consequence of the hugely prolonged extraction activities, almost all traces of ancient production sites have been erased and it is difficult these days to delineate the evolution of the activity over time (Dolci 1995; Dolci and Nistri 1980; Paribeni and Segenni 2015).

Similar is the case of Carcassonne (as described in Paper IV), where the different building materials are distinguishable in the architecture and can be associated with different construction campaigns. Nevertheless, the origin of the materials is unknown because of the recent extensive urbanization of the Aude plain and the destruction of most ancient quarry sites.

In a sense, the importance of a quarry can be established without locating the archaeological site itself but rather observing the materials extracted, their volume and morphology. A detailed examination of a workshop's products discloses information about the organisation of labour and specialisation of the manufacturing process, even if the original site was missing.

Since the 1980s, the study of quarry sites has been deeply influenced by the introduction of scientific practice in the study of materials. This approach engendered a better understanding of the microscopic qualities of materials, creating solid links between sources and products. However, the hard scientific approach to the study of quarry sites has sometimes resulted in a biased, slightly deterministic vision of the production stages that does not take into account the empirical experience of quarrymen and their work. In Paper V, an effort has been made to merge the spectroscopic analytical investigation of a quarry and a few stone artefacts with a typological study of the artefacts and the detection of technological marks that could provide information about the stages of production.

2.2 Painted cliffs

In Scandinavia, rock paintings and carvings are characteristic landscape markers that play an important role in the popular imagination. While the study of quarries and the production of stone artefacts is focused on the transformation of materials and movements (transport/trading) the archaeology of painted stones could seem a

rather static subject, dealing with figures depicted on an immobile surface. However, Scandinavian rock art testifies to a broad network of movements of people, goods, behaviour and technical knowledge, being situated on important roads or meeting places.

The iconography of Scandinavian rock art has been studied extensively and scholars have adopted different approaches to tackling the problem of identification and attribution of complex panels or isolated figures. Publications on the subject are copious, dealing with contrasting interpretative hypotheses about the practice of rock art in prehistoric society. In this context, scientific analysis is primarily used to date the panels (Bednarik 2002), or in planning restoration and preservation work (Bakkevig 2004; Tratebas, Cerveny, and Dorn 2013; Watchman 1990). A detailed characterisation of pigments is rarely carried out as it often requires the application of destructive methods on pigment sampled from the rock surface. Few examples of analyses carried out on paint fragments have highlighted how weathering processes could have altered the composition of certain pigments (Cerveny et al. 2016; Ramanaidou et al. 2008; Rogerio-Candelera 2016; Vázquez et al. 2008). The characterisation of pigments directed towards the description of phases of painting and the configuration of the panels has been conducted in situ using non-destructive techniques such as X-Ray fluorescence spectrometry (XRF) or Raman spectroscopy (Chalmin, Menu, and Vignaud 2003; Gay et al. 2016; Gomes et al. 2013; Newman and Loendorf 2005; Olivares et al. 2013; Ospitali, Smith, and Lorblanchet 2006; Prinsloo et al. 2008). Near infrared spectroscopy can be applied as an imaging technique to highlight figures otherwise not visible due to rock surface weathering (Liang 2012). However, portable spectrometers are also useful for performing measurements directly on single sections of paintings. Paper I, included in this collection, is a pilot study of pigment composition using a portable NIR probe on a panel in Northern Sweden. In this instance, NIR measurements are not appropriate for gathering quantified elemental information but rather to highlight similarities and dissimilarities between figures on the same panel. The study also focused on the rock surface and the influence the support could have on the spectral signal. In a way, this is an attempt to understand how painting and cliff interact: the red ochre might be partially washed away, absorbed in the rock's pores or react with the cliff's surface, creating new materials. The technique adopted is portable and could be used in other case studies to include the characterisation of pigments in traditional stylistic studies of painted pictures.

2.3 Stone tools

The characterisation of raw materials plays a crucial role in the study of lithic tools. The supply of materials is the first fundamental stage in the *chaîne opératoire* (Leroi-Gourhan 1964b) and often represents the only proxy for deciphering movements, trading and exchanges of prehistoric people. Analytical data are fundamental to revealing information about crafting stages and are crucial to characterising the provenance of raw materials. Countless papers, monographies and edited volumes have been published on the subject, including studies of specific materials or geochemical methods (see, for example, the debate on the use of XRF published in the *Journal of Archaeological Science*, Frahm 2013; Speakman and Shackley 2013). Most of these studies aim to acquire detailed geochemical characterisations of lithics, sometimes at the cost of neglecting the bigger picture. It is noteworthy that the most interesting results regarding the socio-cultural meaning and usage patterns of certain materials are achieved through routine analysis applied on a large number of items, generating conspicuous datasets that can be modelled to highlight the movements and distribution of tools and techniques (Pétrequin et al. 2012; Tykot 2004). A NIR-based fingerprint approach to raw material classification is particularly useful when dealing with large datasets. The use of NIR imaging on large lithic datasets can contribute to determining the categories of raw materials even when their sources are unknown. This is the case for the Mesolithic dwelling site analysed in Papers II and III where hyperspectral imaging has been used for classifying the raw materials of more than 2,600 items.

Other stone tools have also been studied in order to understand long-distance trading routes. Millstones represent a particular case. As they are primarily made of very hard stones, such as metamorphic or igneous rocks, they are frequently produced at large quarry sites and exported over long distances. The phenomenon was well known in the Roman and pre-Roman period in Italy and Central Europe (Gluhak and Hofmeister 2011; Renzulli et al. 2002).

2.4 Coffins

Stone coffins, in various shapes and materials, have been part of funerary practice for millennia. Their use is part of a ritual form that involves the deposition of one or more bodies in a sarcophagus, literally, *flesh-eating*. The Greek definition of stone coffins as sarcophagi is probably due to the fact that the stone boxes would allow the circulation of air and microorganism-activating taphonomic processes. Bodily fluids would be consumed or leak out through fissures and pores in the rock, while bones would remain in the coffin. Burials in stone sarcophagi were quite common

throughout Europe in various chronological contexts. Coffin production appears to be quite specialised and it has been frequently observed that some quarries were adapted for the extraction of large blocks for sarcophagi (see Paper V; Boudartchouk 2002; Büttner and Henrion 2009; Lorenz and Lorenz 1983; Morleghem 2010). Coffins are generally carved in soft stone that is easy to work and carry, usually limestone (or marble) or porous volcanic rocks. The degree of finishing and decoration of stone coffins depends on the style, the status of the buyer and the skills of the craftsmen. Most of the time, coffins were partially carved near the quarry to lighten the large blocks but still avoid making them too fragile for transportation.

2.5 Building materials

The approach to production and distribution of stone as a building material in historical archaeology is linked to economic history. In the last twenty years, scholars have assimilated the socio-economic and technological aspects of the archaeology of quarries and masonry. The history of studies in this field varies from country to country. The Italian tradition of building archaeology is rooted in the typological and stylistic analysis of the construction (Bianchi 2004; Boato 2008; Brogiolo 1996), in conjunction with a degree of collaboration with archaeometricians (Franzini and Lezzerini 1998; Lezzerini, Antonelli, et al. 2016; Lezzerini, Ramacciotti, et al. 2016). The French approach to the subject is different, focusing instead on the importance of raw materials and human-environment interaction. Particularly relevant are the proceedings of the CTHS conference (Congrès National des Sociétés Historiques et Scientifiques) published in 2008 and 2011 (Blary, Gély, and Lorenz 2008; Gély and Lorenz 2011). These volumes bring together case studies in which the analysis of building materials starts with the geological characterisation of artefacts integrated with the archaeological analysis of structures and landscapes, focusing on quarries and the transportation of goods. The work of French scholars has been particularly inspiring for this thesis, suggesting the idea that the study of stone masonry ranges from socio-economic history, history of technology to geological exploration. Moreover, sound knowledge of materials is crucial for understanding a building site, its cycles and dynamics (Edensor 2011). Labour is organised around different types of stone and the specific technical requirements of the materials. The schedule for mixing of mortars, supplying stone and making the sculpture has to be regulated in order to streamline the workflow and achieve a satisfactory result.

3 Between social archaeology and archaeometry: a transdisciplinary struggle

The application of scientific protocols to the characterisation of material culture has often been considered a subsidiary discipline, with procedures carried out by specialists, independent of the main body of theoretical archaeological reflection. However, one cannot have scientific practice without theoretical thinking and agnosticism towards archaeological theory simply does not exist (contrary to what is argued by Martínón-Torres and Killick 2015).

The choice of which scientific analytical methods and sampling strategies to apply in a certain context is based on a theoretical assumption, whether or not researchers are willing to acknowledge it.

Although the geochemical characterisation of stone artefacts has become almost a routine practice over the last 60 years (especially for certain chronological periods and cultural contexts, as outlined in Chapter 2), the development of scientific applications has not always been grounded in a theoretical reflection on the implications of this practice for the interpretation of archaeological records.

The archaeological sub-discipline called archaeometry, comprehensive of all scientific approaches used in archaeological research, was conceptualised in the late 1950s. Before this time, the chemical analysis of archaeological materials was already being performed in different contexts and the emergence of an interest in the composition of ancient objects can be traced back to the end of the 18th century (Caley 1951; Pollard 2013). Early studies into the chemistry of materials were mostly driven by curiosity rather than being associated with broader archaeological research projects (Chaptal 1809; Klaproth 1798; Pearson 1796). From the second half of the 19th century, scientists started appreciating the historical value of the compositional analysis of ancient artefacts. During this period, analytical appendices were published that were associated with reports of archaeological excavations (Goebel 1842; Layard 1853; Schliemann 1880). Most of these early chemical investigations were conducted on ancient metals with very few studies dedicated to other materials. The first study of lithic artefacts was carried out by Damour (Damour 1865). For the first time, the scholar compared the geological characteristics of ancient objects and outcrops, attempting to link resources, population movement and manufacture.

During the second half of the 20th century, the application of archaeological sciences developed and expanded into all types of contexts and chronological frameworks. The *Journal of Archaeometry* (from the Research Laboratory for Archaeology and History of Art at Oxford University) was founded in 1958 as a research bulletin and

acquired increasing importance for an international scientific audience. The ISA (International Symposium on Archaeometry) started in 1961 as a meeting on geophysical prospections and was repeated in subsequent years, becoming the largest international meeting of archaeological scientists (Pollard 2012). The journal of Archaeological Science (JAS), published by Elsevier, was established in 1974 and discusses advances in the application of scientific techniques to archaeological research. JAS is currently one of the most important international archaeological journals. Other journals have explicit interdisciplinary goals and leave increasingly more space to contributions that include a scientific approach to the analysis of archaeological contexts. Similar is the case of major international archaeological conferences at which increasingly more sessions are organised around scientific topics.

If the integration of scientific methods within archaeological practice appears to be expanding (Killick 2015), the development of archaeological education in scientific topics still leaves much to be desired. The rapid advancement of scientific research is not supported by the adaptation of university programmes. A consequence is that new and old generations of archaeologists struggle to keep updated with the latest methods. As a result, the gap between archaeologists and applied scientists has widened, leaving few opportunities for the maturation of a theoretical body and a consistent toolbox for interdisciplinary practice. While the study of materials is developing significantly, it is also moving away from the interpretation of cultural phenomena, to the point at which the extreme specialisation of analytical procedures barely even deals with problems related to representative sampling and the contextualisation of results (an exhaustive analysis of this phenomenon can be found in Killick 2015). In this scenario, the pursuit of a concrete interdisciplinary profile in archaeology and archaeological science is difficult, while there is an increasing risk of promoting two different sorts of archaeology: a social archaeology, which deals with cultural phenomena and a scientific archaeology, focused on the modelling of quantitative data collected through analytical procedures.

In the late 1950s and 1960s, as the application of scientific techniques to the analysis of archaeological materials was being established, archaeologists were discussing the rise of a new archaeological paradigm. The theorisation of *new archaeology* or *processual archaeology* certainly influenced the early development of archaeometry. In fact, the new archaeologists promoted a more scientific approach to archaeology and material culture, applying routine protocols to the investigation of past phenomena and ethnographic records (Binford 1962; Binford 1965; Binford 1978; Schiffer 1975). This theoretical input fostered the development of collaborations between

archaeologists and materials scientists that resulted, in some cases, in the production of important publications (including Schiffer 1987, which was decisive for my own education as a student) and in other cases to the circulation of studies that focused on scientific practice and which were not totally relevant to questions concerning archaeological and historical research. For processual archaeologists, the study of materials was important as a repeatable practice, capable of supplying objective data that could be used to infer general patterns of past human behaviour. As a reaction to the neo-positivist perspective of the processualists, the paradigm theorised in the United Kingdom during the late 1970s and 1980s and called *post-processualism* endorsed a different approach to archaeological science. The new movement was rooted in the theories of philosophers of science emphasising the social construction of scientific practice and facts (for example, Latour and Woolgar 1979), drawing attention to the subjective character of archaeological interpretations. Post-processualists emphasised the symbolical and ideological value of artefacts, promoting a phenomenological approach to materials (Cummings and Whittle 2004; Hodder 1982, 1988; Shanks and Tilley 1987; Tilley 2004). During the 1980s, *cognitive archaeology* was introduced to indicate a theoretical model to infer behavioural patterns of past human societies from material remains and scientific practice, which only provide data that is valuable for interpreting the past (Mithen 1996; Whitley 1992). Cognitive archaeologists challenged the post-processualist paradigm and the use of textual metaphors referred to as material culture (Hodder 1988; Tilley 1991) highlighting how material culture should be interpreted not as representation but rather as embodiment.

Over the last twenty years, researchers have started inquiring into the role of archaeological material from a new perspective, questioning the agency of raw materials and natural resources in shaping ancient societies (Boivin 2008; Jones 2004; Shackley 2005). The implications of an interdisciplinary approach in archaeology have been researched, referring to the new materialistic theories (Boivin 2008; Boivin, and Owoc 2004; Hodder 2012; Jones 2001, 2004; Joyce 2012; Joyce et al. 2015), suggesting that an integrated interdisciplinary approach to the study of archaeological materials could help break down some conventional distinctions between specialisations within the discipline (Boivin, and Owoc 2004). These archaeologists, with a broad understanding of the methods of archaeological science, highlighted how humans and materials are mutually constituted by their interactions and transformations over time. Within this perspective, the material world does not necessarily symbolize anything else (Boivin 2008) but is a player in the development of past societies: “*human action and the materials through which humans act cannot be divided*” (Jones 2004, 335).

The study of technology also fulfils an important role in understanding the relationships of human societies and materials. The analysis of technological choice and material/cultural constraints was started by the archaeologist-anthropologist André Leroi-Gourhan in his book *L'Homme et la Matière* (Leroi-Gourhan 1964a) in which he analyses several *moyen élémentaires* (elementary means), simple gestures (such as pulling/pushing/hitting) that people in different geographic, chronological and cultural context have used in order to interact with raw materials. Through his classification, Leroi-Gourhan was able to recognise some general patterns of “technological choices”, a theme also discussed by Sillar and Tite (2000), considering the example of pottery making. As they argue, an artefact is the result of the interaction between social factors (such as status of the maker and the end user), economy and trading, mechanical performance and availability of materials (some of these factors have been discussed in Chapter 2).

According to Jones, these factors could be incorporated into the concept of materiality, defined as: “*a notion that encompasses the view that material and physical components of the environmental and the social practices enacted in that environment are mutually reinforcing*” (Jones 2004, 330). Focusing on an object’s materiality, the artefact no longer becomes a clue to be decoded but an essential component of the past characterised by material properties that must be explored in detail (Allios 2004; Boivin 2005; Jones 2004).

Stones certainly have a symbolic role in many cultures and play an important part in various rituals. This has been documented in several ethnographic and archaeological research works (see paragraph 3.3 in this chapter; Boivin 2008; Boivin and Owoc 2004; Taçon 2004). Less importance has been given, for instance, to the material agency of stones, described as the ability of rocky materials to influence the development of cultural phenomena. An example of the material agency of stones and mountains is the case of the marble industry in the previously mentioned area of Carrara, in Tuscany (Italy; Fig. 2). In the Apuan Alps, the presence of high-quality white marble geological formations has probably motivated the beginning of the exploitation. Nevertheless, a local community has developed around the extraction and production of marble artefacts, establishing a vital connection with the mountains. Generations of quarrymen have lived with the marble, in houses built of the same stone they carved every day. In this instance, the environmental conditions (presence of good materials) favoured the establishment of the settlement although the multi-layered associations created by the stone (even used for food preparation and a whole range of daily activities) developed over time, slowly shaping the social structure, economic system and community behaviour. Materials helped shaping past cultures (and still do). Thus, their selection, transportation and transformation

are key concepts in revealing patterns of human behaviour and socio-economic trends. Considering this information potential, it seems that archaeological practice should be more engaged in material analysis. Materials study must not be confined to the role of an auxiliary discipline. On the contrary, it should be fully included in all archaeological investigations in order to unlock the information potential contained in mineral configurations. The socio-cultural environment and a geological description are equally necessary in order to describe an artefact. In this sense, cultural interpretation and archaeometric data are necessarily intertwined.

3.1 Material itineraries

Stone artefacts form part of a dense network of exchanges with human societies and should be investigated through an interdisciplinary approach, taking into account both object morphologies and material properties. The artefacts' shape can provide information about the function of materials, while an interpretation of the archaeological context relates to their cultural background. For example, a single stone slab could be interpreted as a coffin cover or a tile depending on its archaeological context; a coffin-covering slab could also be reused as a masonry element in a cathedral, although its previous usage will perhaps be marked by inscriptions on its surface. Markers of production, use and disposal can be documented on an object's surface while analysis of the materials provides information about origin, trading network and stone type preferences. In Fig. 5, the levels of information that can be collected from a stone artefact during different stages of its life are schematised (partly inspired by Sillar and Tite 2000, 6). All these stages and interactions contribute to delineating an itinerary, a composite record of information that characterises the object itself. Every artefact tells a story; from the quarry outcrop to the moment in which we observe it, every object has a trajectory consisting of occurrences comparable to life events. This is the biography of an object (as defined by Kopytoff 1986; Gosden and Marshall 1999), an open narrative, involving past circumstances and potential future developments. The contribution of the science of materials to this itinerary narrative is important and is too often relegated to the role of only finding the origin of the rock.

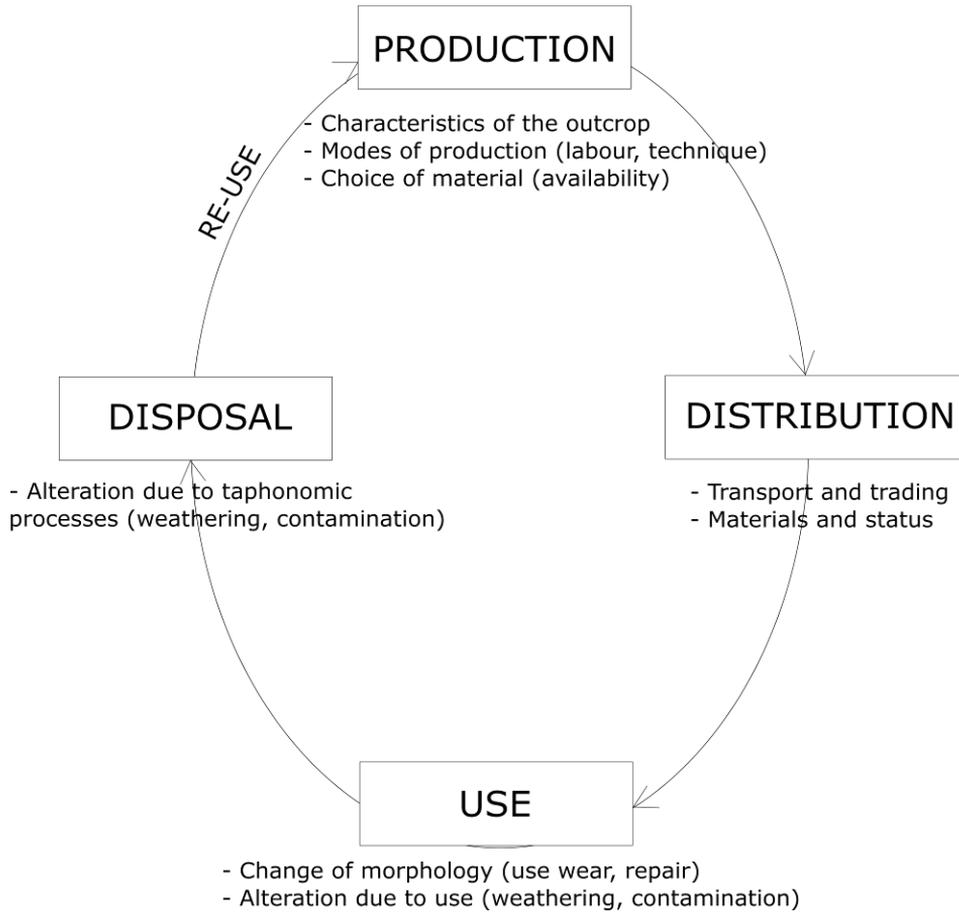


Figure 5. Stages in an object's itinerary showing how materials are chosen, how they move and evolve. Analysis of materials can provide useful information for revealing details about production, distribution, use and disposal.

In archaeology, the analysis of stone types is primarily discussed using a specific set of terms that carry implicit theoretical connotations: sourcing, provenance/provenience. It is quite clear that the concerns of archaeologists and scientists involved in analytical protocols have mainly been to locate the geological deposits from which the artefacts originated. Linking an object to its source is certainly important. However, this represents only one phase in the life of an artefact. The origin and find spot are mainly treated as the beginning and end of an object's trajectory, without saying much about the intermediate stages. The terms *provenance* and *provenience* should be used in reference to distinct concepts rather than as a spelling variation (Boivin 2005; Joyce 2012; Malainey 2010; Price and Burton 2010; Sciuto, forthcoming).

The term **provenience** is used by archaeologists and chemists to principally indicate the place of origin of the raw material from which an artefact is made. *Provenience* is defined by Price and Burton (2010, 214) with a postulate stating that: “*the chemical differences within a single source of material must be less than the differences between two or more sources of that material, if they are to be distinguished*”. In these terms, the study of an artefact’s *provenience* poses only geochemical analytical problems because of the definition of the signature of the materials and sources. The success in the application of this postulate is contingent upon the following: type of materials, geographic distribution of sources, analytical techniques, budget/costs, time, and type of artefacts (Malainey 2010; Price and Burton 2010). The postulate definition is simplistic (a risk that the authors themselves point out) and completely ignores the cultural factors involved in production choice that could influence material output (should we consider large quarry sites including outcrops on two different geological formations, with common extraction procedures, as two distinct sources? And again, shall a quarry that was re-opened after a period of abandonment with different setups and manufacturing procedures be considered a single source?). The variability of materials and modes of production needs to be defined according to the case study and cannot be stated *a priori* without falling into contradiction. The approach defined by Price and Burton, focusing on the methodology rather than the contextualisation of the data, aims to discuss analytical procedures, instruments or the characterisation of specific materials. This heavily geology-driven approach has often been adopted in a prehistoric context and related to the study of stone tools production in which raw materials and artefacts are geochemically linked together to pinpoint networks of trading and movements (Andrefsky 1994; Ericson and Purdy 1984; Gramly 1980; Waelkens, Herz, and Moens 1992).

The term **provenance** appears to concern a broader range of meanings. In art history and museum/archive policies, provenance is the history of the object defined through its successive ownerships (DeAngelis 2006; Drucker 2013). The artefact’s *provenance* is not fully considered to be a property of the object itself but rather linked to possession and purchase. *Provenance* is traced to establish the monetary value of the object rather than its historical significance. In archaeology, *provenance* is also used to indicate the place of origin of an artefact’s raw materials (Malainey 2010) although it appears to be referring to a more general concept, related to the geochemical characterisation but yet open to including socio-cultural factors. As Joyce mentions, *provenance* should refer to the complete itineraries of objects as they are *constituted primarily of raw materials and are continuously altered changing their properties and becoming other things* (Joyce 2012, 124). Thus, it is more appropriate to apply the term *provenance* to biographies of things, as series of transformations and contexts. It also involves the

co-occurrence of physical characteristics and acquired meanings that shape a stone artefact and cause it to become the result of the entangled history of humans and materials.

In conclusion, *provenience* is the analytical chemical and geological characterisation of two objects to determine whether or not they are part of the same formation. It aims to identify a direct link which ties together the quarry and artefact and focuses on analytical issues. Finding the origin of raw materials should not be the ultimate aim of materials study but rather represent one stage of a more comprehensive narrative of *provenance* (as outlined in Fig.5). The geochemical characterisation of stone artefacts has much more to contribute to archaeology than just establishing links. It can be applied to describing longer processes of use and transformation of objects in various environmental and cultural contexts.

3.2 Analysing materials in a changing world

The interaction of past societies with geological resources generated objects that appear solid, static, but that are also in a continuous state of transformation through interactions with the surrounding environment. Viewing ancient stone artefacts as standing totems of an immobile past would be a limitation. Rocks are not passive witnesses of human stories. Far from being immobile observers, stones move and evolve as active players in ecological systems.

Stone artefacts are to be considered components of the long narrative of interaction between people and environments, things encompassing social phenomena and geological processes. Objects interplay on different scales of matter (Bennett 2009), an idea that can be related to Schiffer's work (Schiffer 1987) on the formation processes of the archaeological records. Starting with the distinction between systemic contexts and archaeological contexts, Schiffer classifies the processes that can physically influence artefacts.

The ecosystems in which artefacts and sources are situated are in a continuous state of change and this transformation play an important role in defining a stone's geochemical signature. Quarries, mines or outcrops are exposed to weathering and buried sites are influenced by post- depositional processes (modification of the sediments once they are in place), both of which are capable of causing chemical changes in geological materials.

As mentioned in the introduction, archaeological records are biased towards lithic materials. This relative surplus of stone artefacts is explained by the physical properties of the materials themselves, which are more resistant to weathering and decay than others. This more enduring aspect of stones does not mean that they are

immune to contamination and transformation. Physical and chemical traces of erosion and alteration are valuable for understanding an object's itinerary. For example, the presence of a certain amount of sea salt in the pores of a statue could relate to its exposure to weathering agents in a location not far from the sea for a certain amount of time. A stage in the object's itinerary is invisible but still recorded in its material characteristics.

An interesting instance of post-depositional transformation is represented by the architectural mortars found in the Roman settlement of Massaciuccoli (Tuscany, Italy; Fig. 2). The structures, unearthed during a campaign from 2011–2012 appeared to be constructed using an architectural mortar that was extremely low in calcium hydroxide. Analysis of a few samples demonstrated that the $\text{Ca}(\text{OH})_2$ percentage was significantly reduced by the circulation of acidic water in the archaeological record in which the structures were buried. The mortars which, at the moment of discovery, seemed to be poorly made and quite fragile, had been heavily transformed by taphonomic processes and the modification of the materials' properties could be demonstrated through the observation of microscopic features in the thin section (Sciuto 2013). Changes in the materials of artefacts not only occur when the items are exposed to weathering factors (buried or not) but could also occur in protected environments, such as museums. Unfortunately, all too often we learn about objects being damaged in museums because of inadequate preservation conditions. Regretfully, lack of funding or appropriate structures has caused much historical information to be lost.

An awareness of the context in which the artefact is situated, whether a peat bog or sandy beach, is a good practice for the analysis of materials. Analytical results are usually influenced by the conditions in which the object has been preserved. A lack of knowledge of such environments could lead to misinterpretation of the results. The risk of inducing these kinds of problems is greater when archaeologists and archaeological scientists work independently, the former in the field and the latter in the laboratory (Killick 2015). Isolating samples from their context might cause analytical problems and misleading results, influencing scientific accuracy and the archaeological understanding of the record. The issue of weathering and contamination has unfortunately been quite neglected in archaeological science, while a large debate over stone alteration and erosion exists in architectural and conservation studies (a good compilation of the effects of atmospheric agents on monuments can be found in Winkler 2013).

In the case studies included in this project, the change in materials and the uncertainty of classification have been taken into account. The sampling protocol

has been planned taking into account the alteration in certain materials. Paper V states that a number of the analysed objects presented superficial weathering that have been avoided by ensuring that only the unaltered stretches are measured. In Paper III, surface alteration in lithic stone tools influenced the outcome of the study with a significant number of the fragments' raw materials resulting unclassified. The changes recorded are probably due to the characteristics of the original nucleus and secondary mineral formations within the podsollic archaeological record. The data augmentation produced by the hyperspectral imaging scan provided a better description of minerals and geological types but also incorporated many spectral information that influenced the classification (the analytical and statistical approach to recording and modelling uncertainty is further discussed in Chapters 4 and 5).

3.3 Walking stones: ethnoarchaeology of materials procurement in Nagaland.

When considering the agency of materials in stone procurement and the formation of social structures, the ethnographical record makes an essential contribution. A good example is the influence of ritual practices in the choice of materials for megalithic monuments in Nagaland (India)⁴.



Figure 6. Area visited during the study, Southern Nagaland, India. Image Uwe Dederig at German Wikipedia, CC BY-SA 3.0.

⁴ In February 2018, an international workshop and fieldwork was organised in the state of Nagaland in north-eastern India by the University of Kiel and the Nordic Graduate School in Archaeology Dialogues with the Past at the University of Oslo.

This area is known for the presence of several megalithic sites, which form part of the traditional rituals of the indigenous Naga people (Jamir 2016; Oppitz et al. 2008; Shimray 1985; Wunderlich 2017). In the villages of Khonoma, Maram Khullen, Khezakeno and Willong Khullen, situated in the mountainous region close to the border with Myanmar (Fig. 6), the recent practice of raising megaliths was integrated into ritual activities until the 1980s and is still carried out at some locations and on special occasions



Figure 7. Monolith in Khonoma (Nagaland, India). The large stones form part of daily activities and are integrated into the village landscape.

5. In this area, megaliths were erected as funerary monuments or to commemorate a “feast of merit”, a community celebration through which a member of the community gains social influence by organising a series of feasting. In Maram Khullen and Khonoma, megaliths form part of daily life; they are landscape markers between paddy fields, used for storing woodpiles and even as large cutting boards (Fig. 7). The stone types used for megalithic monuments appear to be varied, procured from more than one supply source. This evidence can be explained by the ritual practice as it is apparent in Nagaland that the stones themselves play a particular role in their raising rituals.

A man who wants to raise a stone for his celebration should wander in the forest to search for an attractive block. Once he has found it he should touch the stone with his bare hands and then return to the village. The stone itself would then speak to the man during his sleep. If he has positive dreams, the stone wants to be taken. However, if he has negative dreams (involving war or killing) the stone will not allow him to remove it and the quest has to start again from the beginning. The description of this ritual recurs in various interviews⁶ and is attributable to an animistic vision of rocks, moving and transforming according to their needs. The agency of stone is linked to its transformative power and its ability to communicate its wishes to humans. This belief is translated in the use of materials, not collected from a single source but rather recovered from various outcrops.

⁵ Data collection has been carried out in collaboration with Gangotri Bhuyan, postgraduate student at the North-Eastern Hill University in Shillong (India).

⁶ Thanks to *Gaon Bura* Mr. V. Punyü, Mr. Rang Thomas and the elders of the village of Khezakeno for sharing their knowledge.

4 Data collection methods

It has been argued that the characterisation of materials plays a crucial role in archaeological investigations and should therefore be fully integrated into research planning. A research plan should be arranged in order to satisfy the needs of archaeologists and archaeological scientists, to construct a common strategy inclusive of various approaches and capable of viewing archaeological objects from multiple perspectives. Interdisciplinary research is designed by taking into account expected outcomes and available techniques in order to collect significant datasets and obtain the best results.

As previously acknowledged, several scholars refer to the divide between archaeologists and scientists as being caused by the separation between the two worlds: if archaeologists primarily work in the field, scientists, on the other hand, are largely confined to laboratories in which they receive samples collected by other parties during excavation/survey campaigns (Engelmark and Linderholm 1997; Hayashida 2003; Jones 2001; Pollard and Bray 2007). This physical gap contributes to a lack of communication and causes misunderstandings between specialists, ultimately influencing the results of the analysis and the interpretation of data. A poor understanding of the context could cause scientists to make errors when interpreting the analytical results for materials altered by environmental conditions. At the same time, archaeologist might conduct sampling incorrectly, not knowing the analytical procedures, causing specimens to be contaminated. This is particularly important for delicate materials and highly accurate analysis. Stone artefacts may appear to be more resistant to change, at least on a macroscopic level, but invisible salts and secondary minerals can fill the rock pores while water can dissolve elements and cause erosion (see paragraph 3.2). Considering that multiple factors could interfere with analytical procedures and influence results, it is crucial to know the formation processes of an artefact's archaeological record. A good understanding of an object's context can be accomplished by merging the empirical knowledge of fieldwork with scientific expertise, creating an approach to archaeological evidence in which the analysis of materials is considered part of the research core rather than an additional activity. By adopting a resolution based on a collaboration between archaeologists and scientists both in the field and in the laboratory, the entire experimental design and data collection strategy can be optimised to recover information already comprehensive of interdisciplinary research questions.

Various methods and tools can be combined to collect information about the materials of stone artefacts, through a selection process carried out according to the

context and research goals. The chemical and mineralogical characteristics of artefacts can be recorded by referring to standard systems and using analytical techniques that can be destructive or non-destructive, portable or fixed. I argue that NIR spectroscopy application in provenance studies could provide an interesting alternative to some of the instruments commonly used in archaeological science. NIR-based instruments allow the collection of large spectral datasets in various contexts and can be implemented in different archaeological research projects. The documentation of morphology with details of production-related marks can be carried out in various ways using analogue or digital techniques. In all the case studies presented, different methods have been combined in the field and in the laboratory for collecting, analysing and visualising a comprehensive set of information on the artefacts. The methodologically common denominator for the case studies is the application of near infrared spectroscopy to characterise stone types. The various procedures adopted are listed here and described in detail: analytical techniques, protocols for observing morphology, determining the typology of objects and tools for documenting manufacturing processes.

4.1 Analytical methods

Several chemical analytical methods are used for characterising stone artefacts in archaeological contexts and the number of new instruments is constantly growing. The evaluation of an artefact's characteristics and research goals are crucial to planning data acquisition campaigns. The choice of an analytical method depends on a series of conditions (Shackley 2008):

- The geological features of the raw materials. Sedimentary, igneous or other types of rocks have different petrographic characteristics that can best be recorded using specific techniques;
- Portability. Certain archaeological objects cannot be sampled or moved to the lab and require analysis using portable instruments;
- Sampling. Some instruments are flexible and allow the collection of more samples, others are more accurate and provide a good analytical result with few measurements.

Another aspect to consider when choosing instrumentation is funding. Often, archaeology departments are not equipped with much analytical instrumentation and some devices have to be borrowed from other institutions. Most of the time, the cost of analysis is independent of the cost of human labour, but relates to equipment, chemicals and sample preparation. Costs must be included in the budget provision when envisaging interdisciplinary research and this economic aspect might influence the choice of analytical procedures.

4.1.1 A matter of sampling

An important concern in environmental archaeology and in archaeological science in general is choosing an efficient sampling strategy. The selection of a representative dataset is crucial for collecting information that is illustrative of a certain phenomenon. In archaeology, the sampling dilemma has too often been underestimated, with several studies focusing on analytical techniques to the detriment of the evaluation of information sources (Beardsley and Goles 2001; Orton 2000).

Most provenience studies rely on a convenient sampling method based on a project's budget, research questions, available methods and the objects' characteristics (Orton 2000).

A good selection of samples is fundamental when characterising stone artefacts. For example, when trying to determine the origin of a certain raw material, the analysis is usually articulated on two distinct targets: artefacts and supply sources. Sampling should be planned in order to obtain a dataset that is representative of the variability of both the archaeological objects and potential quarry outcrops in order to identify matching mineral characteristics. The size, shape and location of the study object will also influence the analytical protocol: sampling to characterise building materials in large architecture will differ from the strategy applied to lithic tools. Smaller objects can be analysed in a laboratory using stationary instruments while the characterisation of outdoor features requires an invasive collection of samples or the use of portable equipment. In some cases, archaeological features (notably quarry walls or portions of masonry) are difficult to access, imposing constraints on sampling opportunities. Analytical methods that are adopted to characterise stone materials also play a role in the definition of a sampling strategy. Most of the geochemical methods applied in provenance studies (for example, petrographic analysis, mass spectrometry and instrumental neutron activation analysis, i.e. INAA) are destructive, expensive and time consuming. As a consequence, analysis can only be performed on a limited number of items.

The sampling issue is not only technical but represents a theoretical dilemma, because by selecting variables to record and items to measure, archaeologists are influencing the questions that could be posed to materials (Boivin and Owoc 2004). There is no final and valid solution to the sampling debate, but some specific issues can be addressed to improve analytical results. Firstly, sampling strategies need to be explicitly discussed, highlighting the issues and benefits; the samples collected should be sufficient for assessing the variability of the geological features but not too much in order to avoid the accumulation of redundant data (Beardsley and Goles 2001).

For destructive analysis, the issue of representativity is crucial: if objects are destroyed, the number of items to be analysed will be as little as possible. The task is easier when dealing with the sourcing of uniform rock types (such as obsidian, see Tykot 2004) but becomes much more complex when practiced on extended outcrops or materials that present a substantial variability (for example, sandstone/conglomerate).

Although portable and non-destructive methods allow the collection of a large number of measurements in a short time, the procedure should always be controlled to avoid problems with data processing. Non-destructive methods can be used extensively for characterising the geological variability of an object's surface, defining the chemical topography of a rocky face. In this way, weathered or altered spots can also be included in the samples in order to assess the variation resulting from external factors.

A good experimental design relies on the experience and the capacity of observation of the researchers who should know the context and formulate pertinent research questions in order to choose samples in the most expedient manner. Even the fastest and most reliable analytical technique cannot replace the observation made by an experienced observer (Caraher 2015). For this study, the analytical techniques applied are all non-destructive and some instruments are also portable. Sampling has always been planned according to the physical characteristics of the items in order to collect a dataset that is comprehensive of the research questions and theoretical implications, based on context and research needs (Boivin 2005). In each article presented, the sampling strategy is indicated and discussed, taking into account materials and analytical techniques. In the following paragraphs, the properties of the instruments are described in detail together with a discussion about data collection.

4.1.2 Near infrared spectroscopy

Near infrared spectroscopy has been applied to all the presented case studies. It is a technique that is relatively new in archaeology for the characterisation of materials although it has been largely used in other disciplines and on other targets. NIR spectroscopy is a technique based on a material's molecular vibrations in the near infrared band of the electromagnetic spectrum recorded through different types of detectors. It is a non-destructive method that requires the use of a controlled light source and digital captors (such as cameras or probes). NIR spectroscopy does not require sample preparation and can be implemented in virtually every context.

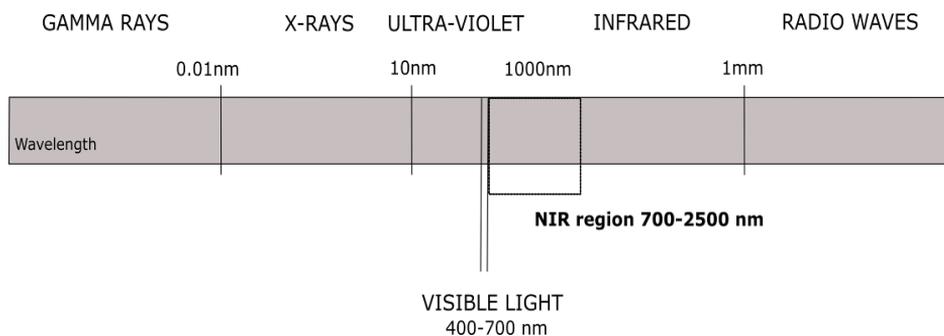


Figure 8. Electromagnetic spectrum and near infrared bands.

NIR covers the region of the electromagnetic spectrum situated between 700 and 2500 nm (close to the interval of visible light; Fig. 8) and contains absorption bands corresponding to overtones and combinations of fundamental C-H, O-H, and N-H vibrations. Absorption is recorded as spectra with peaks that, in some cases, can be identified and assigned to the vibration of specific molecules but more often result in broad features that are the result of aligned peaks (Bokobza 1998). NIR spectroscopy is not used for tracing or quantifying specific elements but as a fingerprint of materials. The shape and intensity of the absorption features are given by the structure and distribution of the particles analysed, as well as their molecular composition. In this way, measurements recorded through this kind of spectroscopy provide a broad overview of the material qualities of the objects and offer other significant advantages.

As previously mentioned, NIR spectroscopy is completely non-destructive and does not always require the captor to be positioned in contact with the target (this is generally valid for aerial and satellite photography while the probe and cameras used in this research had to be positioned relatively close to the target, see description below). Recording spectral information takes a few seconds and measurements can be multiplied several times. Moreover, the spectra collected can be evaluated in real time through PCs/tablets connected to the spectrometers, to detect any errors. These characteristics make NIR spectroscopy a method with interesting application potential in archaeological research, being portable in the field and convenient for scanning objects in the laboratory.

Hyperspectral images (HSI) are the result of several overlapping images at different wavelengths taken on a target. In hyperspectral images, each pixel represents one spectrum. The combination of images and spectral dimension results in a hypercube including spatial and chemical information (Fig. 9).

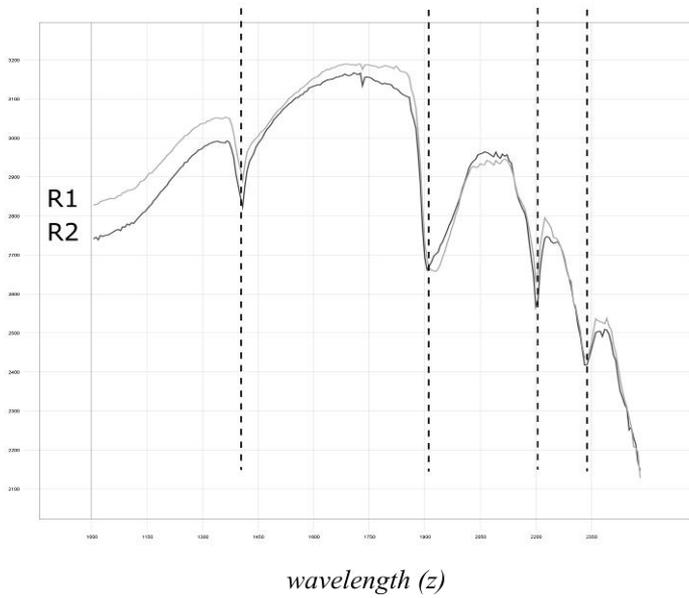
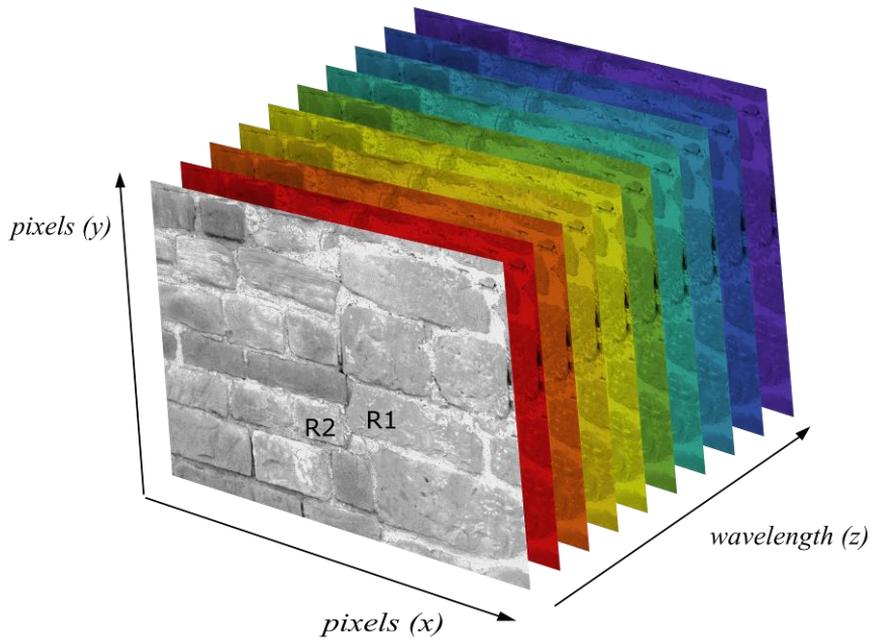


Figure 9. Structure of a hyperspectral image

This research is aimed at testing the application of NIR spectroscopy in archaeological provenance studies although the same technique is largely used in other fields. NIR spectroscopy is particularly suitable for detecting organic materials and is used in routine analysis as a control test in pharmaceutical industries, agriculture, food production and geology. In particular, the protocols applied to various case studies are inspired by NIR spectroscopy geological applications used for mineralogical mapping of the lunar surface (McCord and Adams 1973), mineral exploration (Goetz, Rock, and Rowan 1983), mineralogical analysis of drill cores (Kruse 1996), field mapping of expansive soils (Chabrilat et al. 2002) and field mapping of mineral assemblages for gold deposits exploration (Bierwirth, Huston, and Blewett 2002). In geological surveys, the use of near infrared imaging has been associated with the detailed documentation of surfaces (through LIDAR scanning) to describe morphology and mineralogical variation in vertical quarry faces (Kurz, Buckley, and Howell 2012). NIR spectroscopy has been successfully applied to determine heat treatment in silica rocks (Schmidt et al. 2013) and traces of organic residues (Prinsloo et al. 2008). NIR spectroscopy has also been tested for sourcing chert artefacts, adopting a fingerprint approach (Parish 2011).

Exploring the application of near infrared spectroscopy in archaeology was the target of the Mobima project, with the specific goal of developing an experimental analytical protocol for materials analysis that could be applied to artefacts directly in the field or the semi-automated screening of large datasets in a laboratory setting. The use of an NIR-based instrument represents an analytical challenge that permits the collection of large datasets of fingerprint information that can be processed to highlight patterns that are useful for archaeological purposes (Linderholm et al. 2013; Linderholm and Geladi 2012).

Light, portable and quick image-based techniques are convenient for mapping the surface chemical characteristics of materials, multiplying the number of measurements and evaluating rock characteristics.

NIR spectroscopy is a versatile tool and its use can be adapted to the research target and goal. It can be used as a unique tool for characterising different types of raw materials or as an exploratory method for highlighting similarities and dissimilarities in composition of materials and to set sampling strategies for elemental analysis. Spectral datasets provide unbiased information about an artefact's materials that could substitute optical observations. For the purpose of this research, within the Mobima project, two different NIR devices have been used in order to evaluate applications for outlining the provenance of archaeological materials.

- 1- A portable micro probe produced by Jdsu (now Viavi), which covers a wavelength range of 908–1676 nm with two integrated tungsten lamps and a captor that is 5x3 mm wide. The probe is connected via USB to a tablet and handled using software to evaluate and store the records. The instrument is calibrated every 5 minutes (on average) using a black and white reference (respectively, 100% and 0% absorbance). The white reference (100% reflectance) is made of spectralon, a fluoropolymer with the highest diffuse reflectance of any known material in the near- infrared region of the spectrum. For the black reference (0% reflectance), the manufacturer's recommendation for the JDSU probe is to calibrate the black reference by pointing the spectrometer at a dark corner. In field conditions, finding a spot with no natural or artificial light disturbance could prove difficult. Thus, an easy solution has been adopted by placing the probe on a sheet of fine-grained black sand paper. The portable probe is light, easy to carry and handle, making it particularly suitable for research directly in the field (Sciuto, forthcoming).
- 2- Hyperspectral imaging systems are larger and more complicated to move. The sisuCHEMA pushbroom short-wave infrared camera covers a wavelength range of 1000–2498 nm and is used in a laboratory mounted on a conveyor belt. The camera is calibrated on a spectralon white reference (moving in front of the captor by means of a robotic arm) and takes images of adjustable length, field of view and focus. This camera is mostly used to scan large sets of artefacts, sample series or long, continuous samples, such as soil cores. The NEO HySpex-320m-e (Norsk Elektro Optikk, Skedsmokorse, Norway), linescan pushbroom camera with 256×320 HgCdTe sensor and a PGP filter was used in the field for the case study described in Paper IV. The camera records wavelengths from 1,000 nm to 2,500 nm on 256 channels. The instrument was calibrated using a spectralon white reference and mounted on a rail to move horizontally through a computer-controlled stepping motor. The recorded image stripes can be overlapped to form larger hyperspectral pictures.

The instruments that were tested and combined offered good flexibility for collecting data in different situations, as well as adapting to various conditions and research goals. Hyperspectral imaging allows total coverage of spectral information on an analysed surface. For every hyperspectral image, the object's spectral characterisation takes place on a pixel grid. In each image a spectrum corresponds to one pixel, which means that the details of the information are determined by the amount of material included in one pixel. Pixel size depends on the distance between

the object and the camera and the resolution of the camera itself (for example, satellite hyperspectral images released by ESA have a resolution of 10 m² per pixel⁷). Not all information in a hyperspectral image is valuable. For this reason, an image can be reduced to some relevant clusters, generally applying multivariate image analysis techniques (MIA, explained in detail in paragraph 4.3). Acquiring point measurements using a portable MicroNIR probe is different. This fast and light instrument can be used for collecting multiple measurements on the rock's surface, achieving a good coverage to represent the mineral variation of the geological facies. For example, on quarry walls of sedimentary bedrock the probe is often used to record several vertical profiles, set up directly in the field according to morphological and geological features (see Paper V). For single small objects, for example, masonry elements, five probe measurements are collected in random spots, usually taking care to avoid altered portions. Larger objects are characterised by more measurements but, generally, the number and position of the recordings are chosen taking into account the rock type and characteristics of the artefact itself.

Hyperspectral imaging and a MicroNIR probe can also be used as complementary methods of elemental analysis for characterising stone artefacts. The diffused spectral information provides a map of an artefact's geological variability that can be taken into account when planning a sampling strategy. NIR spectra provide detailed information about the molecular characteristics of a lithic artefact. Thus, samples for destructive analysis can be collected in such a way as to acquire the most relevant chemical information without damaging the target more than necessary.

Working directly in the field has some disadvantages. Hyperspectral cameras and hand-held MicroNIR probes have constraints dictated by their technical characteristics. Both instruments record spectral features of objects positioned close to the captor; the distance can vary up to 1 m for the hyperspectral camera while the probe is usually placed against the object's surface. This factor limits the use of these methods for objects or outcrops positioned further away from the user. An example of this issue can be found in Paper III. In Carcassonne, only the lower portions of the defensive walls were analysed, taking care to measure masonry elements associated with all the identified construction phases. Field conditions are not constant, especially in terms of humidity, temperature and light. Thus, calibration has to be constantly repeated in order to obtain reliable measurements. Light conditions could represent a problem. The MicroNIR probe is equipped with a built-in technical solution: tungsten lamps are integrated into the captor in such a way that when the probe is placed on the object's surface the light used for the recording is only the light that is emitted by the lamps. Similar to this is the case of hyperspectral

⁷ <https://sentinel.esa.int/web/sentinel/missions/sentinel-2/overview>

cameras in which the target has to be struck by a strong source of controlled light. This procedure is easily conducted in a laboratory but working in the field, the variation of sunlight intensity could influence the spectral recording. For gaining better control of the light source, the masonry measurements in Carcassonne were carried out in the shade, with a lamp mounted on the camera to illuminate the target.

4.1.3 Energy Dispersive X-Ray Fluorescence (ED-XRF)

The accuracy of NIR portable spectrometers and imaging systems can be assessed using an internal validation model and through comparison with other non-destructive and portable techniques. The method that has proved to be suitable for the correlation, in accordance with our project aims and goals, is portable Energy Dispersive X-Ray Fluorescence spectroscopy (ED-XRF). The use of ED-XRF in archaeology has a time-honoured tradition; applications of this technique to the characterisation of materials have been developed from the 1960s, together with new advancements in the disciplines of geochemistry and petrography. Various papers have been dedicated to the discussion of this method (Frahm 2014; Frahm and Doonan 2013; Hughes 1998; Olson 2011; Shackley 2011; Shackley 2012a; Shackley 2012b; Speakman and Shackley 2013), especially after the so-called “revolution” of reasonably-priced portable instruments becoming available on the market (Shackley 2011). In recent years the spread of hand-held XRF spectrometers used in archaeological investigations has caused a significant increase in elemental analysis in archaeological contexts (on soils and artefacts). Nevertheless, the archaeological-scientific community has, on various occasions, emphasised that limited knowledge of the instrument’s specific functioning could result in errors and misinterpretation of chemical data. The discussion about the reliability and treatment of XRF data has focused on the application of calibration systems. Portable XRF devices are sold as in-built “off the shelf” calibration models. The instrument’s signal is calibrated through a fundamental parameter procedure against samples of known composition. Nevertheless, detailed information about calibration algorithms and calibration samples are not always available to users. The application of these calibration algorithms could result in an erroneous quantification of certain elements, particularly when the target composition is quite different from the calibration sample. Even though the use of portable XRF appears to be easy and quite straightforward, it should be remembered that “off- the-shelf” calibrations are often not sufficiently accurate enough to quantify the elements. This type of calibration is good for a presence/absence assessment but sometimes not accurate enough for using the quantified result for sourcing materials (Shackley 2012a).

For the studies described in Papers III and IV, a portable Niton XL3t (Thermo Scientific Niton, Billerica, MA, USA) ED-XRF analyser has been used, collecting the surface measurements of various stone artefacts. The ED-XRF instrument can be carried in field operations or positioned on a stationary test stand so measurements can be performed in both field and laboratory settings. ED-XRF measurements have been repeated two or three times in different locations on an object's surface. The measurement's number and position were chosen according to the object's size and the characteristics of the material (more or less homogeneous). The instrument is supplied with a 50 kV X-ray tube, an Ag target and a Si-PIN detector with a resolution of 175 eV at Mn K α (5.895 keV). The set acquisition time for every ED-XRF spectrum is 120 seconds. The resulting measurements comprise three different intervals of 40 seconds each in which the radiation is recorded using three different filters and X-ray tube settings. The light mode is designed for measuring light elements (for example, Si and P). The first part of the measurement is performed without filter with a tube voltage of 6.2 kV. The low filter mode uses a tube voltage of 20kV while main filter modes use filters with tube voltages of 40 kV. ED-XRF measurements resulted in a 4,000 data points spectra, being collected from 0 to 60 keV at intervals of 0.015 keV (bins). In order to avoid the application of integrated calibration algorithms, regarded as misleading for the specific materials object of the case studies, the collected spectra were downloaded as raw data and modelled statistically using a fingerprint approach. The raw spectra were reduced by removing bins with no, or low, signals and resulted in spectra made up of light, with low and main filter sizes of 0–2.8 keV, 2.8–7.0 keV and 7.0–19.0 keV, respectively. The point values were then normalised to the X-Ray tube current by dividing each spectral value (counts/second) with the corresponding tube current value obtained from the instrument parameter file. The spectra obtained were plotted to identify relevant peaks and evaluate the measurements while classification was performed by means of multivariate statistical modelling (Thyrel 2014).

4.2 Documenting stoneworking processes

The visible attributes of an object, such as details of its shape, can inform about specific techniques of manufacture of a certain material. For this purpose, various documenting methods have been applied for recording morphological details useful to reconstructing specific aspects of technical gestures and production modes.

The study of technology and production is primarily carried out through the analysis of marks left by stoneworking techniques. These techniques vary significantly according to time, materials and the organisation of labour. Thus, evidence of craftsmanship is useful for understanding the socio-economical context in which an artefact was manufactured. The archaeological practice of recognising and

differentiating production techniques originates in the theories of structural anthropologists and the ground-breaking work of André Leroi-Gourhan. As suggested in his book *L'Homme et la Matière* (Leroi-Gourhan 1964a), technical tendencies can be traced through different societies, recording variations in the production process (see Chapter 3). If the message of Leroi-Gourhan has been largely received in prehistoric archaeology with a definition of subsequent stages of lithic tools *chaîne opératoire*, historical archaeology appears to still be grounded in a more typologically traditional classification that favours the stylistic study of shapes and decoration rather than production processes. A systematic study of stoneworking techniques was published by Bessac (1986), creating a complete catalogue of tools for stoneworking, their use and relative marks. Bessac adopts an approach based on Leroi-Gourhan's classification system for describing human gestures and the material results of the impact of tools on stone. In Bessac's book, stages of production are described in detail, referring to existing evidence found at historical sites. Similar to this is Rockwell's manual on stoneworking techniques (Rockwell 1993), which focuses on sculpture rather than other types of artefacts. The repository of Rockwell's archive, made available online through a project at King's College London (Pasin et al. 2012; Wootton, Bradley, and Russell 2013) is an important digital reference for stoneworking techniques and toolmark identification⁸.

Digital and analogue methods of recording the shape of toolmarks and the volume of objects have been utilised in the various case studies. Documentation has mainly been carried out through photogrammetry. This method has proven to be extremely useful for documenting archaeological evidence in surveys associated with GPS (Global Positioning System), as shown in Paper V, as well as for producing 3D models that can be annotated with information from various observations (such as toolmarks, masonry description, etc.).

4.2.1 Paintings and lithics

The processes of painting rock faces in prehistory are mostly known through ethnographical comparisons. Chemical analysis of pigments has shown that in most cases the red colour is obtained through the oxidation of iron-rich sediments. Red ochre powder, usually obtained by heating fine-grained soil portions, can be mixed with organic or non-organic binders to create a moist mixture. The formula for each ochre batch is unique and the same pigment can be used for one or more figures. This characteristic makes it easy to detect paintings that were realised simultaneously, using the same pigment, from the chemical fingerprint. In order to distinguish the

⁸ <http://www.artofmaking.ac.uk/>

spectra of every stretch of painting in Flatruet (Paper I), a high-definition documentation methodology was adopted. The painted panel was documented using photogrammetry to capture the morphology of the rock face, while the exact position of the point measurements collected with the MicroNIR probe was recorded in situ using a tablet. The information on the tablet and the photogrammetric image were subsequently merged, annotating the tridimensional model using ArcScene (by Esri). Using this documentation system, spectral measurements are associated with the exact portion of painting with a margin of error of 2–5 mm because of the size of the captor itself.

A 3D GIS platform was used for analysing the distribution of lithic fragments at the Mesolithic dwelling site of Lillsjön (Paper III). The analysis of rock fragments resulting from the knapping process was carried out a few years after the excavation. The position of the finds in the excavated record, originally documented on paper, was visualised using the existing documentation from the excavation implemented in a 3D GIS platform, using Esri ArcScene. The tridimensional projection of lithic categories allowed us to highlight patterns in the use of raw materials during the lifespan of the site, pinpointing the change in supply sources between the earliest and latest phase of occupation.

4.2.2 Buildings

The production of building materials is without doubt the main reason for stone exploitation, at least in terms of volume. Free-standing structures are often made of different kind of materials that may have different origins. The use of stone or bricks usually varies from one chronological phase to another resulting in mixed architecture, products of restoration and reconstruction. The archaeology of architecture is crucial to reconstruct the chain of building events highlighting gaps in the construction process. This sub-discipline of archaeology is strongly linked to the history of architecture and relies on the stratigraphic documentation of standing structures. Buildings archaeology has a long-standing tradition, particularly in Western/Central European and Mediterranean countries (especially Italy, Spain, France, Germany, U.K. and Greece). The analysis of buildings is based on the description of stonework, its typology and chronology in order to date the various stretches of masonry and understand the organisation of the working site (Bianchi 2004; Blary, Gély and Lorenz 2008; Boato 2008; Brogiolo 1996; Brogiolo and Cagnana 2012; D’Ulizia 2005; Ferris 1989; Mannoni 1990). Morphometry and the identification of toolmarks have been used for interpreting stone dressing techniques and marks resulting from the organisation of working sites, as well as equipment for lifting and positioning the blocks. In Papers IV and V, spectral data are systematically

associated with the corresponding construction phase. This combined approach allows the assessment of the use of materials in a particular stretch of architecture. The position of measurements is recorded in the field by using orthophotos of the walls (projected from photogrammetric 3D models) on which spectral measurements and chronological phases are reported.

4.2.3 Quarries

Stone extraction often leaves voids detectable in the landscape. A specific characteristic of quarrying sites is that they are carved instead of built, formed by erosion (induced by humans) rather than deposition. A direct evidence of ancient quarrying is quite rare as it is often erased by subsequent activities. A constant removal of materials systematically obliterate earlier traces. Thus, dating a quarry is generally very difficult. Quarry faces that still show ancient toolmarks are most relevant to understanding extraction processes. Accurate detection of the use of different tools in a quarry can provide information about the products, management strategy and social structure of the working site. The interpretation of toolmarks helps in understanding the phases of exploitation in a quarry and provides important data for understanding the structure of an entire subsection of the community that lived and worked around stone extraction and processing (Allios 2014; Bourin, Gardel, and Guillot 2014; Lamesa 2011). The quarry site described in Paper V has been documented using photogrammetry. Measurements of the extracted modules have been annotated on the model as well as the dimensions and directions of toolmarks.

5 Mind the data: statistics, databases and visualisation.

This section is dedicated to the discussion of techniques for reducing, processing and visualising data in order to create platforms for integrating information collected from various sources. The association of different methods results in composite datasets, including quantitative and qualitative data that can be studied using statistical methods or organised in databases for visualization and analysis.

5.1 Raw data, preservation and dissemination

Data collected using analytical instruments present a compatibility issue. In fact, there are no common standards for the collection and storage of archaeometric data and every study somehow remains an isolated experience. Every analytical technique comprises precise technical details and specific instruments produced by specific companies often have different detection limits and settings. The result is that almost every researcher or research group working on provenance studies are independent and generally gather isolated datasets, describing one specific phenomenon. The situation regarding raw data storage and open repositories is not different for NIR spectra. While the technique is widely used in geology, archaeological applications are still scarce and reference data for some specific artefacts are almost impossible to find. Several geological spectral libraries that contain references from different materials are already available as an open source from various agencies: the United States Geological Survey (USGS) has a database comprising over 1300 spectra⁹, including minerals, man-made materials, vegetation, etc., while the Jet Propulsion Laboratory ASTER Spectral Library version 2.0¹⁰ also contains more than 2400 spectra of various materials.

Unfortunately, dimensions and intervals of NIR spectra are determined by the specific settings of the instrument used for collecting the data. For example, the spectral bands recorded using the MicroNIR are those included between 908 and 1676 nm, while the SisuCHEMA hyperspectral camera records spectra at a greater interval, between 1000 and 2498 nm. Intervals between data points can also vary as well as calibration procedures, making it almost impossible to directly compare spectra collected using different instruments. Generally, the identification of spectral features is achieved through comparison with systematised atlases of reference

⁹ <https://speclab.cr.usgs.gov/spectral-lib.html>

¹⁰ <https://speclib.jpl.nasa.gov/>

(Weyer and Workman 2012), although archaeological-specific materials are not included in these anthologies. As one aim of this project was to demonstrate the value of NIR-based analysis for archaeological investigations, a future outcome could be the systematisation and implementation of the collected data, with the relative metadata, in an online repository available for other researchers working with these techniques.

5.2 Multivariate statistics and chemometrics

NIR spectroscopy can be used extensively as target-specific portable spectroscopy or an imaging technique that provides a detailed overview of the physical-chemical characteristics of the materials. The broad range of applications and the user-friendliness of these devices make it easy to collect large spectral datasets. Archaeometry, like the rest of archaeology, is involved in the big data challenge (Gattiglia 2015), both from a computational and a theoretical point of view. Statistical and interpretative tools have been improved to process large volumes of information in order to find correlations rather than causations. In such large volumes of information the sampling bias becomes less influential and patterns can be highlighted using data reduction procedures. The approach adopted is based on data mining, consisting of applying statistical models to classify information through hierarchical and cluster models. Statistical tools can be used to detect similarities and patterns among the data, cross referencing large datasets and creating clusters of aggregated data (Boyd and Crawford 2012).

Statistical data reduction techniques are fundamental to deriving meaningful information from a multitude of values and variables. Nevertheless, these procedures should be applied with caution. The augmentation of data size increases the volume of information that is potentially extracted but also broadens the level of uncertainty. In choosing statistical models to apply to datasets, researchers are influencing the results as much as in the planning of sampling. Data trends visible with a certain model might not be clear in another model. Thus, interpretation of the analytical results might be different. Use (and misuse) of statistics is adapted to the type and format of data, the characteristics of the technique and the expected correlation outcomes.

NIR spectroscopy can be applied as a digital screening method to easily and quickly highlight the geological characteristics of materials. Nevertheless, data collected in a short time need to be processed in accordance with specific protocols elaborated for the purpose (see next paragraphs). In every hyperspectral image the number of spectra is equal to the number of pixels, while the single spectra collected with the

portable MicroNIR can be multiplied several times (for example, the first field test application of the NIR probe for the field study of rock paintings, partially discussed in the first paper, allowed the collection of 1,432 spectra from six different sites in five working days). A consequence of the extensive use of NIR spectroscopy is the creation of big data matrices containing a lot of information and a considerable number of variables. The data collected with the hand-held probe are visualised on the laptop/tablet in real time during the acquisition for quality control purposes. This first in-field assessment is crucial to estimating the nature of the recordings and detecting incorrect measurements. Real-time visualisation helps to gain an initial understanding of the dataset and the spectral features that characterise the materials. The data matrices are usually exported as comma-separated files with the corresponding variables/wavelengths. The hyperspectral camera mounted on a fixed support is also equipped with a computer for performing direct data evaluation. The images are then exported as raw files in folders containing the metadata and the white reference information for calibration. The option to visualise spectral data directly during the acquisition via digital tools represents a substantial difference between NIR spectroscopy and other analytical methods. The molecular characteristics of materials can be revealed while looking at the artefacts through the spectrometer lens providing a useful tool for sampling and work planning.

In order to reduce the amount of information contained in these files and highlight interesting spectral features, data analysis is usually carried out through methods used in chemometrics. This discipline was born in the 1960s to statistically manage the increasing volume of chemical datasets generated by powerful new devices (Wold and Sjöström 1998). Chemometrics has its roots in multivariate statistics, focusing on the development of models for analysis and the prediction of chemical data and is particularly useful for processing spectral information. NIR technology associated with chemometrics has become a powerful tool for screening and evaluating big datasets, by working on associations among different materials in a way that would not have been possible with other types of elemental analyses. Exploratory analysis in archaeological datasets by means of multivariate statistics is conducted in archaeological science in different contexts (Baxter 1994). The multivariate models used for processing data matrices are described here with specific reference to the potential applications. The software used for multivariate modelling are Evince (by Prediktera¹¹), and SIMCA (by Umetrics¹²). For the purpose of this study, several statistical models have been applied to datasets to classify items and visualise clusters. For each paper an explicit goal was to use the

¹¹ <http://prediktera.se/>

¹² <https://umetrics.com/products/simca>

easiest and most effective statistical solutions for acquiring the maximum amount of information through relatively simple computational steps.

5.2.1 Principal Component Analysis (PCA)

Principal Component Analysis (PCA) is probably the most common algorithm for data survey and dimension reduction. This projection method is valuable for detecting variation in a data matrix. In spectroscopy it is used as an exploratory tool to detect patterns in the data and identify outliers. Through the application of PCA, a matrix of observations and variables can be reduced to a low dimensional plane that describes relationships between observations and variables (Eriksson et al. 2013). Through this data exploration procedure, a multivariate space is reduced to a bidimensional plot representative of selected components with the highest percentage of spatial variation explanation. The evaluation of the variables involved in the score plots is conducted by looking at loadings (lines or plots) to visualise spectral features and the relationships among them.

Spectral data are usually pre-processed and scaled in order to be suitable for analysis via PCA. NIR spectra are often influenced by scattering effects, which result in noise in the recorded data. To improve the exploratory analysis of measurements collected on lithic samples, the scattering correction applied is usually a Standard Normal Variate (SNV). This transformation is applied individually by scaling each spectrum by its own standard deviation. Spectroscopic data matrices are mostly mean centred, a standard procedure for data pre-processing consisting of calculating the mean for every variable and subtracting it to the value, without changing the scale. ED-XRF data are usually pre-processed by applying a Pareto scaling algorithm which results in a mix between Unit Variance Scaling (UV), reducing the importance of high-variance variables, and mean-centring, preserving the data structure.

5.2.2 Multivariate Image Analysis (MIA)

Hyperspectral image files can be quite laborious to process. The dimensions of one single data file exported from a hyperspectral imaging system can vary from about 50 to 500 MB (raw format). Datasets made up of various images can be very heavy. For example, when working with lithics from the Mesolithic site described in the second paper, the total memory space of the entire image datasets reached up to 22 GB (for a total of 2612 single stone fragments analysed). Processing information delivered by HSI requires specific stages that can be batched in order to reduce the size of the files without losing spectral accuracy.

Multivariate Image analysis (MIA) is the most common procedure for treating multi- and hyperspectral images. Every picture is divided into a system of x and y coordinates depending on the number of pixels contained in the file, while z is determined, in our case, by the spectral bands (in wavelengths) covered by the device. Image treatment consists of reducing the spectral information by highlighting the relationships between variables and contracting the dataset (Geladi and Grahn 1996; Grahn and Geladi 2007). MIA has been developed since the 1980s to detect structures hidden in image data, quickly becoming the common ground for chemometrics and image processing. Images can be analysed by taking into account information associated with single pixels or evaluating the entire image (Prats-Montalbán, De Juan, and Ferrer 2011). The most common MIA model is PCA, which is calculated on the entire data matrix. The image is visualised as a colour composite and a new value is associated with each pixel according to the information highlighted by the PCA components. Fig. 10 shows how by switching from component 1 to 3, the image appears different. This effect is due to the spectral features used as variables for pixel classification.

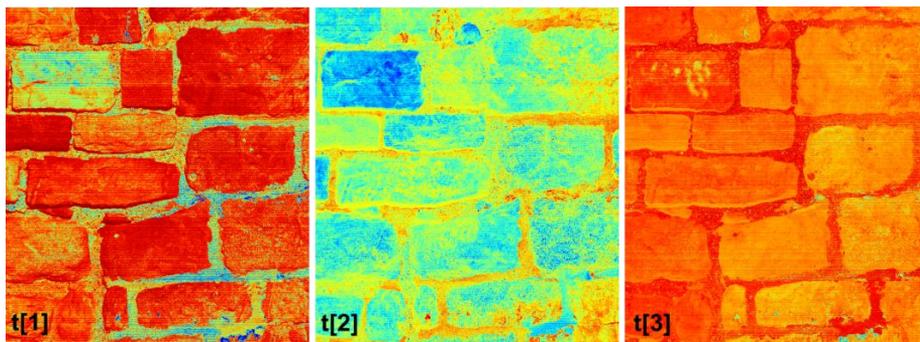


Figure 10. PCA on a hyperspectral image from the wall of Carcassonne (Aude, France). The colour composite score images shows the different spectral feature highlighted by the first three components (respectively $t[1]$, $t[2]$ and $t[3]$).

The pixel values in the PCA can be visualised as a score density plot, often made unclear by the high volume of data involved but helpful for showing large clusters of pixels. By combining the image plane and score plot it is possible to identify clusters, evaluate the spectral features and replace the pixels on the image space (Wise and Geladi 2000). In Fig. 11 the same stretch of masonry is associated to the score plot. The pixel cluster selected in black on the top image corresponds to the spectral response of mortar that results highlighted on the score image below.

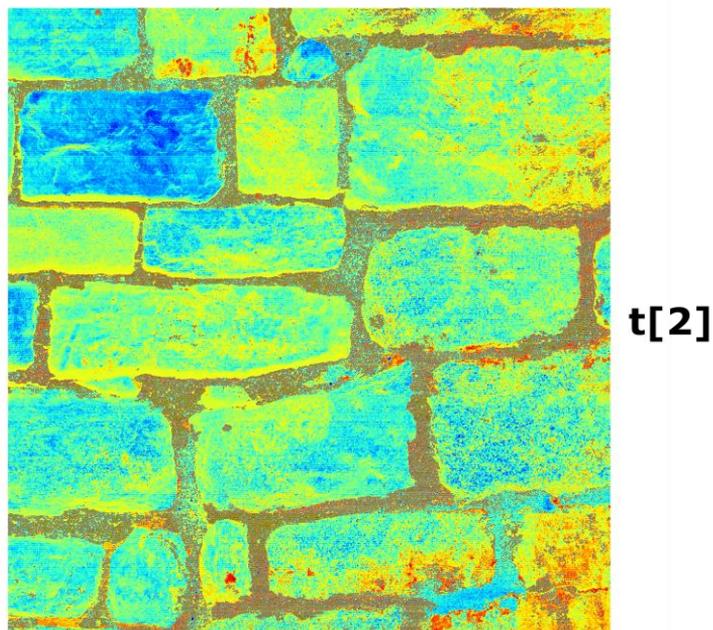
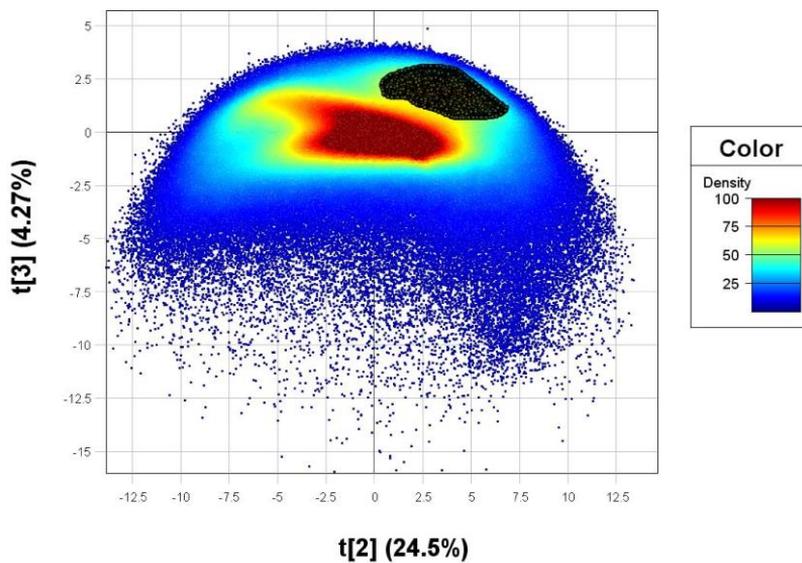


Figure 11. Combination of score image and score plot. The pixel cluster selected on the score plot corresponds to mortar, as automatically highlighted in the image below.

A PCA model of images is a useful tool for assessing image quality and detecting any problems in the recordings. For this reason a lighter version of Evince software, called Breeze (by Prediktera) has been connected to the SisuCHEMA Pushbroom infrared imaging system in the laboratory. The software allows the user to create on-line PCA models of the acquired images in order to evaluate the dataset and determine the strategy for further measurements. Moreover, it allows the calculation of predictive models for performing classification directly on the newly-acquired images.

5.2.3 Soft Independent Modelling of Class Analogy (SIMCA)

Multivariate statistics are also useful for performing the classification of unknown spectral data. Various models are available for prediction and classification and their use depends on the dataset and the spectral features involved in the analysis. The first step stage in making a conscientious choice of a classification model is the evaluation of the dataset, variables and training set. Classification of lithic materials based on infrared spectral characteristics can be problematic because of the high variability of a spectral dataset and the large number of variables involved in the analysis. The models applied should be based on the evaluation of similarity and larger clusters.

For analysing datasets of the spectra of stones a valuable method is Soft Independent Modelling of Class Analogy (SIMCA). SIMCA is a PCA-based model particularly useful for the classification of unknown objects according to independent categories. Every category identified is plotted against the unknown in a local PCA to calculate the distance to the model (DmodX). Each observation is classified according to this distance (Eriksson et al. 2013). SIMCA has been used for classification in two of the four case studies (Papers III and V).

5.2.4 Partial Least Squares Regression and combined matrices (PLS and O2PLS)

Regression is commonly used in statistics for evaluating the relation between variables. Projection to Latent Structures by means of partial least squares (PLS) is a common method for correlating two datasets with noisy variables. This model is useful for datasets where the number of independent variable is larger than the number of data points. PLS supplies diagnostic tools for interpreting the regression model.

Orthogonal PLS (OPLS) is also used for regression analysis while O2PLS is useful for data fusion (Eriksson et al. 2013; Trygg 2002). O2PLS is generally used for spotting common variability in two datasets collected with different analytical instruments. This model is particularly useful for understanding which information

contained in two blocks overlap. It has been used in two case studies (Papers III and IV) to compare NIR and XRF spectra (see Marini et al. 2016 for another example of combination of NIR spectroscopy and XRF).

5.3 Maps and annotations

Both in field and laboratory settings the data collected varied: single artefacts or entire rock outcrops correlated to stratigraphic information from excavations (horizontal) or architectures (vertical). The implementation of these data in a consistent digital platform was challenging due to the variability of geometry, topography and reference scale. The system has been adapted to each and every case, creating platforms of deep mapping to highlight specific material information about the artefacts. Support for the mapping has differed, two or three dimensional, according to the specific tools available and the analysed features. Photogrammetry has been applied in order to record the morphology of artefacts on different scales: to capture the topography of large architectures, in the case of Carcassonne's city walls, or to describe the small details of toolmarks in the case of the quarry in Montescaglioso. Due to publishing constraints, the models have mostly been reduced to orthophotos, also useful during the documentation phase. In fact, photogrammetry has been a reliable tool for creating models on which location of NIR and ED-XRF measurements could be precisely annotated. Moreover, in certain cases, the texture of the models could be replaced by the colour composite images in order to visualise the spectral information in its masonry context (Fig. 12).

Geographic Information System (GIS) platforms have been used for some of the case studies. In the case of the Mesolithic dwelling site in Lillsjön, data from the excavation have been implemented in a 3D GIS platform including information about the stratigraphy, geochemical characterisation of sediments, dated materials and finds (more details can be found in the next chapter). Handling data with a relevant height value (Z), the use of ArcScene resulted in a much more effective visualisation (Berggren et al. 2015; Dell'Unto et al. 2013, 2016).

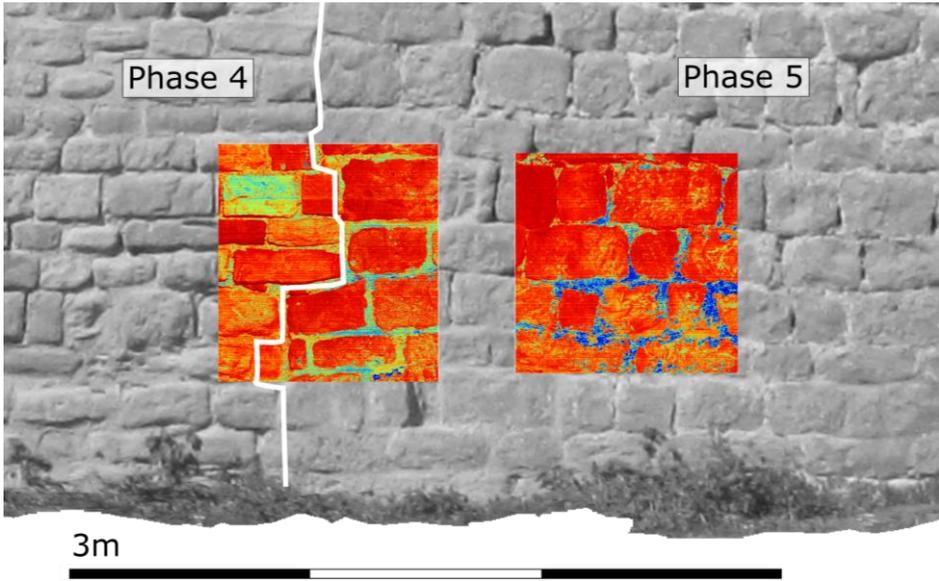


Figure 12. Portion of the inner wall of Carcassonne in an orthophoto. The hyperspectral images have been replaced in the corresponding spots and highlight some peculiar details of the chemical characteristics of building materials. Phase 4 and 5 correspond to two moments in the life of the construction (see Paper III for details).

6 Papers

Each of the case studies in this thesis present specific characteristics: different types of materials, archaeological context and research background. The published and unpublished papers collected here have been written with a focus on analytical procedures and details about data processing. In this chapter, the goals and arguments of every paper will be summarised to highlight relevant novelties, theoretical implications and interdisciplinary approaches.

The papers are listed according to a scale of inquiry, providing details of analytical methods and their application in each scenario. The first article concerns a pilot study carried out in the early stages of the project. The paper is specifically aimed at demonstrating the feasibility of on-site and non-destructive red painting characterisation. The following papers (Paper II, III, IV and V) seek to strengthen the link between analytical data and the archaeological context, combining near infrared spectroscopy with other analytical techniques. The second and third papers consider the study of Mesolithic stone tool raw materials at a dwelling site. The possibility of performing a geological characterisation of artefacts is assessed in Paper II, while in Paper III the distribution of the raw materials of artefacts is combined with site topography, demonstrating how the geography of materials can be informative about the use of space in a prehistoric context. The fourth paper is about a large project that is aiming to develop a protocol for the characterisation of building materials in standing structures. The case study is the fortification wall of the citadel of Carcassonne. The construction and restoration of the wall has been extended over several centuries and the characterisation of building materials has shown how different supply strategies have been adopted in different socio-economical contexts. The last paper focuses on a landscape archaeology project in the area of Montescaglioso (Basilicata, Southern Italy). A protocol for survey documentation of stone artefact was created in order to collect morphological and analytical data on artefacts directly in the field. This technique helps the process of gathering data on a larger scale, outlining landscapes of stone exploitation.

Each paper included in the appendix is discussed here, highlighting important outcomes and results. For every case study a brief context is supplied together with some supplementary data that could not be included in the papers but that are relevant for understanding the archaeological context. In the following table (Table 2) each paper is illustrated according to the scale of analysis, methods used for characterising the materials and for documenting/analysing morphologies and topographies.

Table 2. List of papers in appendix organised according to scale and methods used for mineral characterisation, documentation and data analysis.

Paper	Site/s	Scale of the study	Analytical techniques	Documenting techniques
Paper I	Flatruet rock paintings	Rock paintings panel 3–4 m ²	MicroNIR probe	Photogrammetry and tablet documentation to record figures and shape of the rock face
Paper II	Lillsjön Mesolithic dwelling	Dwelling feature and immediate surroundings 1,000 m ²	Hyperspectral imaging	
Paper III	Lillsjön Mesolithic dwelling	Dwelling feature and immediate surroundings 1,000 m ²	Hyperspectral imaging and ED-XRF	3D GIS to analyse distribution of lithics according to raw materials classes
Paper IV	Carcassonne	Citadel's defensive wall, about 3 km long	Hyperspectral imaging, ED-XRF, MicroNIR probe	Photogrammetry to record the variability of masonry
Paper V	District of Montescaglioso	Surveyed area, about 50 km ²	MicroNIR probe	Photogrammetry to record surface morphology and to document toolmarks

6.1 Stones and mind: rock paintings in Northern Scandinavia.

The first paper is particularly important as it constituted an experimental study of the on-site analysis of archaeological materials using a MicroNIR probe. The study has been published in a specialised journal (Journal of Near Infrared Spectroscopy) with the goal of setting the specifics for sampling archaeological features in surveys and processing data. The paper considers issues and potentialities in performing *in-situ* NIR analysis of earth-based pigments on a stone background and discusses statistical models for highlighting trends and clusters in a data matrix comprising measurements performed on both the painted surface and the background rock wall. This method is discussed using a case study as an example, namely, the rock paintings in Flatruet.

The study area is situated on the southern point of the Flatruet Mountain in Härjedalen, Central Scandinavia (Fig. 2), with a lithology characterised by feldspathic sandstone, meta-arkose, metaconglomerate, marble, metatillite (Neoproterozoic) (Geological Survey of Sweden-SGU). The site at Flatruet is situated on a cliff (about 850 m high) overlooking the valley below. Contrary to most Scandinavian rock art sites, the panel in Flatruet is not located in proximity to water. This site, situated on the eastern limit of the ridge separating Sweden from Norway, surmounts the mountain pass and could have been a reference point for ancient groups moving

across the land and perhaps following animal migration routes (Lahelma 2008). The site consists of a painted panel featuring around 30 figures (20 animals, 5 human figures and 5 indeterminate, Fig. 13). Among the animals are elks, reindeer and one bear. A small excavation was made in front of the painted outcrop by A. Hansson (Hansson 2006; Sognes 2008). In the investigation, a few broken quartz arrow points were found and interpreted by archaeologists as ritual projectiles. The arrow points could have been fired at the painted figures as part of a propitiatory ceremony for hunting (Hansson 2006) or to harm the spirit that dwelt in the mountain (Lahelma 2008). The objects have been dated on typological bases to the Early Bronze Age (approx. 1500 BC), while traces of fires have provided charcoal fragments dated from 4000 BC to 1200 AD. The figures appear to have been painted on more than one occasion and have been dated to the periods described as Forsberg's phase II, III and IV, a chronology generally referring to 3000–2000 BC (Forsberg 1993; Sjöstrand 2011; Sjöstrand 2012).



Figure 13. Detail of the painted figures in Flatruet.

Several interpretations have been proposed for the scene: according to Hallström (Hallström 1960), the paintings on the cliff depict a hunting scene, while Lindqvist (Lindqvist 1994) supports the idea that the human figures represent shamans. Most of the depictions can be found in the central part of the composition, sometimes overlapping, and are generally painted with a red-coloured earth-based

pigment (more details on the materials can be found in Paper I, p. 227). Elks and reindeer in the central and southern portion of the scene measure around 18–29 cm in width and are 8–15 cm high. The various shapes on the rock surface are characterised by stylistic traits: the majority of the figures are just outlined; the figures' contours are generally between 0.8 and 2.5 cm wide (with a few washed out depictions in which the traits are no longer measurable); the elks on the central and right-hand part of the panel have lines characterising their bodies; some of the animals have straight legs while others are depicted with bent limbs. Measurements using the MicroNIR probe were conducted in several spots for each painted figure, collecting a dataset that was comprehensive of the entire panel. The study demonstrates that spectral information pertinent to the painting can be separated from the geological background. The article shows in detail how the spectral features can be used as fingerprints for characterising single stretches of painting according to their pigment's composition. Moreover, spectral measurements can be used for distinguishing painted figures from each other and for characterising single stretches in one image. This is particularly evident when looking at the PCA clusters in Fig. 8 in the paper (p.233), in which a few selected depictions have been analysed in detail to show similarities and dissimilarities in the pigments used. The results of this study are significant from a methodological perspective of measuring rock painting with non-destructive and portable instruments. The potentialities of this approach can be demonstrated by linking the NIR data to the typological interpretation of the panel and the historical context in which it has been painted.

6.2 Stones and place: lithics classification and intra-site distribution in a Mesolithic semi-subterranean dwelling

The second case study presented is a Mesolithic dwelling site located in Ångermanland (Northern Sweden). The study of this site forms part of a larger research project being conducted at Umeå University to outline settlement dynamics during the early phase of occupation of northernmost territories in Scandinavia. Two papers included in this collection focus on the study of lithic tools gathered during the excavation of this prehistoric dwelling. Paper II is part of a peer-reviewed edited volume containing the proceedings of a conference held in Faro (Portugal) in March 2016 (Pereira, Terradas, and Bicho 2017). The short article demonstrates that hyperspectral imaging can be used to characterise different types of raw materials, in this instance, quartz, quartzite, slate and flint, which were scanned using a SisuCHEMA Pushbroom short-wave infrared camera. Spectral peaks characteristic of each material were indicated and the classification was performed through

multivariate statistics. Paper III is based on these preliminary results to complete a broader analysis of lithic raw materials and procurement during the occupation of the dwelling. The site is situated on the shore of the lake, at Lillsjön, and is labelled as RAÄ 260 according to the classification system of Swedish heritage sites. The large elliptical semi-subterranean dwelling is not the only feature of this kind to be found in the area, in which several large huts have been identified and mapped. Although a few of these sites have been excavated (Lundberg 1997; Olofsson 2003), the precise dynamics of the use of space within the dwellings are still partially undefined.

The site, RAÄ 260, was investigated during two different campaigns in 2010 and 2012 by a team of researchers and students from Umeå University as part of a Master's programme in environmental archaeology. Examination of the area around the Anundsjö river system was undertaken with a specific focus on environmental issues, highlighting how the so-called "pioneer" settlers in the north adapted to the local environment and made use of natural resources. One of the research project targets was to monitor site preservation studying the mechanical disturbance caused by forest ploughing and other maintenance practices conducted by the state forestry management department. During the excavation, soil samples and finds were collected on 5 cm deep spit levels on a grid of trenches. The soil samples (842 in total) were analysed using different methods (ED-XRF, total organic content, total phosphate content, Inductively Coupled Plasma optical emission spectrometer¹³) in order to gather information about geochemistry and record ephemeral traces of human activity and soil formation processes. The finds were divided into categories of faunal remains, fire-cracked stones, and lithics and quantified according to type and volume. All the data collected were stored in a database associated with a 3D GIS platform, created in ArcScene and representing the volumes of the excavated trenches¹⁴. A considerable quantity of lithics (2612 objects) was collected during the excavation. The stone fragments, primarily made of quartz and quartzite, were scanned using a hyperspectral imaging system and the images were processed to extract an average spectrum for each object. The dataset was then classified, applying PCA-based models, in four classes of raw materials, two types of quartz and two types of quartzite. The hyperspectral screening of lithics was conducted as a semi-automated process and resulted in a descriptive spectral dataset representative of the material properties of each fragment.

¹³ All analyses of soil samples were carried out by Guido Mariani (PhD) during his intership at the MAL laboratory in spring and summer 2016.

¹⁴ Niccolò Dell'Unto and Giacomo Landeschi from the DARK Lab at Lund University are to be acknowledged for their help in developing the 3D GIS platform. The database was implemented by Lorenza La Rosa during her intership at the Mal laboratory during the years 2016–2017.

The geological configuration of the area close to the site is such that an accurate location of sources is virtually impossible. In fact, as shown in Fig. 14, the surrounding bedrock formations contain quartz and quartzite associated with different lithologic facies. Moreover, the area in which the site is located is characterised by moraine deposits made up of pebbles of assorted materials. In the surroundings of the site, quartz and quartzite are available in various forms and the procurement activities could have covered quite large areas. Even though it seems difficult to establish material supply pathways, the characterisation of raw materials and their distribution within the dwelling reveals information about occupational patterns at the site.

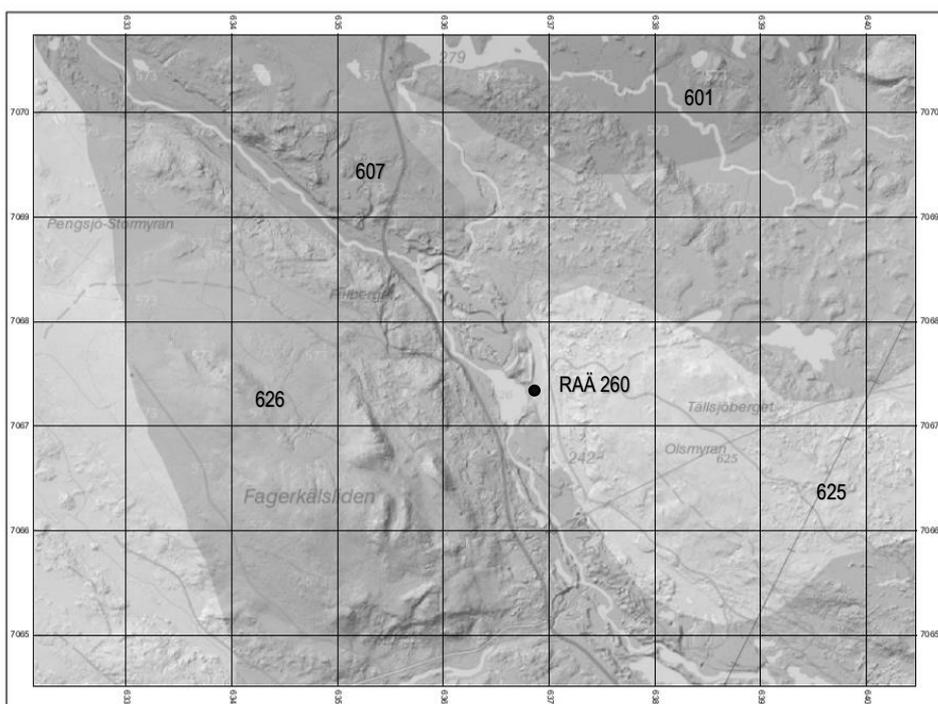


Figure 14. Geological map of the study area. Scale 1: 50000¹⁵. Geological information from the Geological Survey of Sweden (SGU, www.sgu.se); map ©Lantmäteriet.

¹⁵ 626: Granitoid and subordinate gabbro, metamorphic; Svecokarelian orogen; Bothnia-Skellefteå lithotectonic unit; 607: Granite, pegmatite; Svecokarelian orogen; Bothnia-Skellefteå lithotectonic unit; 625: Metagreywacke, mica schist, graphite- and/or sulphide-bearing schist, paragneiss, migmatite, quartzite, amphibolite; Svecokarelian orogen; Bothnia-Skellefteå lithotectonic unit; 601: Granite, granodiorite, syenitoid, quartz monzodiorite and metamorphic equivalents; Svecokarelian orogen; Bothnia-Skellefteå lithotectonic unit.

The data on the spectral characteristics of lithics were implemented in the GIS platform and visualised in the tridimensional reconstructed record. The 3D visualisation shows how clusters of different types of quartz and quartzite appear to be associated with at least two different phases of occupation of the site. The category distribution of the raw materials shows evidence of a change in the supply of stone for lithic tools: different types of quartz and quartzite have been used throughout the lifespan of the site. The reason for the change in material usage during the Mesolithic period is uncertain. However, there appears to be a link between the passage of time and transitions in the selection of supplies that certainly provide a clue for understanding the behaviour of ancient settlers. This change could be linked to larger scale dynamics of adaptation and procurement of raw materials. This hypothesis could be confirmed by adopting a similar approach to the analysis of lithics at other dwelling sites and comparing the results.

The two studies are certainly valuable as a demonstration of the potentialities of the application of near infrared-based analysis of lithic sets. Moreover, Paper III demonstrates that including large-scale materials analysis in archaeological investigation can help understand the record, drawing attention to the links between humans, technology and raw materials.

6.3 Stones and time: stories of building materials in the walls of Carcassonne

Paper IV deals with the characterization of building materials used in the inner defensive wall in Carcassonne (Aude, France). The citadel of Carcassonne has been inscribed on the UNESCO World Heritage Monument List since 1997. The site is now visited every day by a huge number of tourists and has been taken over by souvenir shops and various attractions with a medieval theme. The commercialisation of a mythical idea of the Middle Ages does not leave much space for a more complex archaeological narrative that encompasses the long history of the city, its inhabitants and the landscape. A 2-years study (collective research project- PCR) of the military architecture of the fortified citadel, was aimed at studying architecture, archive documents and building materials (Allios et al. 2016; Faucherre et al. 2016). During the project it was possible to collect new archaeological data to define the long-term processes that contributed to shaping the fortress in the way we are able to observe it today. The material sources for reconstructing past events that took place on Carcassonne hill were primarily the architectural remains. A detailed archaeological analysis of the standing structure of the citadel is a long and complex operation, based on the observation of masonry and the documentation of building techniques. The starting point for this operation

was the systematic analysis of the inner defensive wall showing several phases of construction spanning from the Roman period-late antiquity to contemporary restorations. The wall is more than three kilometres in length with a variable height. The materials used in the masonry could be described visually according to colour and particular geological features, typical of sandstone formations. Nevertheless, a more accurate characterisation of stone types would allow a greater understanding of the dynamics of materials supply during the construction phases and contribute to a general understanding of natural resources management in the region.

Taking into account technical issues resulting from the size of the monument, a new experimental approach was tested, combining various non-destructive methods for performing *in-situ* characterisation of building materials. Hyperspectral imaging, portable ED-XRF and the MicroNIR probe proved to be effective tools for analysing masonry elements and distinguishing between the types of raw materials used in the construction. The study presented in Paper IV represents a methodological test that allowed the formulation of a number of hypotheses on the organisation of the working site. The dataset collected will be implemented in the future with additional spectral analysis of stone elements in order to delineate and quantify geological materials that are used on a large scale.

The experiment also highlighted the importance of associating information on the materials with the archaeological analysis of standing structures in order to understand construction procedures and the link between the ramparts and potential quarrying sites in the surroundings. Spectral data collected in a subsequent phase of the project demonstrate that a link exists between the construction campaigns and transformation of the landscape, in particular of Carcassonne hill. The bedrock morphology appears to have been modified in the upper portion, close to the wall's foundations (Fig. 15). At several points around the fortress we were able to document outcrops of sandstone showing quarrying marks and proving that local bedrock had been used as a material for the construction.

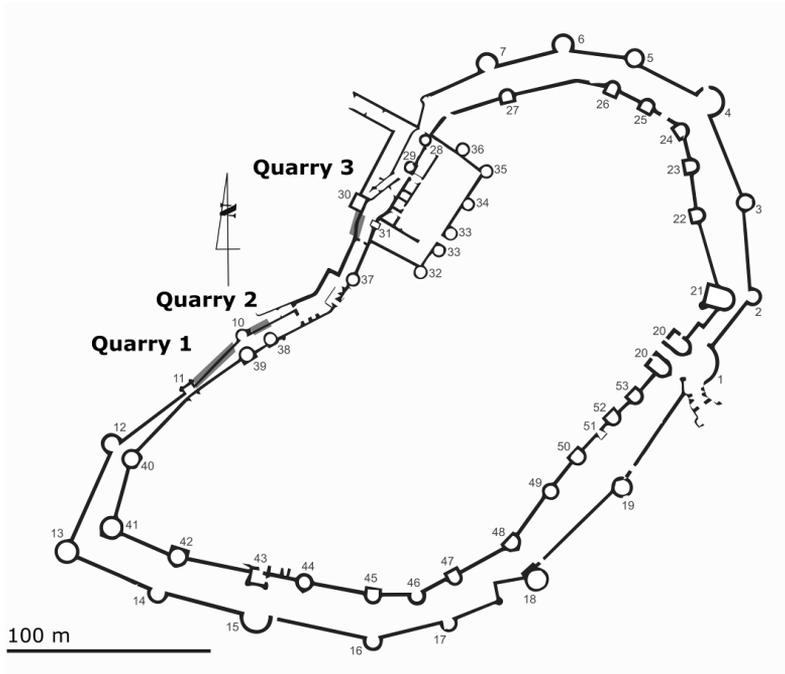
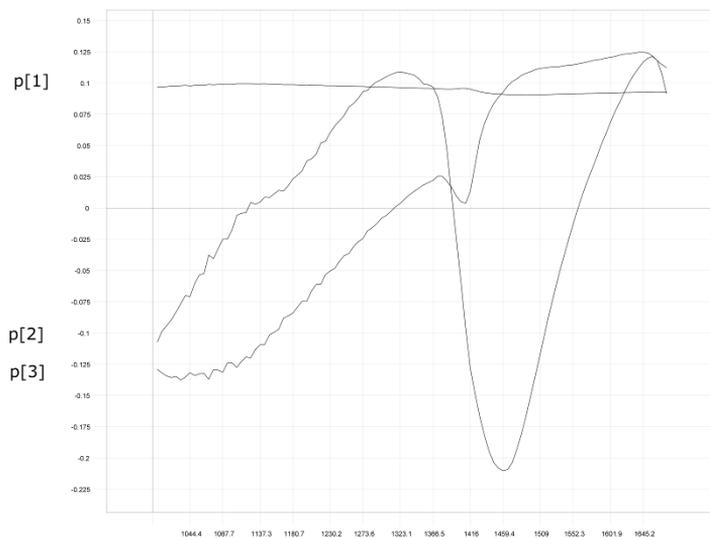
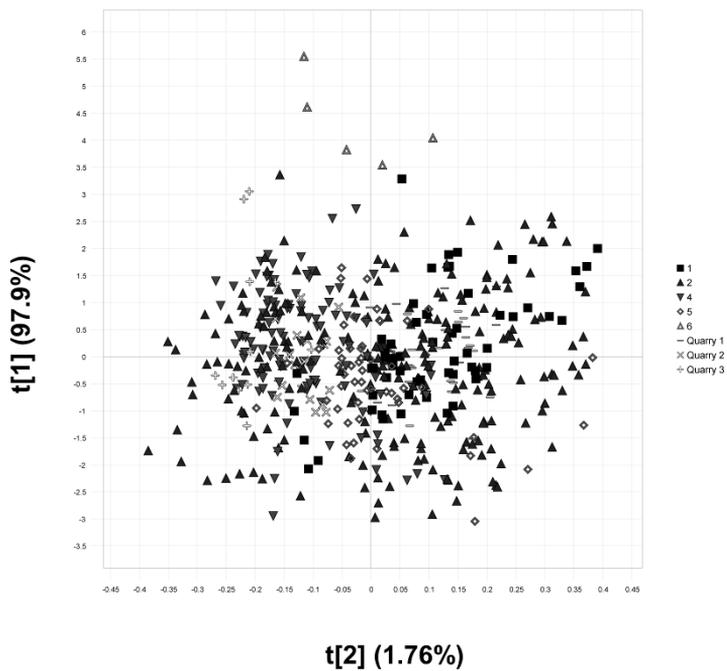


Figure 15. Position of the sandstone outcrops with quarry marks, analysed using ED-XRF and MicroNIR.

The size of the exploitation is not visible today and has been partially hidden by later sediments. A medieval faubourg, later destroyed during the Albigensian crusade, existed in the area that currently appears as a smooth hill. The presence of dwellings could have resulted in the use of outcrops as quarries, supplying building materials on site. This hypothesis has been reinforced by spectral measurements performed using MicroNIR at the quarry sites and suggests a strong similarity in the composition with some of the raw materials used in the masonry. Considering the position of the outcrops, which are rather difficult to approach, measurements could only be performed with instruments that were easy to carry (the hyperspectral camera could not be used). It is therefore difficult to make a complete comparison with the rest of the data collected for the case study presented in Paper IV. The PCA plot of MicroNIR measurements of three outcrops that display quarry marks and measurements collected on the masonry show interesting trends (Fig. 16). Elements of phases 4 and 1 in particular appear to overlap with measurements performed on quarry 2/3 and 1, respectively (phase 1 corresponds to the most ancient evidence of construction during the Roman period/late antiquity, phase 4 to 11th/12th century; see the chronology of phases in detail in Paper IV).



Line

Figure 16. PCA and loading line of MicroNIR measurements collected on masonry elements and quarry outcrops in Carcassonne.

The dataset collected in Carcassonne is still incomplete and the data patterns should be considered a circumstantial evidence rather than a final result. Nevertheless, the data show how the information on materials can contribute to the historical narrative. It demonstrates that the morphology of a site admired by hundreds of thousands of tourists is the process of a slow transformation of landscape and societies: while the construction changed over time according to different historical events, the landscape was also transformed. The cycle of use and reuse of materials can be traced through extraction, construction of the walls, destruction and cleaning of debris thrown down the slopes. All these stages entail a great transformative power on the landscape and urban network, impacting, through the construction, the entire district of Carcassonne.

These various aspects of materials were also echoed in the large installation realised by Felice Varini called *Cercles Concentriques Excentriques*. The work of art, visible on the ramparts from 4 May to 30 September 2018, was constructed using the concept of anamorphosis, a perspective distortion of a geometric composition. A spectator standing at a vantage point at *La porte d'Aude* (the Aude gateway) could admire the shape of thirteen yellow concentric circles projected on the architecture (Fig. 17).



Figure 17. Preparation of the installation *Cercles Concentriques Excentriques* by F. Varini. Photo by D. Allios.

Moving around the citadel and changing angle, the spectator could discover contrasting perspectives in which the form is fragmented and distorted. The geometry was created by drawing circles projected from a light source and filling the

sections with painted aluminium sheets. The flexible material adapted perfectly to the ridged surface of the masonry, without giving the impression of flattening the volume, but rather enhancing it, adjusting to the various holes and cracks. The yellow geometry was not conceived as a work of abstraction but rather as a celebration of the single stone blocks that form part of a multifaceted historical object. Varini's work does not just overlap on the architecture but is rooted in it, allowing the spectator to discover the monument from different angles, accompanied by the yellow stripes.

Varini's perspective has been constructed to be observed from the gateway that links the citadel to the modern city, a vantage point often chosen by Viollet-le-Duc for his drawings. The famous architect was commissioned in 1850 to lead the citadel's restoration project, resulting in an extensive reconstruction of part of the walls (Viollet-le-Duc 1881). It is probably not a coincidence that *La porte d'Aude* is the perspective from which the shape of Varini's circles has been assembled and Viollet-le-Duc's vision of medieval architecture is often displayed. When leaving "La porte d'Aude" to explore the wall's circuit, the visitor will discover that the citadel is far from having an ideal architecture. However, the entire citadel comprises a multiplicity of masonry fragments, stretched and deformed by time and events – stones piled up over the centuries that form one of the most famous sites in Southern France.

6.4 Stones in the landscape. Mapping the production and circulation of Calcarene artefacts

The aim of Paper V was to map and describe a landscape of stone exploitation and trading in artefacts in Basilicata, Southern Italy, in a territory marked by the presence of limestone cliffs, canyons and hills. In this stretch of mountainous landscape, between the town of Matera and the Ionian coast, several settlements and production sites from the 6th to the 3rd century BC have been mapped. A dense network of archaeological evidence delineates the pattern of the Greek colonization of Southern Italy, in a territory already inhabited by an indigenous Lucanian population. The goal of the CHORA project (CHORus of Resources for Archaeology), led by the Graduate School of Archaeology, University of Basilicata, is to collect information about the ancient occupation and its relationship to the landscape. The project aims to compile data from various proxies to describe the dynamics of habitation and production in the region, adopting an interdisciplinary approach. The case study presented in Paper V focuses on the understanding of the production and circulation of Calcarene artefacts and is included in the larger experimental landscape archaeology project. The study involved the evaluation of

the usability of a portable NIR probe for collecting spectral information during surveys. The area sampled was the territory around the town of Montescaglioso, where a few sites with stone artefacts had already been identified from previous research.

The experiment was conducted in order to test the use of MicroNIR in field surveys in an area quite rich in various stone artefacts, still visible and scattered on the surface, marking ancient settlements. This particular configuration is partly due to the relatively light population in the area around Montescaglioso. The district is actually situated in the core of a region, Basilicata, in which the process of urbanisation came rather late, compared to other areas of Italy. In this area, it is still possible to observe marks of past land use through the cultivated crops and ancient artefacts that have been integrated into contemporary rural life. Stone coffins re-used as flower pots, millstones as building materials, quarries as landfills – all coexist together with archaeological objects unearthed during excavations and now left visible, circumscribing ancient and new spaces. In this scenario, the use of MicroNIR appears to be effective for collecting information about artefacts that are sometimes found with no clear archaeological context. This portable and fast analytical method can provide important data about the characteristics of materials, tracing their origins, uses and trajectories.

The extraction of stone blocks and the production of various artefacts are an essential part of the history of the territory around Montescaglioso, an essential proxy to understanding the transformations of settlements and landscape. Acquiring spectral data of materials means disclosing information about scales of manufacture, distribution and trading network but it also highlights the role these objects play in the metamorphoses of landscape and its actual configuration. Integrating the characterisation of materials into survey operations permits the collection of important data. Nonetheless, measuring artefacts in field conditions can present problems. Weathering is a major obstacle for performing in-field measurements and finding an unaltered spot to make recordings of an artefact or outcrop can be difficult. The strategy adopted was to also perform NIR measurements on altered portions of stones, creating a referential dataset and compiling information on chemical and biological activity on the rock surface (the difference between altered and unaltered rock surfaces is further discussed in Chapter 8). In future developments of the CHORA project this analytical method will be applied on a larger scale, compiling a dataset that includes the spectral signatures of many more artefacts and outcrops. The area of study will be enlarged, including the territory of relevance to the Greek colony of Metaponto, as well as some medieval settlements,

in order to illustrate the dynamics of extraction and manufacture of stone artefacts in a large area throughout the centuries.

The case study described in Paper V was presented at the international conference YOCOCU 2018 (Youth in Conservation of Cultural Heritage) and will be published as part of the conference proceedings.

8 Discussion

During this research, several methods have been combined to reach different scales of accuracy. Working with a diverse set of case studies helped to pinpoint the potentialities and limitations of the proposed approach. NIR-based analytical tools have proven to be a reliable support for the analysis of archaeological materials in various situations.

The MicroNIR is a perfect tool for surveys, extremely portable, resistant and performs very well in quite extreme conditions. Nevertheless, the wavelengths that can be measured with this instrument are limited, creating difficulties in the identification of specific minerals. The bands measured are between 900 and 1,600 nm, including the third, second and part of the first overtone regions. The peaks registered in these bands are usually quite broad and result in several overlapping molecular bond vibrations.

The size of the MicroNIR captor could also add a degree of uncertainty to the measurements. It might actually make it difficult to precisely position the window (4x2 mm, placed in the centre of a 4 cm wide holder) on small targets (as was the case in the rock paintings in Paper I).

Weathering and biological activity on the rock's surface could interfere with measurements, particularly in field conditions in which these phenomena are more frequent. Nevertheless, an accurate sampling of the surface for analysis allows for good reliability of measurements. Fig. 18 shows an example of measurements on white and dark coatings on limestone (calcarenite) blocks in Montescaglioso (described in Paper V). On the top, the actual state of alteration of the masonry with ongoing chemical and biological activity; in the centre, a PCA model calculated on measurements performed on altered and unaltered spots shows that the weathering can be distinguished from the other measurements. It is easy to detect lichens or other organisms on a stone surface through infrared imaging because of the clear response of organic materials to infrared bands. However, the characterisation of chemical weathering can be more complex and depends on the type of processes involved and the level of analysis (pixel resolution sufficient to spot micro accumulations of salt or other contaminants).

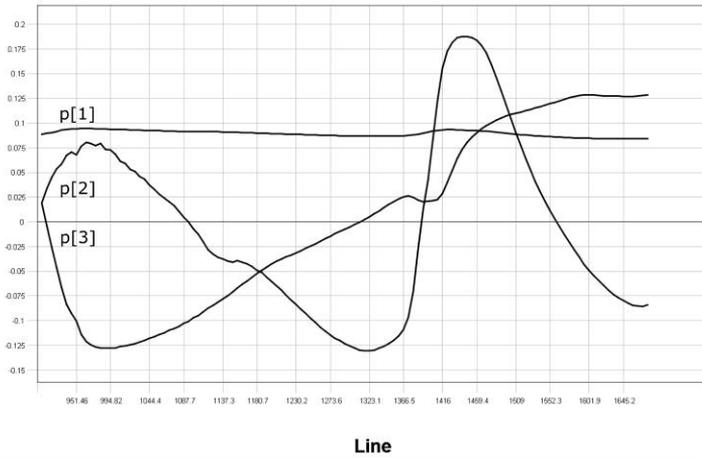
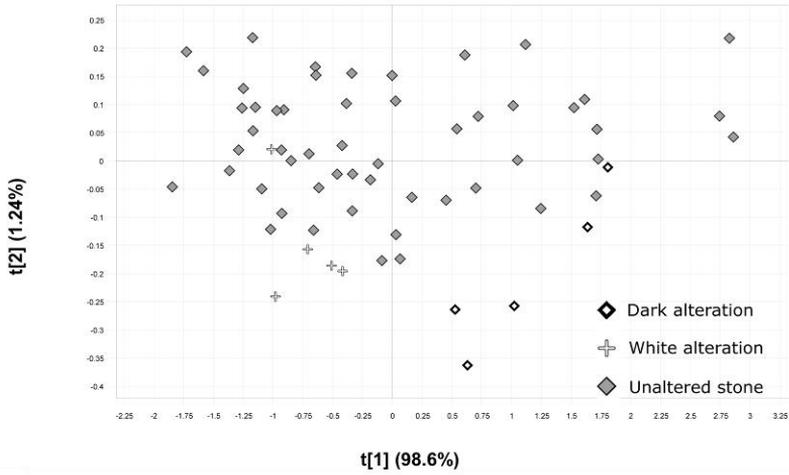


Figure 18. Portion of the southern wall analysed in Montescaglioso (Paper V). The limestone blocks present superficial alterations that have been measured using a MicroNIR. In the centre is a PCA model of black/white coatings and unaltered stone.

Hyperspectral cameras allow the rapid acquisition of image recordings, delivering complex and rich data matrices. Although this instrument is relatively easy to use and calibrate and also performs well in a controlled environment, the use of hyperspectral cameras in the field can present some technical issues. The camera used for the study of the citadel's walls had to be transported in a van (already limiting the portability of the instrument) and had to be installed on a system of rails powered by a portable power unit. The recordings collected using the hyperspectral camera include more spectral bands than those obtained using the MicroNIR, with vibrations in the first overtone and combination bands. Many peaks that indicate the mineralogical composition of rocks are located in the interval between 1500 and 2500 nm, making the hyperspectral recordings more valuable for characterising stone types and particular differences in the molecular composition of materials.

Hyperspectral images contain a series of contiguous spectral data, helpful for mapping the surface of alteration in rocks. Fig. 19 shows the detail of a masonry element in Carcassonne's defensive wall. The stone block presents some superficial alteration resulting from exposure to heat. For several years, this portion of the wall was enclosed by a dwelling constructed in the space within the two concentric walls. A stove or oven in the house was probably located adjacent to the wall and left traces of fire directly on the sandstone blocks. These days there is almost no remaining trace of this house except for the superficial alteration on the masonry. The image on the top shows a PCA model calculated on the hyperspectral image with a darker area in the right-hand lower corner (the grayscale indicates the $t[1]$ values). Below, a PCA scatter plot calculated on the same spectral pixel values shows two distinct clusters, one indicating the unaltered sandstone (on the right) and the other indicating the fire-altered portion (on the left).

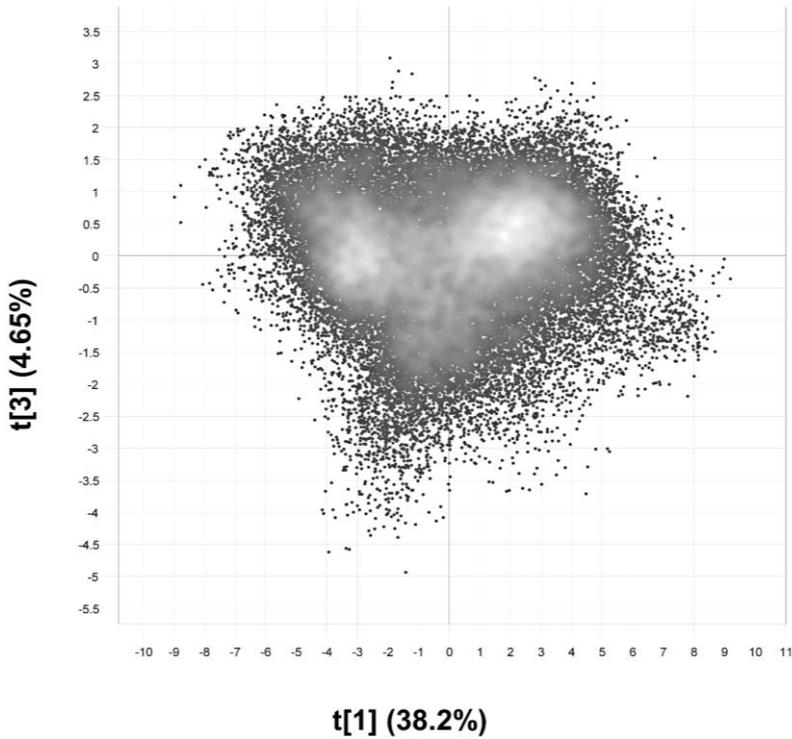


Figure 19. Detail of an element in the masonry of the defensive wall of Carcassonne analysed using a hyperspectral camera. On the left, PCA on the image, the $t[1]$ values are represented as a grayscale. On the right is a 2D score plot of the pixels in the image. Intensity values are represented as grayscale.

Measurements performed using ED-XRF proved to be less significant than NIR spectroscopy in characterising stone types. The in-built calibration system of the Niton XL3t was not considered to be suitable for analysing quartz/quartzite (in Paper III) and sandstone (in Paper IV) and a fingerprint approach was adopted. Moreover, the sampling strategy chosen for the measurements was adapted to the strategy used for NIR measurements in order to compare the results. Portable XRF instruments have been proven to deliver a rather high background signal and have scarce sensitivity to trace elements. ED-XRF was used to measure rather small quartz/quartzite flakes (Paper III) and different sandstone types (Paper IV). In both cases classification was performed according to the main elements and was not accurate enough to distinguish between stone types that were so similar.

As argued, uncertainty in classifying objects can arise from the material properties themselves, such as weathering, contamination due to taphonomic processes in buried records, or the detection limits of analytical tools. Statistical models can help to process ambiguous data, adapting the algorithms to the characteristics of the datasets. The statistical models used for processing the collected datasets have been chosen by adapting to the targets. PCA has been shown to be a useful screening tool in every case study for spotting patterns and similarities between the analysed objects. After data reduction and evaluation, the choice of classification algorithms has been diversified according to the data types.

In Paper I, the distinction between red painting and background was highlighted through the application of a PLS-DA (Partial least-square regression discriminant analysis) model. This algorithm is used to maximise the described variance between the two sets of measurements (in our case, the rock surface and the red figures). The application of this statistical model helped to enhance patterns that were not visible using a simple PCA model.

In Paper III we dealt with the classification of large volumes of materials, as well as overlapping and scattered categories. The classification was initially reduced to a subset in order to identify the mineralogical characteristics of objects and divide them into classes according to the clusters. The most optimal model for predicting the entire set of lithics was then considered to be SIMCA, in order to be able to assign items according to their distance to the pre-defined classes.

Methods used for recording morphologies and visualising analytical data have also been adapted to the research goal. Photogrammetry and 3D GIS are powerful tools that allow the storage of information on stoneworking techniques. This software should be used as a tool for data analysis and annotation support. However, tools for multivariate statistical analysis, including the algorithms described in Chapter 5,

have not yet been integrated into photogrammetry and GIS software. This technical limitation makes the full integration of morphology and spectral analysis impossible at present. Analytical results can be linked to the model but the evaluation and modelling of data cannot be performed directly on the 3D rendering. The development of a platform that allows real interdisciplinary analysis is a necessity in order to conduct data processing and visualisation of the molecular characteristics of stone artefacts directly on tridimensional models.

The theoretical discussion about the role of materials in shaping ancient societies may appear to have been somehow disregarded in certain papers included in this dissertation. This choice is in response to an editorial need to describe the details of a relatively new application of analytical techniques, leaving little space for discussing the theoretical implications. This introductory text aims to complete and correlate methods and data within a broader discussion about rock's agency. All the papers demonstrate how combining the methods of geochemical analysis and documentation is crucial for gathering data about the use of materials over time, outlining the set of tools for an interdisciplinary study protocol. The value of these methods has been tested in various contexts, each time moving towards a more informed integration of materials analysis and archaeological practice. The application of NIR-based analysis paves the way for the characterisation of stone artefacts in disparate contexts, enhancing the significance of materials science in archaeological research.

9 Conclusion

Understanding the association of human societies and geological resources can help archaeologists shed light on past behaviours. However, reconstructing the relationship of past societies with geological resources is a complex endeavour. The diachronic evolution of the link between humans and materials can be outlined by deconstructing modern cultural references and reassessing the correlation between communities and natural resources. The process of decolonising materials is not easy and can only be conducted through a meticulous collection of archaeological data in different chronological and cultural contexts. An interdisciplinary approach that is focused on collecting information about the geological characteristics and cultural meaning of rocks is necessary in order to understand the ecological system of object modifications and cultural transformations. A flexible approach to materials analysis could help to disclose more information about processes of supply, manufacture and disposal (see diagram in Chapter 3). A large-scale analysis of data collected using the outlined procedures could reveal trends in the use of materials that depend on cultural and economic factors.

This thesis represents a first step towards acknowledging the agency of rocks in the development of communities, showing how the availability of materials can influence cultural choices and that supply can be a symptom of societal transformations. A systematic implementation of analytical, typological and ethnographic data could provide datasets for assessing larger scale dynamics of the exploitation of geological resources and shed light on the long-term alliance of humans and rocks. All the case studies presented highlight how a diversified supply of raw materials could correspond to a transition, a change in lifestyle for the people living at the sites; in some cases, the modification is linked to transformations within the sites (as is the case of the political change in Carcassonne), or in the surrounding environment.

In this introduction and the attached papers, it is argued that the adoption of new analytical procedures could support the acquisition of geochemical data on archaeological artefacts. Various applications of NIR spectroscopy combined with tools for documenting an object's morphology have been reviewed and discussed, highlighting the advantages and disadvantages. The use of NIR spectroscopy to characterise inorganic archaeological materials is quite unique, especially when considering the fact that the technique is primarily used in the study of organic substances. NIR spectroscopy offers a range of interesting options: portable spectrometers and imaging systems are versatile instruments that can be used as

short-range remote sensing tools both in the laboratory and in the field. The technical peculiarity of this type of spectroscopy makes it possible to distribute the measurements on the surface of the objects, allowing a diffuse evaluation of chemical properties and the direct observation of their variability. The adoption of a more extensive and adaptable sampling strategy helps reduce researcher bias and allows the collection of a different range of information. Visualising and exponentially increasing the number of measurements performed allows the construction of an analytical protocol that adequately represents the variability of chemical-geological information, including alteration and contamination.

Monitoring the weathering and deterioration of stone artefacts is an interesting research output for NIR-based instruments. In the case study described in Paper V, a number of reference measurements were collected on altered portions of an object's surface. By repeating these measurements after some time has elapsed, we could attempt to establish if the deterioration has progressed and also determine the weathering rate of the exposed limestone artefacts. Recordings conducted with portable instruments could help in planning the preservation and restoration of individual objects and monuments, providing important data for cultural heritage management.

This thesis shows that the application NIR spectroscopy for characterising stone artefacts can deliver interesting results. However, their application depends on good knowledge of the spectral signatures of various materials, as well as statistical methods of data reduction. The experience has not been without issues and the creation of an analytical protocol stems from a thorough experimental design. The analytical protocol was elaborated after repeated observations, considering methodological constraints and various approaches.

The application of NIR technology represents a viable resource for studying the intangible strands of an artefact's biography. Like other digital analytical tools, NIR spectroscopy can help record the traces of past events registered in the mineral characteristics of a stone artefact. Data collection, processing and interpretation is the challenge to which researchers must now respond.

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