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Alessandro Aleo

Assessing and comparing technological complexity of Neanderthal and modern human adhesives

ASSESSING AND COMPARING TECHNOLOGICAL COMPLEXITY OF NEANDERTHAL AND MODERN HUMAN ADHESIVES

Dissertation

for the purpose of obtaining the degree of doctor
at Delft University of Technology
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by

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Contents

Summary

Adhesives are essential components of everyday life and have been for thousands of years. The history of adhesives begins with a stone tool covered in birch tar found at Campitello Quarry, Italy, dating to around 200,000 years ago. This find demonstrated the use of adhesives by Neanderthals and their ability to manufacture materials through transformative processes. In Southern Africa, modern humans have been producing compound adhesives by mixing plant and mineral materials since at least 70,000 years ago, tailoring adhesives to different environments and uses. In recent years, the number of identified prehistoric adhesives has grown, and adhesive technology has become a proxy for discussing technological complexity across different hominin species. However, to fully evaluate and compare the adhesive technology of other human groups across space and time, more research is needed on the selection of adhesive materials, adhesive recipes, and the context in which adhesives and their tools were used.

With this dissertation, I contribute to enlarging the sample of identified prehistoric adhesives by analysing tools with adhesive residues from Steenbokfontein Cave (South Africa, Later Stone Age), Morín Cave (Spain, Middle-Upper Palaeolithic), and the Dutch North Sea (the Netherlands, Mesolithic). I employ a multi-analytical approach encompassing optical microscopy and experiments with the integration of data from chemical analysis of residues. The results of the analysis, combined with data from available literature, provide insights into several questions that enhance the debate on the technological complexity of Neanderthals and modern humans. What materials and additives were used by Neanderthals and modern humans to manufacture adhesives? Was there a difference in adhesive recipes depending on the context of use of the hafted tools? Was there a preference for hafting specific tools? Is there a difference between Neanderthal and modern human adhesive technologies in terms of raw materials exploited, use of additives, and context of use? Does adhesive technology reflect differences in technological complexity between Neanderthals and modern humans?

All the analysed adhesives were used by Neanderthals and modern humans to fasten their tools to organic handles. Adhesive residues have been identified on stone and organic projectile points, as well as on ‘common tools’ used in domestic tasks, strongly suggesting that adhesives were integrated into the

domestic economy of Neanderthals and modern humans. No relevant differences in the use of adhesives were observed depending on the tools' raw materials or functions.

Adhesives were mostly produced from natural resources available in the surrounding environment. At Steenbokfontein Cave, South Africa, adhesives were made using the resin or tar of conifer trees, specifically *Podocarpus* or *Widdringtonia*, both of which were available near the Cave and mixed with (mineral) additives. Similarly, at Morín Cave in Spain, the resin from a tree of the genus *Juniperus*, largely available in the environment, was likely used. However, there is evidence that some adhesives were selected over others equally available for their material properties. For instance, birch bark tar was preferred over pine resin for hafting bone points at the Dutch North Sea, a trend seen at many other Mesolithic sites. Furthermore, increasing evidence suggests that Neanderthals used additives, primarily iron oxides, to alter the material properties of their adhesives, similar to contemporaneous modern humans in Africa. This reflects Neanderthals' and modern humans' understanding of available natural resources, their distinct material properties, and the effects of their combinations.

Adhesive technology requires good knowledge of natural resources and their material properties, control of fire, enlarged cognitive functions, and forms of cultural transmission and social learning, qualifying it as a complex technology. The examination of adhesive remains in this thesis demonstrates that Neanderthals and modern humans share considerable technological parallels, highlighting Neanderthal technological sophistication. How Neanderthals selected, transformed, and employed adhesives suggests analogous procedures and reasoning to modern humans. Consequently, these insights likely reflect that Neanderthals had comparable cognitive and technological skills to anatomically modern humans.

Samevatting

Lijm is al duizenden jaren een essentieel onderdeel van het dagelijks leven van de mens. De geschiedenis van lijm begint met een 200,000 jaar oud werktuig dat is gevonden in de Campitello-groeve in Italië. Deze ontdekking toont het gebruik van lijmstoffen door Neanderthalers aan en daarmee hun vermogen om materialen te vervaardigen door middel van transformatieve processen. In Zuid-Afrika produceren moderne mensen al minstens 70,000 jaar samengestelde lijm door plantaardige materialen en mineralen te mengen, waarmee lijmstoffen geschikt worden gemaakt voor verschillende toepassingen, onder diverse omstandigheden. In de laatste jaren is het aantal geïdentificeerde prehistorische lijmstoffen toegenomen en is lijmtechnologie een graadmeter geworden voor technologische complexiteit van groepen mensachtigen. Om lijmtechnologie van menselijke groepen door tijd en ruimte te evalueren en vergelijken, is meer onderzoek nodig naar de selectie van lijmmaterialen, lijmrecepten en de context waarin lijmstoffen en hun werktuigen werden gebruikt.

Met dit proefschrift draag ik bij aan het vergroten van kennis over geïdentificeerde lijmstoffen. Hiervoor heb ik werktuigen met lijmresten uit de Steenbokfontein-grot (Zuid-Afrika, Later Steentijd), de Morín-grot (Spanje, Midden-Boven-Paleolithicum) en de Nederlandse Noordzee (Nederland, Mesolithicum) onderzocht. Ik gebruik een multi-analytische benadering die optische microscopie en experimenten combineert met data uit chemische analyses. De resultaten van de analyse, in combinatie met gegevens uit beschikbare literatuur, geven inzicht in verschillende vragen die de discussie over de technologische complexiteit van Neanderthalers en moderne mensen verrijkt. Welke materialen en toevoegingen werden door Neanderthalers en moderne mensen gebruikt om lijm te vervaardigen? Was er een verschil in lijmrecepten afhankelijk van de context van het gebruik en de montage van werktuigen? Was er een voorkeur voor het monteren van specifieke werktuigen? Is er een verschil tussen Neanderthaler- en moderne menselijke lijmtechnologie qua gebruikte grondstoffen, toevoegingen en de context van gebruik? Reflecteert lijmtechnologie verschillen in technologische complexiteit tussen Neanderthalers en moderne mensen?

Alle geanalyseerde lijmstoffen werden door Neanderthalers en moderne mensen gebruikt om hun werktuigen aan organische handvaten te bevestigen. Lijmresten

zijn geïdentificeerd op stenen en organische projectielen, evenals op 'gewone werktuigen' die voor huishoudelijke taken werden gebruikt. Dit suggereert dat lijm was geïntegreerd in het dagelijks leven van Neanderthalers en moderne mensen. Er werden geen relevante verschillen in het gebruik van lijms waargenomen op basis van de grondstoffen of de functies van werktuigen.

Lijm werd vooral geproduceerd uit natuurlijke grondstoffen die beschikbaar waren in de directe omgeving. In de Steenbokfontein-grot, Zuid-Afrika, werden lijmstoffen gemaakt van de hars of teer van naaldbomen, specifiek Podocarpus of Widdringtonia, die beide in de buurt van de grot verkrijgbaar waren en die gemengd werden met (minerale) toevoegingen. Ook werd in de Morín-grot in Spanje waarschijnlijk hars van een boom uit het geslacht Juniperus gebruikt, die in de omgeving volop aanwezig was. Er zijn echter aanwijzingen dat sommige lijmstoffen boven andere, even beschikbare stoffen werden verkozen vanwege hun specifieke eigenschappen. Zo werd berkenbast-teer verkozen boven pijnhars voor het monteren van benen spitsen uit de Nederlandse Noordzee, een trend die ook op veel andere Mesolithische sites wordt waargenomen. Bovendien zijn er steeds meer aanwijzingen dat Neanderthalers toevoegingen, met name ijzeroxiden, gebruikten om de eigenschappen van hun lijm te beïnvloeden vergelijkbaar met hoe moderne mensen in Afrika, die gelijktijdig leefden, dat deden. Dit toont aan dat Neanderthalers en moderne mensen op vergelijkbare wijze natuurlijke grondstoffen gebruikte, en dat ze kennis hadden van de verschillende eigenschappen van materialen, en van mogelijke combinaties van materialen.

Lijmtechnologie vereist een veel kennis over natuurlijke grondstoffen en hun eigenschappen, controle over vuur, cognitieve capaciteiten en vormen van culturele overdracht en sociaal leren. Daarom wordt het gebruik van lijm als een complexe technologie gekwalificeerd. Het onderzoek naar lijmresten in dit proefschrift toont aan dat Neanderthalers en moderne mensen aanzienlijke technologische overeenkomsten delen, wat de technologische kunde van Neanderthalers benadrukt. De wijze waarop Neanderthalers lijmstoffen selecteerden, transformeerden en toepasten, suggereert procedures en redeneringen die analoog zijn aan die van moderne mensen. Dit toont aan dat Neanderthalers vergelijkbare cognitieve en technologische vaardigheden hadden als anatomisch moderne mensen.

Chapter 1

Introduction

The invention of adhesives is considered an important advancement in the history of technology. An adhesive is any substance used for sticking objects or materials together. The main ingredient of an adhesive is the tackifier (e.g., resin and tar), which provides suitable viscoelastic properties and stickiness. Additives (e.g., filling agents, moisturisers, plasticisers) can be added to the formulation to improve the material properties of the adhesives (Langejans et al., 2022). Adhesives consist of the oldest known plastic materials in history (Langejans et al., 2022). They can be moulded into any desirable shape to create and bond a smooth, regular joint, and they can also be used on their own to create artifacts such as figurines (Kaal et al., 2020). The versatility of adhesives is illustrated by their wide range of archaeological and historic applications. Unlike many other bonding methods, like mechanical fastening, adhesives allow materials that are dissimilar in shape, size, and composition to be joined together. Additionally, they can be used to repair objects and serve as a sealant to waterproof containers, baskets, and wooden structures (Langejans et al., 2022). Today, adhesives play a pivotal role in our lives. Adhesives, like mainly synthetic polymeric materials, are employed daily in the industrial, biomedical, and pharmaceutical sectors due to their outstanding material properties and low production costs (Dinte & Sylvester, 2017; Hartshorn, 2012).

Material traces preserved in the archaeological record can inform us about the dawn of adhesive technology. From the perspective of archaeologists, the study of archaeological adhesives informs us on past populations' technology, know-how, knowledge of material properties, availability of natural resources, and even contains information linked to cognition. Adhesives are frequently produced through transformative processes that combine different raw materials, often irreversibly, to create an entirely new product with different properties (Schmidt, 2021; Wadley, 2013). These transformative processes rely on controlled heat treatment and require advanced executive functions of the brain, such as abstraction and forward planning, which are often considered characteristics of modern humans (Villa & Roebroeks, 2014; Wragg Sykes, 2015). However, the oldest known securely characterised adhesive was produced by Neanderthals and predates by about 100,000 years the first adhesive made by modern humans (Mazza et al., 2006). Therefore, adhesive technology plays a central role in the debate on technological and cognitive complexity in the past. Nonetheless, our knowledge of this technology is still scarce and relies on scattered evidence. To overcome this, my research aims to contribute to the current body of knowledge on prehistoric adhesives by enlarging the sample of securely identified adhesives to allow comparisons of different adhesive traditions. This work focuses on western European and South African Pleistocene and early Holocene adhesives, as they are among the earliest evidence of this technology, and their study can thus enhance the discussion on technological complexity in the deep past.

Moreover, research into ancient adhesives can also provide valuable inspiration for modern-day biocompatible adhesives. New classes of natural materials have attracted increasing scientific attention when it comes to developing environmentally friendly adhesives from renewable sources (e.g., Gaillard et al., 2013; Lang et al., 2018; Xu et al., 2014). The aim is to reduce toxicity and dependence on petroleum and promote a circular economy (Ferdosian et al., 2017; Heinrich, 2019). Therefore, enlarging our knowledge of archaeological adhesives, which were manufactured from natural substances, can help identify new organic resources that can be researched and tested in the future to facilitate this transition to eco-friendly materials.

Technological complexity in the deep past

The term 'technological complexity' is recurrent in the archaeological literature, although there is no clear definition of what it entails. Some technologies, such as composite tools, compound adhesives, mechanically delivered projectiles, and transformative technologies, are theorized to demonstrate technological complexity (Haidle, 2009; Hoffecker & Hoffecker, 2017; Lombard & Haidle, 2012; Schmidt et al., 2022; Stolarczyk & Schmidt, 2018; Wadley, 2010), but the traits that make a technology complex are often not clearly defined, hampering efforts to measure and compare technological traditions. The methods proposed to measure and compare complexity include quantitative approaches, i.e., counting the number of procedural units or manufacturing steps (Perreault et al., 2013), or the techno-units (Oswalt, 1976), graphical representations, i.e., cognigrams (Lombard & Haidle, 2012; Lombard et al., 2019), and system modelling with Petri nets (Fajardo et al., 2022; Kozowyk et al., 2023b).

Among others, complex technologies display the following traits, they are multi-stepped, have a hierarchical structure, require enlarged cognitive functions of the brain, are efficient with respect to energy cost and benefits, and involve some forms of cultural transmission and social learning (Hoffecker, 2018; Hoffecker & Hoffecker, 2017; Lombard, 2019; Schmidt, 2021; Wadley, 2013).

Adhesive technology exhibits many of these traits associated with complexity. They are manufactured from natural resources using several multi-stepped production methods involving the use of fire, and are often transformed irreversibly (Schmidt, 2021; Wadley, 2010). Transformative processes, which results in the creation of new materials, demand advanced technological and cognitive abilities, such as mental fluidity, forward planning, and imagination (Wadley, 2010). Further, the production of adhesives and multicomponent tools requires the collection and processing of different resources. It is consequently more costly than other Palaeolithic technologies. Thus, we can likely use adhesive production, selection of ingredients, and hafted tools design and use as indicators for technological complex processes in the deep past.

Archaeological adhesives

The oldest securely identified archaeological evidence of adhesive dates to about 200,000 years ago. Neanderthals at Campitello (Italy) produced adhesives by destructively distilling birch bark (*Betula* sp.) into tar to haft stone tools (Mazza et al., 2006). While the importance of this discovery was initially overlooked, it supports the high level of technological and cognitive complexity of Neanderthals (Fajardo et al., 2023; Niekus et al., 2019; Schmidt et al., 2023; Wragg Sykes, 2015). In Africa, the oldest securely identified adhesive is found in South Africa and dates to about 65,000 years ago (Soriano et al., 2015). It consists of a mixture of conifer resin, potentially from *Podocarpus*, and ochre used to haft lithic segments (Soriano et al., 2015). It is worth mentioning that older ochre and resin residues likely linked to hafting adhesives have been documented in several African sites, but were never chemically characterised (Rots et al., 2017; Rots et al., 2011; Wadley et al., 2009; Wojcieszak & Wadley, 2018). However, these residues suggest that adhesives were probably used in Africa since at least ~130,000 years ago (Rots et al., 2011). In Africa, early modern humans mixed plant extracts (i.e., resins, latex, and gums) with organic and inorganic additives to create compound adhesives. Compound adhesives are also considered evidence of complex technology, as they require a deep knowledge of the properties of individual ingredients, an understanding of chemical reactions, and controlled use of fire (Wadley, 2013; Wadley et al., 2009). Experimental work has demonstrated that the correct ratio of ingredients must be added to create an adhesive with optimum properties (Kozowyk et al., 2016; Wadley, 2005; Zipkin et al., 2014).

Adhesives are an early example of complex technology used by both Neanderthals and modern humans. Therefore, since their discovery, comparisons between Neanderthal and modern human adhesive technologies have been made (e.g., Schmidt, 2021; Villa & Soriano, 2010; Wadley, 2013; Wragg Sykes, 2015), aiming to shed light on the level of cognitive and technical abilities of each species and the potential differences. In this regard, recent studies have supported the complexity of Neanderthal tar production (Fajardo et al., 2023; Kozowyk et al., 2023b; Schmidt et al., 2023). Furthermore, adhesives allow the combination of dissimilar materials into composite tools. The production of multi-component sophisticated tools is often seen as evidence of technological complexity and had significant consequences on the material culture and evolutionary trajectory of our species (Ambrose, 2010; Barham, 2013; Lombard & Haidle, 2012; Rots, 2010).

Pleistocene adhesives from the European Palaeolithic and African Stone Age are rare in the archaeological record. Being organic materials, they do not preserve well, but they are documented in several assemblages in Eurasia and Africa. Molecularly identified Middle Palaeolithic adhesives (Fig. 1.1), dating between approximately 200-40,000 years ago and associated with Neanderthals, have been identified in 11 sites across Eurasia: Italy (Degano et al., 2019; Mazza et al., 2006), France (Schmidt et al., 2024b), the Netherlands (Niekus et al., 2019), Germany (Koller et al., 2001), Romania (Cârciumaru et al., 2012), Syria (Boëda et al., 2008b; Hauck et al., 2013), and Russia (Doronicheva et al., 2022). Upper Palaeolithic adhesives (Fig. 1.1), dating between approximately 40-12,000 years ago and associated with modern humans, have been identified in six sites across western Eurasia: Italy (Sano et al., 2019), Spain (Bradtmöller et al., 2016; Javier Muñoz et al., 2023), Denmark (Baales et al., 2017), Romania (Cârciumaru et al., 2012), and the United Kingdom (Langejans et al., 2023). In South Africa, modern human Middle Stone Age adhesives (Fig. 1.2) dating between approximately 70-40,000 years ago have been molecularly identified in three sites (Charrié-Duhaut et al., 2013; d'Errico et al., 2012; Soriano et al., 2015; Wojcieszak & Wadley, 2018). More evidence of adhesives is documented during the early Holocene, such as in the Mesolithic sites of northern Europe (Fig. 1.1) (Aveling & Heron, 1998; Bjørnevad et al., 2019; Chen et al., 2022; Gramsch, 2000; Kabaciński et al., 2023; Osipowicz et al., 2020a; Vahur et al., 2011) and South African Later

Stone Age sites (Fig. 1.2) (Charrié-Duhaut et al., 2016; Jerardino, 2001; Veall, 2022).

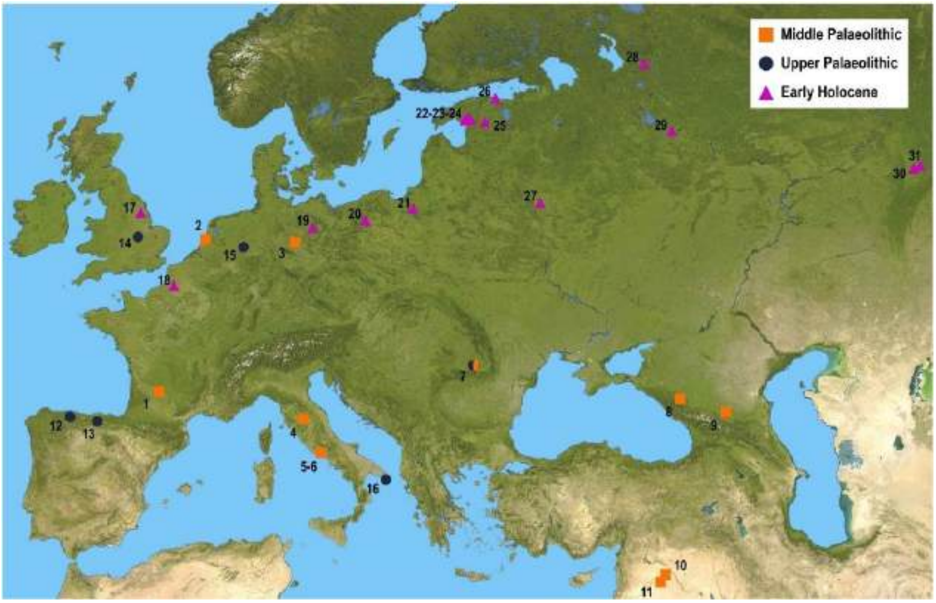

Figure 1.1: Distribution of spectrochemically identified Eurasian prehistoric adhesives. Middle Palaeolithic adhesives 1) Le Moustier (France); 2) Zandmotor (Netherlands); 3) Königsaue (Germany); 4) Campitello Quarry (Italy); 5) Fosellone Cave (Italy); 6) Sant'Agostino Cave (Italy); 7) Gura Cheii-Râşnov Cave (Romania); 8) Mezmaiskaya Cave (Russia); 9) Saradj-Chuko Grotto (Russia); 10) Hummal (Syria); 11) Umm el Tlel (Syria). Upper Palaeolithic adhesive 12) El Buxu Cave (Spain); 13) Morín Cave (Spain); 14) Hinxton (United Kingdom); 15) Bergkamen (Germany); 16) Grotta del Cavallo (Italy). Early Holocene adhesives 17) Star Carr (United Kingdom); 18) Hangest-sur-Somme Gravière II Nord (France); 19) Friesack 4 (Germany); 20) Krzyż Wielkopolski 7 (Poland); 21) Tłokowo (Poland); 22) Sindi-Lodja I (Estonia); 23) Pärnu River (Estonia); 24) Pulli (Estonia); 25) Ulbi (Estonia); 26) Kunda Lammasmägi (Estonia); 27) Aziarnoje 2B (Belarus); 28) Veretye I (Russia); 29) Stanovoye 4 (Russia); 30) Shigir (Russia); 31) Beregovaya 2 (Russia). Base map via Pixabay.

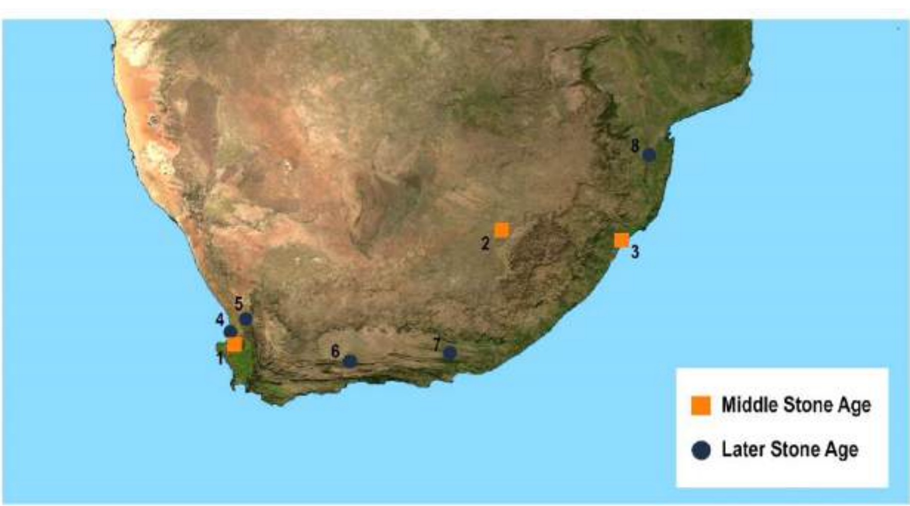

Figure 1.2: South African spectrochemically identified prehistoric adhesives. Middle Stone Age adhesives 1) Diepkloof Rock Shelter (Western Cape); 2) Rose Cottage Cave (Free State); 3) Sibudu Cave (KwaZulu-Natal). Later Stone Age adhesives 4) Elands Bay Cave (Western Cape); 5) Renbaan Cave (Western Cape); 6) Boomplaas Cave (Western Cape); 7) Melkhoutboom Cave (Eastern Cape); 8) Border Cave (KwaZulu-Natal). Base map via Pixabay.

Adhesives have mostly been identified with optical microscopy by documenting their distinctive morphologies, spatial distribution, and association with use-wear traces (Gibson et al., 2004; Lombard, 2007). In the last decades, molecular analyses, mainly spectroscopy and various chromatography methods, have been applied to identify adhesives (a.o. Chasan et al., 2024; Chen et al., 2022; Monnier et al., 2013; Prinsloo et al., 2023; Rageot et al., 2019; Regert, 2004; Schmidt et al., 2024a; Schmidt et al., 2022; Wojcieszak & Wadley, 2018), although not on a regular basis. Even when these analytical methods are applied, they are usually limited to small-sized samples, hampering the recognition of diachronic and geographical trends in adhesive production and use, as well as technological variability. Thanks to molecular analysis, we know that prehistoric adhesives were manufactured from different natural sources, including tree resins and gums, latex, plant and insect waxes, bark and wood tars, and bitumen (see Langejans et al., 2022 for a synthesis). Different adhesive sources were used depending on their availability in the environment, context of use, technological knowledge, material and working properties, and likely cultural traditions. Tar, for example, was likely preferentially chosen by Neanderthals for its superior mechanical properties over other natural adhesives (Kozowyk & Poulis, 2019). Other adhesives may have been preferred depending on the context of use. Some adhesives are better at withstanding impact, while others create stronger bonds or can be reheated and reused (Kozowyk & Poulis, 2019). Therefore, it is assumed that adhesive recipes were task-appropriate and likely adapted to the environmental and climatic conditions (Wadley et al., 2009). Other adhesives may have been selected or modified with additives depending on the tool's raw material. At Sibudu Cave (South Africa), for instance, ochre-loaded adhesives were employed to haft dolerite and hornfels segments, but they were not found on segments made of other lithologies (Lombard, 2007). In this regard, experiments provided supporting evidence of the difference in performance of various adhesive types depending on the substrate (Tydgadt & Rots, 2022). Therefore, by examining the use of specific adhesives within distinct contexts or the preference for certain raw materials over others that were locally accessible for adhesive production, we can infer past human groups' choices and reconstruct their know-how and technological skills. However, it is important to emphasise that some materials may be overrepresented in the archaeological record due to preservation biases. Experimental work has shown the differential preservation of adhesive types (Kozowyk et al., 2020). Collagen glues, for instance, preserve very poorly, and that may account for their absence in the archaeological record until the Neolithic (Solazzo et al., 2016) and their overall rarity.

Research aim

This research is part of the project "Ancient Adhesives: a window on prehistoric technological complexity", which aims to create a new computational method to study and compare technological complexity in the past by focusing on Neanderthal and modern human adhesives. To address this, in collaboration the team members (L. Baron, R. Chasan, M. Despotopoulou, S. Fajardo, P. Kozowyk, J. Postma, and J. Zeekaf), archaeological, experimental, ethnographic, and molecular data on adhesives were collected to be modelled with business process models (Petri net) to assess technological complexity. In this broader context, my work aims to record and compare technologically complex behaviour among Neanderthals and modern humans through the analysis of archaeological adhesive remains and their associated tools.

Despite their central role in the current debate on ancient human behaviour and technology (e.g., Kozowyk et al., 2023b; Niekus et al., 2019; Schmidt et al., 2023; Schmidt et al., 2022; Wadley, 2010), the actual use and reuse of prehistoric adhesives is understudied. Our understanding of past adhesive technology suffers from the lack of a systematic application of molecular methods for adhesive characterisation, specifically on large samples. Without information on adhesive ingredients, additives, loading agents, and context of use, it is challenging to validate current inferences on the complexity of prehistoric adhesives.

To address this missing knowledge, I studied artefacts from archaeological sites in western Europe and South Africa using optical microscopy and incorporated the interpretation of elemental and molecular data of residues collected by other team members and technicians. The European sites, Morín Cave (Spain) and the surface finds from the Dutch North Sea coast (Netherlands), provided contextual data on both Neanderthal and modern human adhesives, while the South African site, Steenbokfontein Cave, only has modern human adhesives. All the selected assemblages had already provided incidental evidence for adhesive remains and were highly suitable for further examinations. Moreover, considering the rarity of preservation for such finds, each site displayed a relatively large sample size. That allows discussions on the variability and versatility of adhesive technology, linked to tools function and material properties, natural resources exploitation, the diachronic evolution of adhesive technology, and intra-group comparisons.

The data will be used to answer the following research questions:

1. How embedded was adhesive technology in the tool kits of Neanderthals and modern humans?
2. Is there a difference between Neanderthal and modern human adhesive technologies in terms of raw materials exploited, use of additives, and context of use?
3. Was there a preference for hafting certain tool types?
4. Did Neanderthals and modern humans use different adhesives depending on the tool's function or raw material?

By addressing these questions, I will be able to answer the main research question that this thesis seeks to answer:

5. Does adhesive technology reflect differences in technological complexity between Neanderthals and modern humans?

Methodological approaches

To characterise and compare Neanderthal and modern human adhesive technologies, I employed a multi-analytical approach combining optical and scanning microscopy with the interpretation of data from spectroscopic and chemical analyses of residues. The analytical techniques used throughout this dissertation are optical microscopy, scanning electron microscopy-energy dispersive X-ray spectrometry (SEM-EDX), Fourier-transform infrared microspectroscopy (micro-FTIR), attenuated total reflectance (ATR-FTIR) analysis, Raman microspectroscopy (micro-Raman), X-ray diffraction (XRD), X-ray micro-computed tomography (μ-CT), and gas chromatography-mass spectrometry (GC-MS). The results of the analyses were integrated into the artefact biography framework, which allowed me to reconstruct the life-history of the tools and their adhesive residues, including their function and transformation through use and reuse.

Optical microscopy: use-wear and residue analysis

Since Semenov's pioneering work in 1964 (Semenov, 1964), use-wear and organic residue analyses have become essential tools for investigating prehistoric assemblages and identifying past behaviours. Use-wear analysis, or traceology, refers to the study of all the traces from use preserved on archaeological tools (a.o. Marreiros et al., 2015; Van Gijn, 2010). The analysis is conducted using different microscopes, with varying magnifications. A stereomicroscope (up to x60) is used to detect macroscopic wear in relation to the overall morphology of the tools (Van Gijn, 2010) and occasionally provides hints as to the hardness of the material worked and the use motion (i.e., 'low-power approach' (Odell, 1977; Semenov, 1964; Tringham et al., 1974)). Meanwhile, a high magnification metallographic microscope (up to x500) enables a more detailed interpretation of the material worked and the use motion based on the characteristics and distribution of polish, striations, and edge-damage (i.e., 'high-power approach' (Keeley, 1980; Keeley & Newcomer, 1977)). By combining the low- and high-power approaches, it is possible to infer the past use of a tool regarding the contact material and the actions undertaken. However, it is important to note that conclusions about tool use can only be regarded as interpretations and not as identifications (Van Gijn, 2014).

Interpretations of the wear traces are made using an experimental reference collection (Rots, 2010; Van Gijn, 1990). Therefore, our understanding of past activities is strongly influenced by the available comparative reference material. Experiments are key to this process, with generalised experiments used to reproduce and study a wide variety of tools, and problem-oriented experiments aimed at reproducing specific types of wear that do not yet have an experimental equivalent (Van Gijn, 2010, p. 30). For this reason, within the remit of this PhD, it was necessary to conduct an experiment to create a small reference collection of used tools made from South African rocks before analysing the Steenbokfontein tools (Chapter 2). Moreover, another experiment was conducted to better understand the characteristics of hafting traces on bone points that served as a reference for the analysis of archaeological bone points from the North Sea (Chapter 5). These experiments, together with the reference collection of the Laboratory for Material Culture Studies of Leiden University were used to infer the functions of archaeological tools with adhesives by comparing several corresponding attributes between archaeological and experimental use-wear traces.

Since its introduction in the late seventies, residue analysis has gradually developed into a valuable method for obtaining novel data on tool function and the use of plants and animals, otherwise invisible in the archaeological record (Langejans & Lombard, 2014; Rots et al., 2016). Residue analysis consists of the identification of organic and inorganic residues present on the artifact. This method uses high-magnification microscopy (up to x1000) to identify microscopic remains of processed materials left on the tool's surface after use (Langejans, 2012; Lombard & Wadley, 2007). Micro-residues, and especially organic ones, are generally poorly preserved on tools. However, their preservation depends primarily on the deposition environment and taphonomic processes (Langejans, 2010); therefore, only under certain circumstances organic residues can survive. Aggressive cleaning procedures, extensive handling, and storage are also factors that may affect the preservation of use-residues (Langejans, 2012). Therefore, specific protocols should be applied to preserve the integrity of organic residues prior to residue analysis.

The study of adhesives has mainly been carried out through microscopy. Adhesive residues have been identified in archaeological assemblages across Europe and Africa based on their morphological features (i.e., colour, shape, interaction with light), spatial distribution, and association with use-wear traces (Dinnis et al., 2009; Gibson et al., 2004; Lombard, 2006, 2007). However, several studies have emphasised the limitations of interpretation based solely on residue morphology and distribution patterns (Monnier et al., 2012; Pedergnana, 2020; Pedergnana et al., 2016). As seen during the analysis of adhesive residues on South African tools from Steenbokfontein Cave (Chapter 3), taphonomic processes can alter residue morphological attributes, leading to incorrect interpretations (see also Baales et al., 2017). Likewise, the spatial distribution of adhesive residues can change after the tool is discarded. Moreover, optical microscopy alone is inadequate in securely differentiating between plant exudates of distinct species despite the correct identification of adhesive residues (Soriano et al., 2015). Nevertheless, optical microscopy is a useful screening method for selecting tools with potential adhesive residues for further analysis and understanding their uses.

Moving forward: novel techniques for adhesive characterization

Due to the limitations of archaeological adhesives identification through optical microscopy, new techniques, including molecular protocols, have been applied in the last few decades to aid material-specific identifications (Bordes et al., 2017; Monnier et al., 2017; Perrault et al., 2018; Regert et al., 2006; Rumiński & Osipowicz, 2014). The methods selected in the project have already been introduced in the field of archaeology and proven to aid the identification of inorganic and organic compounds in adhesive mixtures. They can be divided into non-destructive (SEM-EDX, FTIR, Raman, XRD, and μ-CT) and destructive methods (GC-MS). Depending on the nature of the analysed sample, some techniques are better suited than others.

For the characterisation of the organic fraction of the residues, I applied the following methods:

- Scanning electron microscopy (SEM) coupled with an energy dispersive X-ray spectrometry (EDX) provides useful elemental composition information of the analysed substrate that supports the identification of organic compounds. This technique has been used in several studies to confirm the organic nature of residues and indicate the likely presence of adhesives (e.g., Bradtmöller et al., 2016; Monnier et al., 2013; Pawlik & Thissen, 2011). However, SEM-EDX cannot be used to identify specific compounds; therefore, only broad interpretations of the residue's nature can be made. In this dissertation, SEM-EDX was applied to the study of micro-residue from Morín Cave to confirm their organic nature and narrow down the number of tools to be further analysed with Raman spectroscopy (Chapter 4).
- Infrared spectroscopy (FTIR) and Raman are non-destructive techniques widely employed to characterise archaeological residues and are applicable to a wide array of materials (Artioli, 2010). They are forms of molecular spectroscopy, measuring characteristic vibrational or rotational frequencies of molecules, providing information on chemical bonding and molecular structure of organic and inorganic materials. To enable the analysis of small samples (mm-μm), it is possible to couple a microscope to the FTIR and Raman spectrometer. In adhesive residue analysis, micro-FTIR and micro-Raman are primarily used to characterise the organic components of the mixture (e.g., Brody et al., 2002; Chen et al., 2022; Monnier et al., 2013; Sano et al., 2019; Vahur et al., 2011) but also, to a lesser extent, to verify the

presence of mineral additives such as ochre (Prinsloo et al., 2023; Wojcieszak & Wadley, 2018). These techniques have the advantage of being non-destructive for the residue, which can be further analysed. However, they generally produce broad characterisation and therefore are not suited for mixtures or identifying specific materials (Langejans et al., 2022; Prinsloo et al., 2013). Additional limitations in the analysis of micro-residues with vibrational spectroscopy depend on the nature of the material, such as natural decay of organic compounds, limited sample size, uneven morphology, autofluorescence, and the absence of an extensive collection of reference spectra of pristine and degraded material (Monnier et al., 2017; Prinsloo et al., 2013; Prinsloo et al., 2014). Nonetheless, micro-FTIR and micro-Raman are very efficient in verifying the organic nature of the sample and provide indications for planning future analysis based on targeted protocols and methods. In this dissertation, these methods were primarily used to characterise the organic fraction of residues on stone tools from Steenbokfontein Cave (South Africa) and Morín Cave (Spain) (Chapters 3 and 4). By comparing the spectra of different samples, I could also assess if the nature and thus the source of the adhesives was similar.

- Gas chromatography-mass spectrometry (GC-MS) is one of the most precise methods to characterise unknown organic residues in archaeological samples (Langejans et al., 2022). This technique allows the identification of molecules or groups of molecules that are material-specific and thus serve as biomarkers to characterise different organic materials (Evershed, 2008). GC-MS has largely been applied to the study of prehistoric and historic adhesive residues (Charrié-Duhaut et al., 2013; Isaksson et al., 2023; Niekus et al., 2019; Regert et al., 2019; Regert et al., 2003; Schmidt et al., 2024b; Schmidt et al., 2022). Recently, attempts were also made to link the presence or absence of specific molecules with different production methods of adhesives, specifically birch tar (Chasan et al., 2024; Kozowyk et al., 2023a; Rageot et al., 2021; Rageot et al., 2019). The downsides of this method are its destructive nature and the often complicated interpretation of chromatograms (Langejans et al., 2022). Biomarker identification is reliant on a comparative reference collection and mass spectra, and it is further complicated by natural degradation processes that affect the original lipid composition. Diagnostic biomarkers may undergo transformations related to natural ageing, burial conditions, and anthropic modifications, such as thermal degradation during adhesive production or reworking, which can make their recognition challenging (Polla & Springer, 2022; Whelton et al., 2021). In addition, many biomarkers are not material-specific and are present in various natural sources. Therefore, their identification cannot be considered proof of the use of a particular resource (Polla & Springer, 2022; Whelton et al., 2021). Lastly, different extraction methods or analytical procedures are better suited for some classes of organic compounds than others and may impact the results (Cleland & Schroeter, 2018; Whelton et al., 2021). Liquid chromatography-mass spectrometry (LC-MS), for instance, is particularly suitable for large biomolecules while pyrolysis GC-MS is for very small samples (<1 mg) (Whelton et al., 2021). When dealing with unknown archaeological residues, the archaeological context, environmental data, and dating may help select the most appropriate analytical protocol by informing on species availability and exploitation in a specific environment and period. In this dissertation, GC-MS was applied with a consistent protocol to the characterisations of archaeological residue from Steenbokfontein Cave (Chapter 3), Morín Cave (Chapter 4), and the bone points from the North Sea (Chapter 5).

For the inorganic fraction of the residue, I used the following methods:

- X-ray diffraction (XRD) is a non-destructive method used to analyse the structure of crystalline materials. By studying the crystal structure and crystalline phases present in a material, it is possible to reveal chemical composition information (Artioli, 2013; Franceschi, 2014). In adhesive residue analysis, XRD is primarily used to characterise the inorganic components of the mixture and thus verify the presence of mineral additives, such as iron oxide, clay and sand components (Rosso et al., 2016). It can also be used to identify beeswax due to its semi-crystalline structure. In this dissertation, XRD was applied to verify the use of hematite as an additive in the adhesive mixture at Steenbokfontein Cave, corroborating microscopy observations (Chapter 3).
- X-ray micro-computed tomography (μ-CT) allows the study of archaeological objects and fossil remains non-destructively (e.g., Bernardini et al., 2012; Ngan-Tillard et al., 2018). This system collects thousands of digital radiographs using an advanced X-ray detector, which results in virtual three-dimensional images with resolutions of a few microns (Tuniz & Zanini, 2014). The reconstructed volumetric data can be sliced to gain insight

into the inner microstructural features of artefacts. In adhesive residue analysis, μ-CT is primarily used to explore the inner structure of the residue to check for the presence of additives, by-products of production processes, or soil contaminants (Niekus et al., 2019; Schmidt et al., 2023). Although μ-CT does not characterise the nature of additives, I used it for analysing the Steenbokfontein tools to evaluate the presence and distribution of iron oxide particles within the adhesive to support the use of hematite as an additive, corroborating the XRD result (Chapter 3).

Theoretical framework: artefact biography approach

The data collected in the various chapters will be integrated using the cultural biography approach, which views objects as having biographies analogous to those of humans (Gosden & Marshall, 1999; Joy, 2009; Kopytoff, 1986). The core assumption is that objects' characteristics and functions are constantly transformed during their life. By studying these transformations, it should be possible to reconstruct human-material interactions and some of the choices of past agents (Gosden & Marshall, 1999; Van Gijn & Wentink, 2013).

The notion of cultural biography differs from Schiffer's notion of life-history (Schiffer, 1972) because it emphasises the interplay between people and objects, which is absent in the latter. The life-history approach accounts that artefacts are not static and considers the full life of the objects from raw material procurement, manufacture, use, maintenance, storage, transport, recycling, reuse, and discard to explain technological and performance changes at each stage in the object life-cycle (Schiffer, 2004; Schiffer et al., 2001). The cultural biography approach looks at objects from the perspective of their birth (conceptualization and design), life (use and reuse), and death (deposition, loss, and discard). This approach has been criticised for its linearity, as objects do not necessarily follow a single and straight life path (Hahn & Weiss, 2013; Joy, 2009; Meirion Jones et al., 2016). During the different stages of their life, objects are involved in particular sets of relationships and can act simultaneously in different relationship webs (Kopytoff, 1986). They can enter and leave different spheres of relationships, 'die' several times, or their life can extend over a series of human lifetimes (Joy, 2009). A case in point is the stone axes of the Dani of Papua New Guinea, which, during their life-cycle, go from being used as functional tools, to gifts between wedding partners to seal the relationships, to sacred objects hosting the spirit of the deceased (Hampton, 1999; Van Gijn & Wentink, 2013). This becomes more challenging when dealing with prehistoric objects, since no written documents are preserved, and the available evidence is usually scarce and fragmented (Joy, 2009; Van Gijn & Wentink, 2013). To address these limitations, it is best to avoid considering the biography of things as a linear pathway that can be fully reconstructed. Instead, we should focus on specific stages of an object's life in particular contexts where the available archaeological evidence can shed light on the transformations that can occur due to interaction between people and things (Hahn & Weiss, 2013; Joy, 2009; Van Gijn & Wentink, 2013).

To reconstruct objects' biographies, use-wear and residue analysis are useful tools since they provide evidence of the actual use of the objects and allow us to document the different phases of an object's use life and the associated changes in function and/or meaning (Van Gijn & Wentink, 2013; Van Gijn, 2010). By applying this approach, we can shed light on aspects of tools' life cycle that are otherwise invisible. The 'birth' of the adhesives is informed through optical microscopy and molecular analysis. The characterisation of the adhesive is fundamental to understanding choices and combinations of raw materials, while residue distribution and use-wear can inform the design of the hafted tool. The actual 'life' is investigated through use-wear analysis, which provides clues on how the residues and their tools were (re)used and curated. The 'death' of the objects is discussed in relation to the deposition context. Through the combination of this evidence, we can write biographies of adhesives and their tools and explore the variety of behaviours and choices of past agents (cf. Van Gijn, 2010). Through this framework, useful insights to address the research questions are generated and discussed.

Thesis outline

- Chapter 1 – Introduction. This chapter includes background information on prehistoric adhesives research, a summary of archaeological finds, and the research aim and questions. Additionally, it provides an overview of the analytical and theoretical techniques employed throughout this dissertation to analyse archaeological adhesives.
- Chapter 2 – Building up a reference collection for the study of non-flint lithic raw materials. This chapter presents the results of an experiment aimed at

assessing the comparability of use-wear traces on tools made of South African rocks with those on European flint tools. This experiment was a necessary step before the analysis of non-flint tools from Steenbokfontein Cave. To evaluate whether adhesive recipes are tailored according to tools' function, a traceological study of the tools with residues is required. Therefore, a reference collection representative of the use-wear traces that develop on local rock types is needed. My results highlighted a certain degree of comparability between wear traces on flint and non-flint tools. A flint reference collection can thus be used to make basic interpretations of use-wear on different rocks.

- Chapter 3 – Exploring South African compound adhesives. This chapter presents the results of a multi-analytical study of 30 stone tools with adhesive residue from the entire stratigraphic sequence of Steenbokfontein Cave, Western Cape (South Africa), dated between ~5250 and ~2200 cal BP. This research aimed to document possible diachronic trends in adhesive technology or changes in adhesive recipes due to the tool's material properties and uses. I found evidence for three different adhesive mixtures, yet my results also highlighted the continuity in the use of conifer resin or tar, likely *Podocarpus* or *Widdringtonia*, as the main adhesive ingredient and the flexibility in the use of additives independently of tools' raw materials and functions.
- Chapter 4 – European Palaeolithic adhesives: a case study from Spain. This chapter presents the result of a multi-analytical study of stone tools with adhesive residues from the Palaeolithic site of Morín Cave, Cantabria (Spain), dated between ~45,000 and ~25,000 BP. With its long stratigraphic sequence spanning from the Middle to the Upper Palaeolithic, the site offered the opportunity to directly compare Neanderthal and modern human adhesive technologies. I found a strong similarity in adhesive traditions during the Middle to Upper Palaeolithic transition, which could be attributed to environmental factors, such as the limited availability of species suitable for adhesive production. However, the possibility of technological and cultural exchange between human groups cannot be ruled out.
- Chapter 5 – European Mesolithic adhesives: the bone points from the Dutch North Sea. This chapter presents the result of a multi-analytical study of osseous points with adhesive residues from the Dutch North Sea coast (~13,000-10,000 BP). I reconstructed how the points were used and their hafting arrangements based on experimental comparisons, archaeological

use-wear, and residue analysis. Tar, in combination with sinew or vegetal bindings, was used to haft the points used for aquatic and terrestrial hunting. Here, the choice of tar can be explained by looking at its material properties. Tar is water-insoluble and can be reheated many times, accommodating the extensive reuse of the points, which I also documented during the functional analysis.

- Chapter 6 – Discussion and conclusion. In this concluding chapter, I summarise and discuss the results presented in chapter two to five in light of the research questions of this dissertation. I use data on adhesive ingredients, context of use, and reuse to identify and compare trends in prehistoric adhesive traditions and reflect on the technological and cognitive implications of this technology.

Chapter 2

Building up a reference collection for non-flint lithic raw materials

"Comparing the formation and characteristics of use-wear traces on flint, chert, dolerite and quartz"

Lithic Technology

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Supplementary Information:

Abstract

Use-wear traces are considered to be material specific. The use of an appropriate reference collection is thus fundamental for interpreting tools' function. To test whether a flint reference collection can be used to interpret the function of non-flint tools, I conducted experiments using chert, dolerite, and quartz endscrapers and flakes. I compared wear traces obtained during the experiment with use-wear on experimental flint tools exposed to the same variables (motion, contact material, time). The results highlighted strong similarities in the characteristics and distribution of traces on chert and flint. Dolerite and quartz differ from flint, especially regarding the distribution and appearance of use-polish. However, shared traits were observed in all the raw materials involved in this experiment, demonstrating a certain degree of comparability between use-wear traces on flint and non-flint rocks. Based on the data, a flint reference collection can allow a basic interpretation of use-wear also on different rocks.

Introduction

Use-wear analysis is becoming more and more part of mainstream methodologies in archaeological research across the world. The method greatly developed since its conception sixty odd years ago (Evans et al., 2014; Marreiros et al., 2020 and references therein). Traceology refers to the study of macro and microscopic wear traces on the edges/surfaces of objects. By studying the characteristics and distribution patterns of wear traces it is possible to infer past tools' functions. Experiments form a fundamental component of use-wear studies. Based on experiments, archaeologists can infer past tool function and production in light of the similarities between corresponding attributes of archaeological and experimental use-wear (Van Gijn, 2010). Hence, the interpretation of wear traces is strongly influenced by the available comparative reference collection. One research hiatus is that comparative collections of non-flint knapped tools are still rare. With this study, I want to explore how traces on different lithic raw materials compare to detect whether flint tools can be used as a reference collection for non-flint artefacts.

Non-flint materials, and coarse rocks in particular, generally attract little scientific attention, and technological and typological frames derived from flint have often been applied to them (Knutsson, 1998). The same holds for functional studies. However, raw material properties not only influence knapping attributes, but also the distribution and appearance of the wear traces. Hence, we can expect tools made on different rocks to exhibit different patterns of use-wear traces (Clemente-Conte et al., 2015). Despite the frequency of flint tools, there are regions where good quality flint is scarce or unavailable, and is replaced by other rock types with similar knapping properties, for example quartz and quartzite (Aubry et al., 2016; Knutsson et al., 2016). Outside Europe, numerous fine- and coarse-grained rocks, like chert, quartzite, and silcrete, are used in tool production instead of flint (e.g., Douglass et al., 2016; Holdaway & Douglass, 2015; Nami, 2015; Will, 2021).

The identification and interpretation of use-wear traces on quartzose and heterogenous rocks are often considered problematic. Recently, several experimental programs aimed to broaden our knowledge on the mechanical responses to stress caused by the use and wear formation process on non-flint rocks were created (e.g., Bello-Alonso et al., 2020; Bello-Alonso et al., 2019; Fernández-Marchena & Ollé, 2016). Concurrently, new analytical techniques, such as scanning electron microscope (SEM) and laser scanning confocal microscope, improved the detection and recognition of wear traces on these highly reflective and irregular rocks (e.g., Ollé et al., 2016; Pedergnana et al., 2020). The application of these methods to archaeological materials highlighted the feasibility of functional interpretation of lithic assemblages composed of non-flint artefacts (e.g., Lemorini et al., 2019). In these recent advances, the authors stressed the need to use rock-specific reference collections. Comparisons to use-wear traces on flint are problematic due to significant differences in the raw material properties which influence wear formation (see for example Bello-Alonso et al., 2019).

It is certainly true that wear traces are in part material specific, but there are also similarities in the types of wear analysts encounter (Clemente-Conte et al., 2015). Because building a comprehensive raw material specific reference collection is not always an option, I set out to test the extent to which we can rely on a reference collection of flint tools to interpret the wear traces of non-flint knapped materials. To do so, I designed a systematic experiment in which the same activity, with the same use duration, is performed with flint tools and tools made of other lithologies. As my overarching research interest includes South African lithics, I focused on well-known resources from there: chert, quartz, and dolerite. A description of macro and microwear traces resulting from the work of different materials, was provided for chert, dolerite, quartz, and flint artifacts. After that, I performed a comparative analysis between experimental traces on flint and non-flint tools to investigate which differences in the characteristics of wear traces can be observed and how these influence the interpretation of tools' function.

Materials and methods

Experimental tools: raw materials and tool types

Experimental flake tools and endscrapers were made on rock types that are generally found in the South African archaeological record: chert, dolerite, and quartz (Table 2.1, rocks descriptions SI). All flint tools are non-cortical and made of a fine-grained European flint (Fig. SI1). The tools were made by expert flint knappers, using soft and hard hammer stones. The chert and dolerite rocks were collected in Lesotho and the Kwazulu-Natal province of South Africa. The quartz cobbles are store-bought and collected from locally river-beds in the

Netherlands. All chert tools are made of a fine- to medium-grained chert and are non-cortical (Fig. SI2). Due to the shortage of good quality quartz and dolerite, and the small dimension of the available nodules, tools were made from both cortical and non-cortical flakes (Fig. SI3, SI4). The quartz cobbles had a smooth, rounded cortical exterior with weathering limited to some sheen over the surface. The dolerite cortical exterior is angular and rough.

Table 2.1: Overview of the experimental tools and the variables tested in the experiment.

Exp nr.	Raw material	Tool type	Cortex on the used edge	Handling	Motion	Contact material	Use-duration (min)
2558	Flint	Endscraper	No	Hafted	Scraping	Hide	30
665	Flint	Endscraper	No	Handheld	Scraping	Hide	60
2555	Flint	Flake	No	Handheld	Cutting	Hide	30
2556	Flint	Flake	No	Handheld	Cutting	Hide	60
207	Flint	Endscraper	No	Handheld	Scraping	Bone	30
1810	Flint	Endscraper	No	Handheld	Scraping	Bone	60
48	Flint	Flake	No	Handheld	Sawing	Bone	30
2557	Flint	Flake	No	Handheld	Sawing	Bone	60
428	Flint	Flake	No	Handheld	Cutting	Reed	30
183	Flint	Flake	No	Handheld	Cutting	Reed	60
3827	Chert	Endscraper	No	Hafted	Scraping	Hide	30
3826	Chert	Endscraper	No	Hafted	Scraping	Hide	60 (2*30)
3831	Chert	Flake	No	Handheld	Cutting	Hide	30
3830	Chert	Flake	No	Handheld	Cutting	Hide	60 (2*30)
3824	Chert	Endscraper	No	Hafted	Scraping	Bone	30
3825	Chert	Endscraper	No	Hafted	Scraping	Bone	60 (2*30)
3828	Chert	Flake	No	Handheld	Sawing	Bone	30
3829	Chert	Flake	No	Handheld	Sawing	Bone	60 (2*30)
3832	Chert	Flake	No	Handheld	Cutting	Reed	30
3833	Chert	Flake	No	Handheld	Cutting	Reed	60 (2*30)
3855	Dolerite	Endscraper	No	Hafted	Scraping	Hide	30
3807	Dolerite	Endscraper	No	Hafted	Scraping	Hide	60 (2*30)
3809	Dolerite	Flake	Yes	Handheld	Cutting	Hide	30
3808	Dolerite	Flake	No	Handheld	Cutting	Hide	60 (2*30)
3806	Dolerite	Endscraper	No	Hafted	Scraping	Bone	30

3805	Dolerite	Endscraper	No	Hafted	Scraping	Bone	60 (2*30)
3813	Dolerite	Flake	No	Handheld	Sawing	Bone	30
3811	Dolerite	Flake	Partially	Handheld	Sawing	Bone	60 (2*30)
3812	Dolerite	Flake	Yes	Handheld	Cutting	Reed	30
3810	Dolerite	Flake	No	Handheld	Cutting	Reed	60 (2*30)
3816	Quartz	Endscraper	No	Hafted	Scraping	Hide	30
3814	Quartz	Endscraper	No	Hafted	Scraping	Hide	60 (2*30)
3821	Quartz	Flake	No	Handheld	Cutting	Hide	30
3823	Quartz	Flake	Yes	Handheld	Cutting	Hide	60 (2*30)
3817	Quartz	Endscraper	No	Hafted	Scraping	Bone	30
3815	Quartz	Endscraper	No	Hafted	Scraping	Bone	60 (2*30)
3822	Quartz	Flake	Yes	Handheld	Sawing	Bone	30
3819	Quartz	Flake	Yes	Handheld	Sawing	Bone	60 (2*30)
3818	Quartz	Flake	Yes	Handheld	Cutting	Reed	30
3820	Quartz	Flake	Partially	Handheld	Cutting	Reed	60 (2*30)

The scrapers were retouched to endscrapers and hafted by side-mounting them at one end of a pine wood handle with the aid of a compound adhesive made of pine resin, beeswax, and ochre (Fig. SI5). The hafting design and adhesive recipe are representative of hafted tools in the African Middle Stone Age (MSA) and Later Stone Age (LSA) (Deacon & Deacon, 1980; Lombard, 2007; Wadley, 2005).

In the experiment, I used the hafted scrapers in a transverse scraping motion. The flakes remained unretouched and were handheld. They were used in longitudinal motions (cutting and sawing).

The experiments: contact materials, motion, and time

The experiment can be considered a generalized reference experiment (Van Gijn, 1990). These types of experiments are aimed to reproduce and study a wide range of used tools, and it was not my goal to replicate specific tasks or archaeological objects.

I used the tools to process animal and plant materials (Table 2.1). The contact materials ranged from soft to medium-hard and consist of fresh deer hide (*Cervus elaphus;* soft), green reeds (*Phragmites australis* Trin.; soft-medium), and fresh deer bone (*Cervus elaphus;* medium-hard). I selected these materials because they represent a plausible counterpart of the raw materials that could have been exploited during the MSA and LSA. Reeds are widely known in the ethnographic record for being used as shafts for bone-tipped arrows (Deacon, 1992) and may have been used for the same purpose in the past. Red deer was chosen to replace African medium-sized ungulates (Steele & Klein, 2013).

The hide and bone raw materials were scraped and cut with the scrapers and flake tools respectively. In the hide scraping experiment, the hafted tools were placed perpendicular to the hide and pulled towards the hide-worker (see as reference Konso hide-workers in Ethiopia; Rots & Williamson, 2004) (Fig. 2.1a). The endscrapers were used to clean fresh skins which were cut with flake tools. The cutting motion was done unidirectionally (Fig. 2.1b, c). A downward motion was also applied in the bone working experiments, where endscrapers were used to deflesh and scrape the surface of fresh bones (Fig. 2.1d). The bone-cutting experiments were conducted on the same bones after they had been cleaned and scraped in the previous experiment. The bones were cut, creating deep incisions against the grain of the bone but never cutting through it (Fig. 2.1e). A bidirectional longitudinal sawing motion was applied. Reeds were only cut with flake tools and not scraped. Fresh reeds were cut by placing the flake perpendicular to the reed's stem and using a unidirectional motion (Fig. 2.1f).

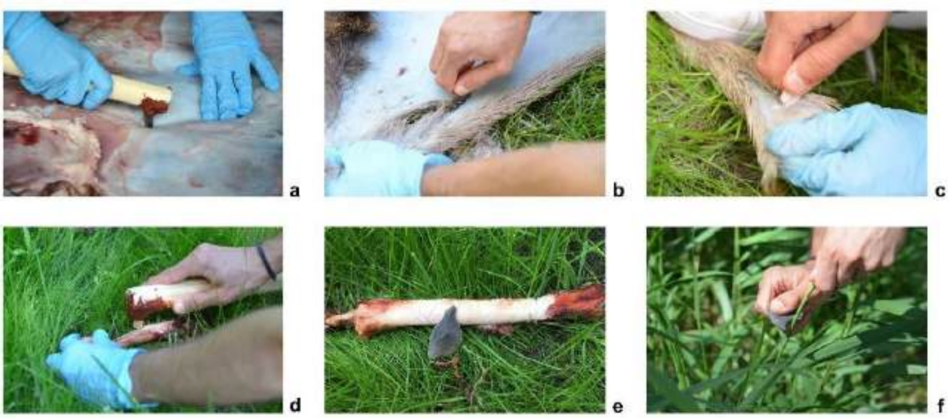

Figure 2.1: Experimental activities carried out. a) Scraping a fresh deer hide with a hafted endscraper; b, c) Cutting fresh deer hide with handheld flakes; d) Scraping a fresh deer bone with a hafted endscraper; e) Deep incisions on a fresh deer bone from sawing with a handheld flake; f) Cutting green reeds with a handheld flake.

Each experiment was conducted twice, at two time intervals: 30 and 60 minutes (Table 2.1). With this sequential experiment the progression of use-wear traces through the use was documented (cf. Ollé & Vergès, 2014). Only one active side or area of the tool was used, concentrating potential wear formation to a specific edge. All activities were carried out by the same person (AA) to reduce the variables related to the experimenter. Regarding the flint tools, I conducted experiments 2555, 2556, 2557, and 2558. The others were selected from the available Laboratory for Material Cultural Studies reference collection which comprises more than 4000 experimental tools.

Methods: sample preparation, microscopy and wear analysis

To capture the development of the traces, all intended active surfaces of the tools were examined and photographed prior to the experiments. I analysed the tools after the 30-minute interval and again after the last 60-minute interval. I aimed to photograph the same locations after each use-session. However, because of edge-removals and crushing during some of the activities, several spots were lost. When that occurred, I selected new photo locations where traces had already started to develop.

Before microscopy the tools were cleaned under running water and in an ultrasonic bath with water and soap for 10 minutes. This was to remove use-residues that hindered the analysis. Some tools needed a further 20-minute cleaning treatment. During the analysis under high magnification, all pieces were cleaned with a 96% alcohol solution to remove any finger grease from handling.

Macroscopic traces were visually analysed using a Leica M80 stereomicroscope with an external light source and magnifications ranging from x7.5 up to x60. Images were taken using a Leica MC120 HD camera. Microscopic traces were observed with a Leica DM6000 M metallurgical microscope with incident light and bright field illumination and magnifications ranging from x50 up to x500. Images were captured with a Leica DFC450 camera and microscopy z-stacking software to overcome the problem of low depth of focus.

For each implement, I recorded the location, distribution and association of the use-wear on the tool's surface, using a system of polar coordinates (Fig. SI6, Van Gijn, 1990). I recorded the following variables: edge-damage (edge-removals, crushing, edge-reduction), edge-rounding, polish, striations, and abrasion. Abrasion corresponds to the disappearance of part of the original surface of crystals due to use. The degree of abrasion and the shape and size of the hollows ('pecking') are indicators of the type of the worked material (Clemente-Conte & Gibaja Bao, 2009; Clemente-Conte et al., 2015; Ollé et al., 2016). Edge-removals were evaluated based on their distribution along the edge and the orientation of the scars. The former is informative of the hardness of the material worked, the latter of the use-motion (Semenov, 1964; Tringham et al., 1974). Polish was described by means of various attributes namely: distribution, texture, topography, brightness and degree of linkage. Polish distribution is influenced by the worked material, the use duration and applied motion. Polish texture and topography are related to the worked material as well as brightness. The description of the micro-polish follows the terminology and methodology developed by Keeley (1980) and further developed by others (Van Gijn, 1990; Van Gijn, 2010; Van Gijn, 2014; Vaughan, 1985). The orientation of the striations in relation to the used edge was recorded. I described the development of edge-rounding, polish, and abrasion as light, moderate, or heavy. The degree of rounding was visually assessed based on the extent of rounding of the used edge caused by working a specific contact material. For polish and abrasion, I observed the extension of the surface area affected by alteration. For the former, I also considered the degree of linkage in the polish. Considering that use-wear traces do not always develop in the same way along the edge, the degree of wear of a specific trace may vary depending on its location.

Finally, a comparative analysis was undertaken to identify shared similarities in the characteristics and distribution pattern of traces. The aim was to explore the possibilities and limitations of using the reference collection of flint tools to infer the function of non-flint tools.

Results

Table 2.2: Overview of the production and use-wear traces recorded on the experimental tools. The description of the experiment type (Exp. type) is compiled as follows: raw material, motion (scraping or cutting/sawing), contact material, time (30 or 60 minutes). Production traces are grouped together. The detailed description of the single experiments is given in the SI.

Exp.	Exp. Type	Traces									
		Production	Use								
			Edge-removals	Edge-rounding	Polish development	Polish distribution	Polish texture	Polish topography	Polish brightness	Abrasion	Striations

2558	Flint/Scr/Hide/30	-striations -polish -edge-damage	-	-light	-light -moderate	-band along the edge -invasive	-rough and greasy	-	-bright	-	-
665	Flint/Scr/Hide/60	-	-	-moderate	-moderate	-band along the edge -invasive	-rough and greasy	-cratered	-bright	-	-
2555	Flint/Cut/Hide/30	-	-isolated	-light -moderate	-light -moderate	-band along the edge -invasive	-rough and greasy	-pitted -cratered	-bright	-	-
2556	Flint/Cut/Hide/60	-	-isolated	-moderate	-moderate	-band along the edge -invasive	-rough and greasy	-pitted	-bright	-	-
207	Flint/Scr/Bone/30	-	-isolated	-light	-light	-line along the edge	-smooth and matt	-domed	-bright	-	-
1810	Flint/Scr/Bone/60	-	-isolated	-	-moderate	-line along the edge -isolated spots	-smooth and matt	-domed -flat	-very bright	-	-
48	Flint/Saw/Bone/30	-	-close	-	-light -moderate	-isolated spots	-smooth and matt	-domed	-very bright	-	-longitudinal
2557	Flint/Saw/Bone/60	-	-close -scalar	-	-moderate -heavy	-isolated spots	-smooth and matt	-domed -flat -pitted	-very bright	-	-longitudinal
428	Flint/Cut/Reed/30	-	-isolated	-light	-moderate	-band along the edge -invasive	-smooth and matt	-domed	-very bright	-	-
183	Flint/Cut/Reed/60	-	-isolated -close	-light	-moderate -heavy	-band along the edge -invasive	-smooth and matt	-flat -domed	-very bright	-	-
3827	Chert/Scr/Hide/30	-	-	-light	-light	-band along the edge -invasive	-rough and greasy	-pitted -cratered	-bright	-	-
3826	Chert/Scr/Hide/60	-	-	-light	-light -moderate	-band along the edge -invasive	-rough and greasy	-	-bright	-	-

3831	Chert/Cut/Hide/30	-	-rare -isolated	-light	-light	-band along the edge -invasive	-rough and greasy	-	-bright	-	-
3830	Chert/Cut/Hide/60	-	-isolated	-light	-light -moderate	-band along the edge -isolated spots -invasive	-rough and greasy	-	-bright	-	-
3824	Chert/Scr/Bone/30	-	-single	-light	-light -moderate	-line along the edge -isolated spots	-smooth and matt	-	-very bright	-	-
3825	Chert/Scr/Bone/60	-	-isolated	-light	-light -moderate	-line along the edge	-smooth and matt	-pitted	-very bright	-	-
3828	Chert/Saw/Bone/30	-	-close	-	-light	-isolated spots	-smooth and matt	-domed	-very bright	-	-
3829	Chert/Saw/Bone/60	-	-close	-	-moderate	-isolated spots	-smooth and matt	-domed	-very bright	-	-longitudinal
3832	Chert/Cut/Reed/30	-	-close	-light	-light -moderate	-band along the edge -invasive	-smooth and matt	-domed	-very bright	-	-
3833	Chert/Cut/Reed/60	-	-close	-light	-moderate	-band along the edge -invasive	-smooth and matt	-domed	-very bright	-	-
3855	Dole/Scr/Hide/30	-polish	-	-light	-light -moderate	-band along the edge -isolated spots -invasive	-granular	-flat	-very bright	-	-
3807	Dole/Scr/Hide/60	-	-	-moderate	-light -moderate	-band along the edge -isolated spots -invasive	-granular	-flat	-very bright	-	-
3809	Dole/Cut/Hide/30	-	-isolated	-light	-light	-isolated spots	-rough and greasy	-flat	-very bright	-	-

							-granular				
3808	Dole/Cut/Hide/60	-	-isolated	-light	-light	-isolated spots	-granular	-flat	-very bright	-	-
3806	Dole/Scr/Bone/30	-	-isolated	-light	-light	-isolated spots	-smooth and matt	-domed	-bright -very bright	-light	-transverse
3805	Dole/Scr/Bone/60	-	-isolated	-light	-light	-isolated spots	-smooth and matt	-domed -pitted	-bright -very bright	-light	-
3813	Dole/Saw/Bone/30	-	-close	-	-light	-isolated spots	-smooth and matt	-	-bright	-	-
3811	Dole/Saw/Bone/60	-	-close	-light	-light	-isolated spots	-smooth and matt -granular	-domed	-bright -very bright	-	-
3812	Dole/Cut/Reed/30	-	-isolated	-light	-light -moderate	-isolated spots -invasive	-smooth and matt	-domed	-very bright	-	-
3810	Dole/Cut/Reed/60	-	-isolated	-light	-light -moderate	-isolated spots -invasive	-smooth and matt	-domed	-very bright	-light	-
3816	Quartz/Scr/Hide/30	-	-rare -isolated	-light	-absent	-	-	-	-	-	-
3814	Quartz/Scr/Hide/60	-	-rare -isolated	-moderate	-light	-isolated spots	-rough and greasy	-	-bright	-light	-
3821	Quartz/Cut/Hide/30	-	-isolated	-light	-absent	-	-	-	-	-	-
3823	Quartz/Cut/Hide/60	-	-isolated	-light -moderate	-light -moderate	-band along the edge -invasive -isolated spots	-rough and greasy	-	-dull -bright	-light -moderate	-
3817	Quartz/Scr/Bone/30	-	-isolated	-	-absent	-	-	-	-	-	-
3815	Quartz/Scr/Bone/60	-	-isolated	-light	-light	-isolated spots	-smoot	-	-bright	-moderate	-

							h and matt				
3822	Quartz/Saw/Bone/30	-	-isolated -close	-light	-light	-isolated spots	-smooth and matt	-	-very bright	-	-
3819	Quartz/Saw/Bone/60	-	-close	-	-light -moderate	-isolated spots	-smooth and matt	-domed -pitted -comet tails	-very bright	-moderate	-
3818	Quartz/Cut/Reed/30	-	-isolated	-light	-moderate	-band along the edge -invasive	-smooth and matt	-domed	-very bright	-	-
3820	Quartz/Cut/Reed/60	-	-isolated	-light	-light -moderate	-band along the edge -invasive	-smooth and matt	-domed	-very bright	-	-

Flint

Hide: Working fresh hide resulted in edge-rounding, rare edge-removals, which occurred only during the cutting activity, and a greasy polish developed in a characteristic band along the edge. The polish is bright and invasive and follows the profile of the working edge exactly (Fig. 2.2a, b). The texture of the polish is either pitted or cratered. Polish on the endscrapers display a transverse directionality. The degree of edge-rounding varies between light to moderate, and it is never heavily developed even after 60 minutes of use. As already demonstrated in previous studies (cf. Collin & Jardon-Giner, 1993; Loebel, 2013; Rots, 2005), working fresh hide creates a less pronounced rounding of the edge and a greasier and brighter polish compared to dry hide.

Bone: Traces produced by contact with fresh bone consist of edge-rounding, edge-removals, and polish. Scraping fresh bone resulted in a slightly developed rounding, a few edge-removals, and a smooth polish distributed in a thin line along the edge (Fig. 2.2c). Sawing caused more edge-removals and no rounding. The associated polish is smooth and matt, it has a localized distribution and a clear longitudinal directionality (Fig. 2.2d, e). Tiny pits are visible in the polish. Longitudinal striations, indicative of the use-motion, were documented on both cutting tools. No striations were documented on the endscrapers, but the polish displays a transverse directionality.

<u>Reed:</u> Cutting fresh reeds produced a slight edge-rounding, unevenly distributed edge-removals, and a band of well-developed highly linked polish (Fig. 2.2f). The polish has a smooth and matt texture and both domed and flat topography. Well-developed spots of polish generally display a flat topography rather than domed. The brightness is very intense, and the polish exhibits a clear longitudinal directionality.

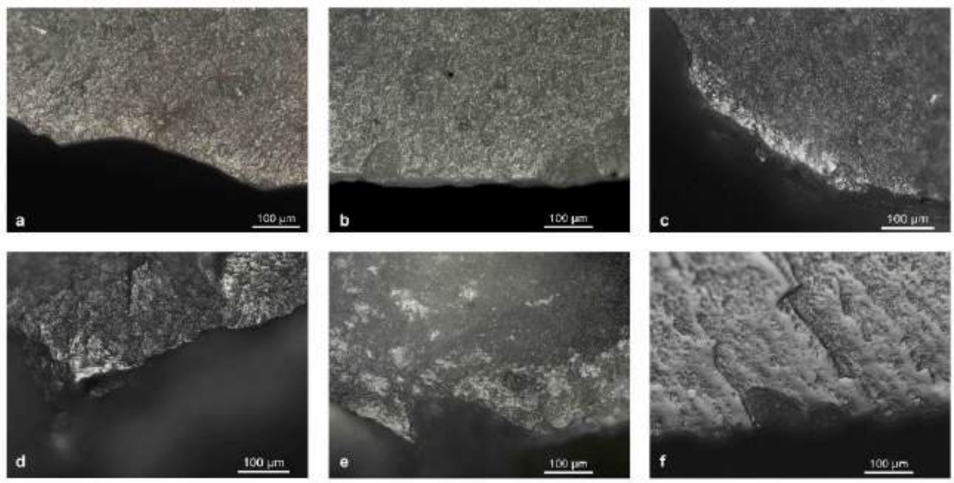

Figure 2.2: Selection of use-wear traces on experimental flint tools. a) Light edge-rounding and greasy band of polish from scraping fresh hide (200x); b) Edge-rounding, isolated edge-removals and band of polish from cutting fresh hide (200x); c) Line of polish and light rounding from scraping fresh bone (200x); d) Domed polish with longitudinal directionality from sawing fresh bone (200x); e) Edge-removal with longitudinal orientation and flat smooth polish from sawing fresh bone (200x); f) Edge-removal oriented longitudinally and highly-linked band of polish from cutting reeds (200x).

Chert

<u>Hide:</u> Traces from contact with fresh hide consist of edge-rounding, edge-removals, and polish. Scraping and cutting resulted in a light edge-rounding. Edge-removals occurred only on tools used for cutting. A rough and greasy polish developed on all scraping and cutting tools. On endscrapers, a continuous band of polish -with transverse directionality- formed (Fig. 2.3a). While on cutting tools, the polish has a more localized distribution but, it is still invasive (Fig. 2.3b). Pits and craters in the polish were documented on one endscraper.

<u>Bone:</u> Scraping fresh bone caused a very light edge-rounding and a few edge-removals. A smooth and matt polish is distributed at the very edge of the endscrapers, while a lightly developed greasier polish extends more into the piece (Fig. 2.3c). Sawing caused more edge-removals than scraping. The continuous crushing of the edge inhibits the formation of edge-rounding. Bone

polish developed in isolated spots and has a smooth and matt texture and a domed topography. Tiny pits are visible in the polish (Fig. 2.3d, e). The polish is bright and has a clear longitudinal directionality. Striations parallel to the edge were documented on one flake.

Reed: Working fresh reeds resulted in edge-removals, lightly developed edge-rounding, and a wide band of polish with a smooth and matt texture and domed topography (Fig. 2.3f). The polish is very bright and displays a longitudinal directionality. The degree of linkage of the polish is higher on the flake used for 60 minutes.

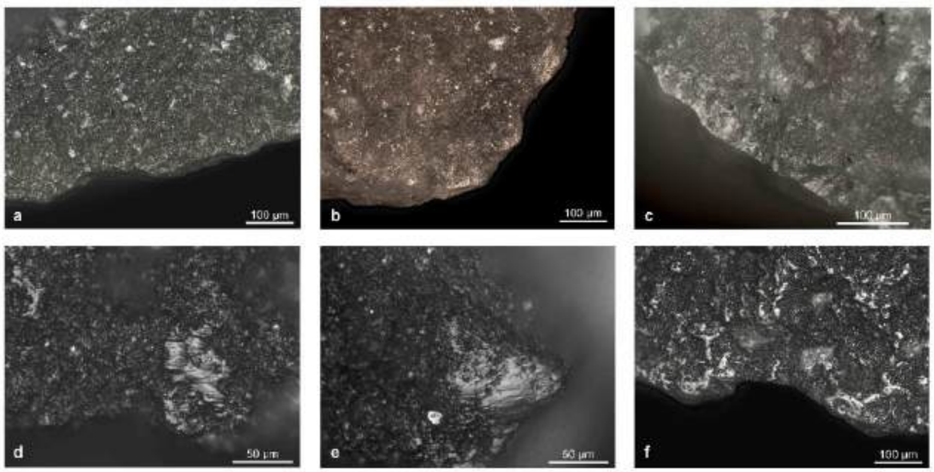

Figure 2.3: Selection of use-wear traces on experimental chert tools. a) Edge-rounding and greasy band of polish from scraping fresh hide (200x); b) Edge-rounding and polish from cutting fresh hide (200x); c) Line of polish from scraping fresh bone (300x); d) Isolated spots of polish with longitudinal directionality from sawing fresh bone (500x); e) Edge-removals and smooth spot of polish with longitudinal directionality and tiny pits from sawing fresh bone (500x); f) Edge-removal oriented longitudinally, edge-rounding and invasive smooth polish from cutting reeds (200x).

Dolerite

Hide: Traces from contact with fresh hide are edge-rounding, edge-removals, and polish. Edge-removals were documented only on cutting tools. Hide polish developed on endscrapers and flakes and its distribution is not uniform along the used edge. Polish developed in isolated patches with a granular texture and a flat topography (Fig. 2.4a, b). On endscrapers, the polish is distributed more continuously along the used edge than on flakes. Hide polish developed both on crystals and on the matrix.

Bone: Working fresh bone resulted in edge-removals, edge-reduction, polish, abrasion, and striations. A light edge-rounding was observed on both endscrapers, while on the flakes, it developed only on the one used for 60 minutes. Severe edge damage occurred during the sawing activity. On both endscrapers and flakes, the polish has a localized distribution, and it developed on the most protruding areas of the edge. The polish has a smooth and matt texture and a domed topography (Fig. 2.4c, e). Pits are occasionally visible in the polish. Fine and small striations perpendicular to the working edge are visible on the surface of a worn protruding crystal on endscraper 3806 (Fig. 2.4d). On both endscrapers, abrasion of the crystals is visible. Abrasion is lightly developed since no very abrasive materials were worked. The hollows are mostly medium-sized and irregular in shape in line with what has been reported in the literature for tools used to process medium and hard materials (cf. Clemente-Conte et al., 2015).

Reed: Traces from contact with fresh reeds consist of rare edge-removals, light edge-rounding, smooth and matt polish with a domed topography, and abrasion (Fig. 2.4f). The polish is invasive and distributed in isolated spots along the working edge. The polish lays on top of the crystals and grains of the matrix, the brightness is very intense, and the polish displays a clear longitudinal directionality. Abrasion is visible on a few crystals along the edge and is lightly developed.

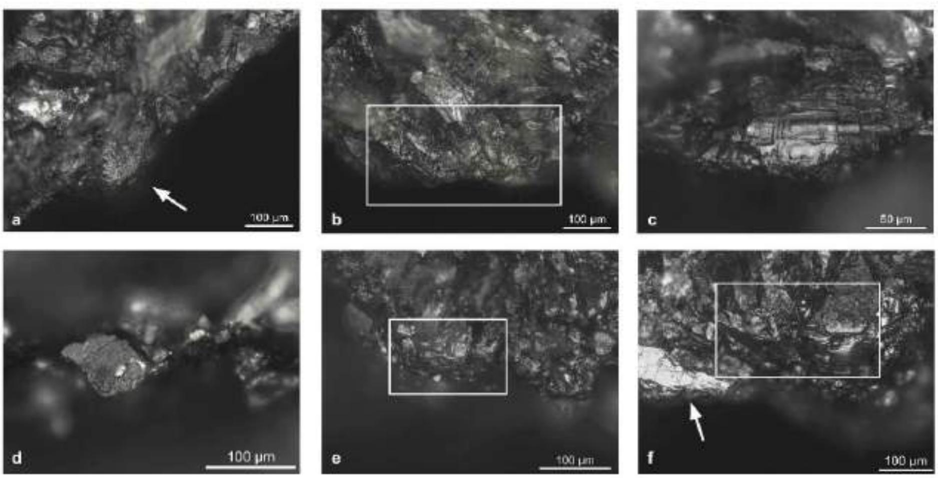

Figure 2.4: Selection of use-wear traces on experimental dolerite tools. a) Edge-rounding and polish with granular texture (white arrow) from scraping fresh hide (200x); b) Edge-rounding and granular polish from cutting fresh hide (200x); c) Smooth polish with transverse directionality from scraping fresh bone (500x); d) Abrasion and striations perpendicular to the edge on a protruding crystal from scraping fresh bone (300x); e) Edge-rounding and domed polish with longitudinal directionality (white square) from sawing fresh bone

(300x); f) Edge-damage, invasive smooth polish (white square) and light abrasion (white arrow) from cutting reeds (200x).

Quartz

Hide: Traces from contact with fresh hide consist of edge-removals, edge-rounding, polish, and abrasion. The degree of rounding varies between light to moderate (Fig. 2.5a). Edge-removals are rare and isolated. Hide polish did not develop on all tools. When present, the polish is distributed in isolated spots or a band along the edge and has a rough and greasy texture (Fig. 2.5c). Polish is mostly located on the dorsal cortical exterior of the flake used for cutting hide. Abrasion developed on endscrapers and flakes (Fig. 2.5b). Due to the state of the worked material (fresh hide), the degree of abrasion is mostly light with small-sized hollows (cf. Clemente-Conte et al., 2015).

Bone: Fresh bone traces -consisting of edge-removals, edge-rounding, polish, and abrasion- did not develop on all tools. Only one endscraper displays traces of use. Scraping fresh bone caused a light edge-rounding and isolated edge-removals. Polish -with transverse directionality- developed in isolated spots at the very edge and abrasion was documented in a few locations (Fig. 2.5d, e). Cutting produced more edge-removals than scraping and very light rounding. A smooth polish, with pits and comet tails, developed on the cortical exterior of one cutting tool and has a localized distribution (Fig. 2.5g). The degree of abrasion is higher on tools used to work bone compared to those used on hide, and the hollows are medium-sized and irregular in shape (Fig. 2.5f) (cf. Clemente-Conte et al., 2015).

Reed: Cutting fresh reeds resulted in isolated edge-removals, light rounding, and a band of smooth and matt polish with a domed topography (Fig. 2.5h, i). The brightness of the polish is very intense and displays a longitudinal directionality. Reed polish is clearly visible both on the cortical exterior and crystalline surface of the tools. However, on the crystalline surface, the polish distribution is less uniform due to the uneven topography, but it is still invasive.

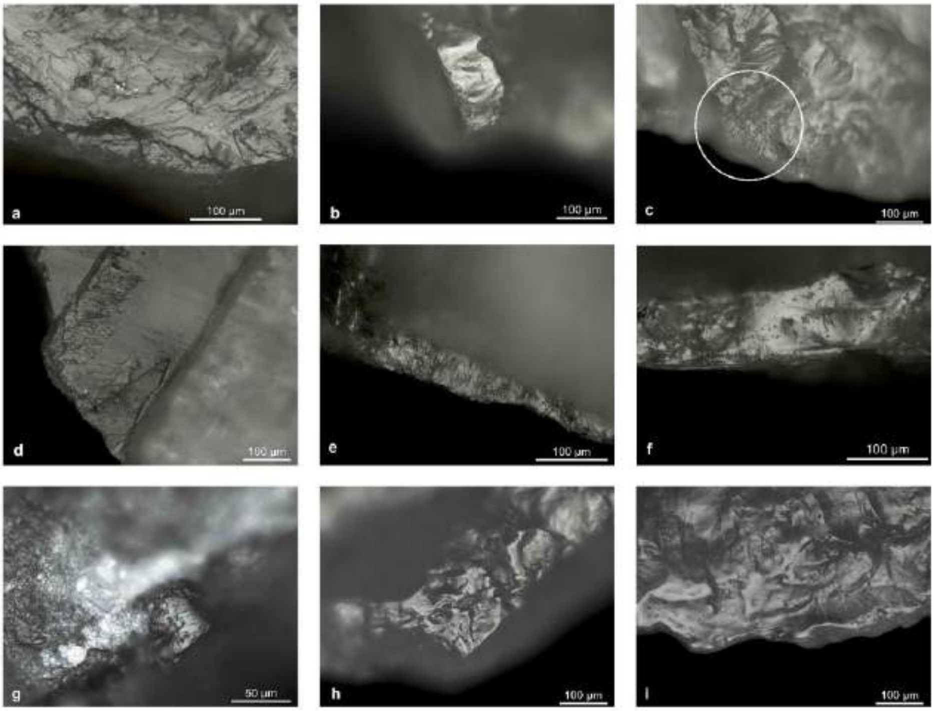

Figure 2.5: Selection of use-wear traces on experimental quartz tools. a) Edge-rounding and polish from scraping fresh hide (200x); b) Polish and abrasion from cutting fresh deer hide (200x); c) Polish (circle) from cutting fresh hide (200x); d) Edge-rounding and moderately developed abrasion from scraping fresh deer bone (200x); e) Polish and linear features with transverse orientation from scraping fresh bone (300x); f) Edge-removals and moderately developed abrasion from sawing fresh bone (300x); g) Moderately developed spot of polish displaying tiny pits/comet tails (500x); h-i) Edge-removals and fluid polish from cutting reeds (200x).

Tool effectiveness

Raw materials properties (such as hardness, roughness, and toughness) and the shape of the tools can influence tool efficiency, and this is directly related to edge maintenance. For use-wear analysis, this means that some raw materials and tool types may have a limited or typical build-up of use-wear. For example, in a brittle raw material, working edges with wear traces may continuously collapse resulting in a limited accumulation of traces. Therefore, I also recorded the degree of tool effectiveness.

I did not observe differences in the effectiveness of flint and chert tools while processing different materials. Flake tools and endscrapers were highly effective in the various activities. Only one chert flake used to cut fresh hide was ineffective. However, chert's lower efficiency depended mainly on the convex shape of the working edge, which is not particularly suited for cutting. The development of edge modifications did not significantly affect the performance of tools.

Not all the dolerite tools proved effective. I noticed a clear difference in effectiveness between retouched and unretouched implements. Endscrapers were overall less effective than flake tools and thick scraper-heads performed worse than thin ones. Endscraper effectiveness further decreased during use since edge-rounding and edge-reduction caused a rapid increase of the active edge angle. Conversely, unmodified flakes were effective cutting tools. However, the effectiveness of flakes used to saw bones rapidly declined during use due to continuous edge-crushing. Edge modifications were mainly caused by the loss of grains rather than fracturing (cf. Gibaja Bao et al., 2009).

Quartz tools were overall effective in the different activities, except for two cutting tools. Exp. 3822 was selected to process fresh bone. However, this tool has a granular internal structure, and during the use the edge quickly crumbled. The convex indented lateral edge of exp. 3823 was unsuitable for cutting fresh hide. The efficiency of the latter depended mainly on the shape of the active edge rather than on the characteristics of the raw material. Due to the hardness of quartz, edge-rounding and edge-reduction formed slower compared to other rock types. That allowed quartz tools to retain their efficiency for a longer period of use. However, the edges tend to break prematurely compared to flint and chert.

Discussion

Comparisons between non-flint and flint tools

Flint - chert comparison

Chert and flint are both microcrystalline varieties of quartz. Due to their similarities, the development of and the traces themselves are thus expected to be similar (SI). According to Nieuwenhuis (2002), the characteristics of use-wear traces on chert, especially coarse-grained chert, were to some extent different from flint. The result of her experiment showed that traces on chert tools are comparable with those on flint tools, but less extensively developed, especially polish (Nieuwenhuis, 2002, p. 36). My study underlines this conclusion. Due to the uneven micro-topography of chert, especially on tools used to process medium and medium-hard materials (reeds and fresh bone), the polish started to develop on the higher areas of the micro-surface and extended gradually on the lower parts through use. Therefore, polish spots have a more localized distribution and a lower degree of linkage when compared to polish on flint (Fig. 2.6).

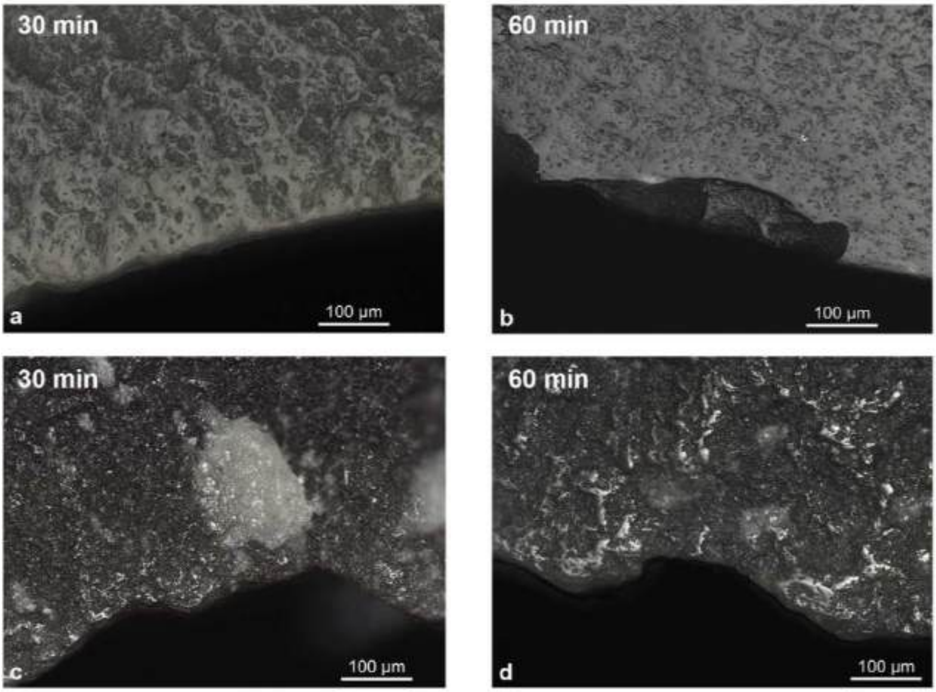

Figure 2.6: Comparison between reeds polish on flint (top a-b) and chert (bottom c-d) distribution and degree of linkage after 30 and 60 minutes of use. Magnification 200x.

Polish distribution, texture, and topography on chert tools are consistent with flint (Fig. 2.7a, b, c). However, polish distribution on chert hide-cutting tools differs slightly from what was observed on flint tools. Instead of a continuous band of polish along the edge, a few spots of moderately developed polish were documented on protruding locations, while a light polish is more spread along the edge (Fig. 2.7a). On both rock types, hide polish has a rough and greasy texture, while bone and reeds polishes have a smooth and matt texture (Fig. 2.7b). Deep craters in the polish, associated with hide as a contact material (Van Gijn, 1990), were documented only on tools used to scrape hide. Tiny pits in the polish are visible both on hide working and bone working tools. Bone and reeds polishes mainly display a domed topography. Only on flint, some heavily developed spots of bone and reeds polish have a flat topography (Fig. 2.7c). The difference in polishes observed on experimental chert tools is thus quantitative rather than qualitative. Experimental traces on chert tools can be interpreted based on the ones observed on the flint reference collection, although a specific chert reference collection would be preferred.

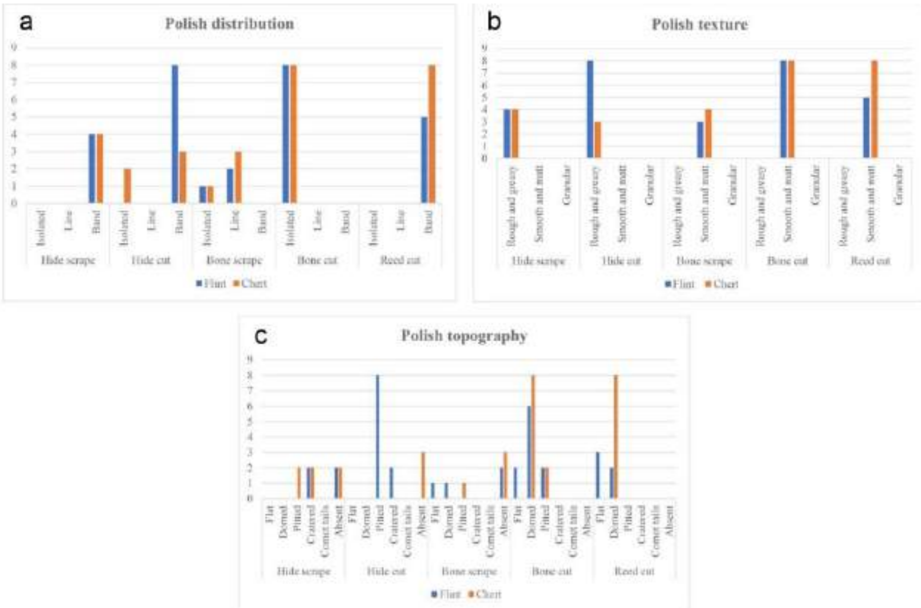

Figure 2.7: a) Column charts displaying polish distribution on experimental flint (blue) and chert (orange) tools. Polish distribution is consistent between flint and chert tools except for chert hide-cutting tools on which the polish has a more localized distribution. Numbers represent the sum of locations on the tool (see section 2.4) where the polish characteristic was observed; b) Column charts displaying polish texture on experimental flint (blue) and chert (orange) tools. Polish texture is consistent between flint and chert. Numbers represent the sum of locations on the tool (see section 2.4) where the polish characteristic was observed; c) Column charts displaying polish topography on experimental flint (blue) and chert (orange) tools. On both rocks craters occur only in hide polish, while pits on hide and bone polish. A domed polish is characteristic of bone and reed as contact materials on both rocks. Numbers represent the sum of locations on the tool (see section 2.4) where the polish characteristic was observed.

<u>Flint – dolerite comparison</u>

Despite their great abundance, specific studies on wear traces on dolerite tools are rare. Dolerite, like quartzite, is a heterogeneous rock therefore, data were interpreted in the light of studies conducted on other heterogeneous rocks and the few available on dolerite and basalt (e.g., Bello-Alonso et al., 2020; Bello-Alonso et al., 2019; Clemente-Conte & Gibaja Bao, 2009; Clemente-Conte et al., 2015; Huet, 2006; Lemorini et al., 2019; Pedergnana & Ollé, 2017; Wadley & Kempson, 2011). On heterogeneous rocks, such as dolerite, edge removals – considered indicative of the hardness of the worked material (Tringham et al., 1974) – are less clear than on flint. When the scars are present, their morphological characters such as shape, initiation, and termination are not easily recognizable due to surface reflectiveness, micro-topography unevenness, and, secondarily, problems in the depth of field (cf. Pedergnana & Ollé, 2017). Rounding developed on the tool only after the active edge had stabilized. Prior to that, the continuous microflaking of the edge hindered the formation of rounding (cf. quarzite tools, Clemente-Conte & Gibaja Bao, 2009). Other experiments conducted with the *Dolérite du Trieux* (a formation from the North Armorican Massif, France) pointed out the relative fragility of dolerite tools' cutting edges. Mechanical tests showed that hardness measurements are much lower on dolerite compared to flint and quartz, which results in a rapid deterioration of the used edge (Huet, 2006) (also see SI). Regarding microwear, both from my experiment and the literature (Clemente-Conte & Gibaja Bao, 2009; Clemente-Conte et al., 2015), I can conclude that polish develops on dolerite slower than on flint, and its distribution is usually localized or restricted to small areas since only higher reliefs of the surface are in direct contact with processed material. The main difference between flint and dolerite concerns hide polish (Fig. 2.8).

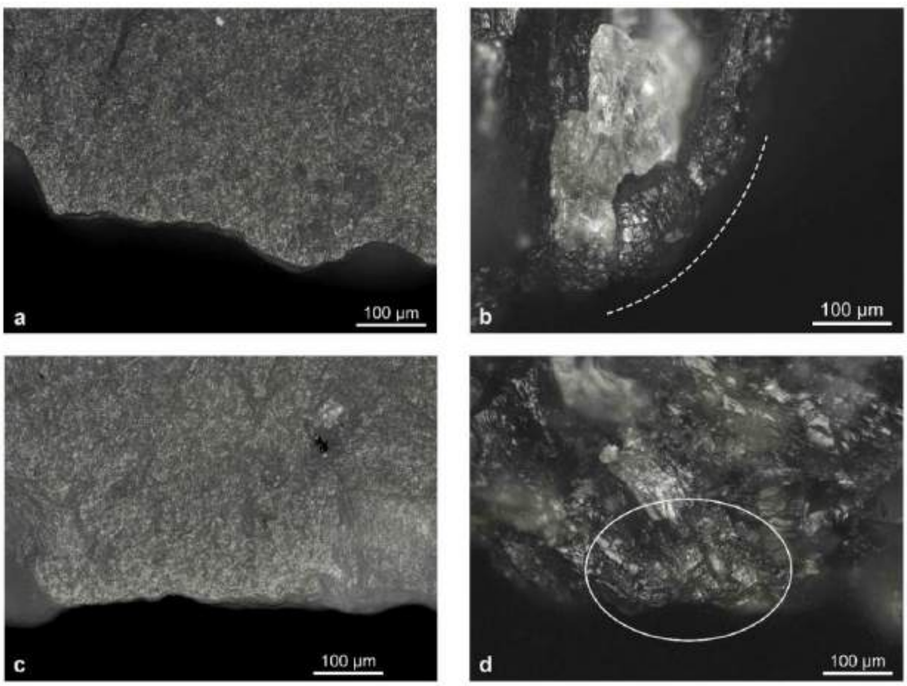

Figure 2.8: Comparison between fresh hide polish on flint endscraper (right, a) and flake tool (right, c) and dolerite endscraper (left, b) and flake tool (left, d). Magnification 200x.

On flint, hide polish is distributed in a continuous band along the used edge. However, on dolerite tools, hide polish developed in isolated spots, both on the crystals and the matrix. A continuous band of light polish is visible only on hide-scrapers (Fig. 2.9a). On dolerite, hide polish is mainly characterised by a granular texture while on flint is mostly rough and greasy (Fig. 2.9b). Hide polish topography is flat on dolerite tools, and the characteristic pits or deep craters observed on hide polish on flint are absent (Fig. 2.9c). On dolerite bone working tools, polish formed only on top of crystals and grains on the highest locations of the surface. On tools used to cut reeds, the polish developed in isolated spots on the crystals and the matrix and extended inside the piece (Fig. 2.9a). On both flint and dolerite, the texture of both bone and reeds polishes is mostly the same (Fig. 2.9b). Dolerite bone-cutting tools mainly display no topography features since polish spots are usually not large enough to display topographical features. On endscrapers, a domed topography was documented. On flint, a domed topography and tiny pits are considered characteristic of bone as contact material (Keeley, 1980; Van Gijn, 1990). In the comparison, reeds polish displays a similar domed topography (Fig. 2.9c).

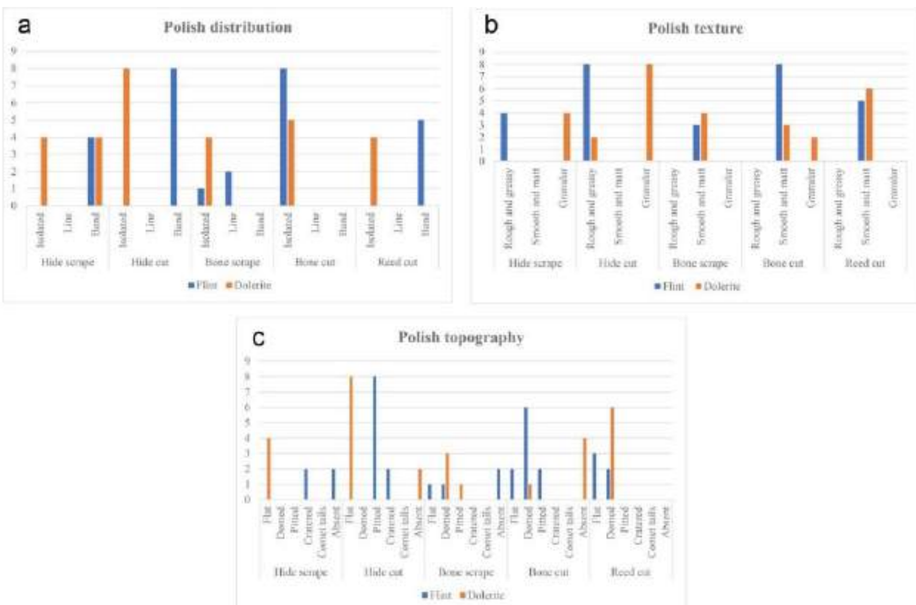

Figure 2.9: a) Column charts displaying polish distribution on experimental flint (blue) and dolerite (orange) tools. On dolerite tools used on hide and reeds, the polish is mainly distributed in isolated spots, while on flint in a continuous band along the edge. Numbers represent the sum of locations on the tool (see section 2.4) where the polish characteristic was observed; b) Column charts displaying polish texture on experimental flint (blue) and dolerite (orange) tools. On dolerite hide-working tools, the polish has mainly a granular texture, while on flint is rough and greasy. Polish on flint and dolerite tools used on bone and reed displays a domed topography.

Numbers represent the sum of locations on the tool (see section 2.4) where the polish characteristic was observed; c) Column charts displaying polish topography on experimental flint (blue) and dolerite (orange) tools. On dolerite hide-working tools the polish has mainly a flat topography, while on flint is either pitted or cratered. Polish topography on dolerite tools used on bone and reeds is consistent with flint. Numbers represent the sum of locations on the tool (see section 2.4) where the polish characteristic was observed.

The characteristics of macro and microwear traces on dolerite tools partially overlap with those on flint. Micro polishes develop in general slower than on flint, and because of the irregular microtopography of the surface, the distribution is isolated, and degree of linkage limited. The difference in bone and reeds polishes on dolerite is mainly quantitative (less polish in localised areas), while for hide polish it is quantitative and qualitative (also a different appearance). Although hide polish texture and topography are different from flint, the invasiveness of the polish, its location both on high and low locations of the edge, and the association with rounding all point unmistakably to soft materials processing. The greasy appearance of some spots of polish recalls animal materials rather than vegetal. Thus, the identification of the type of contact material remains feasible. A flint reference collection is useful in the identification of wear traces on dolerite tools. However, in some cases, additional specific experiments may be necessary for a more confident interpretation of the worked material mainly because of the variation of dolerite rocks' composition and grain size which can affect wear traces.

<u>Flint – quartz comparison</u>

Due to their differences in structure and mineralogical composition, quartz and flint have very different responses to mechanical stress (SI). That affects use-wear traces formation on these rocks. The experiment highlighted a clear difference between traces on the quartz crystal surface and the unflaked cortical exterior. Polishes, with very similar characteristics to flint, mostly form on the cortex/neo-cortex (Fig. 2.10), while the quartz crystal surface mainly displays plastic deformations, linear features, and abrasion. As reported in the literature (Clemente-Conte & Gibaja Bao, 2009; Knutsson et al., 2015), micro-polishes occur on quartz less frequently and much slower than on flint except for activities involving silica-rich materials, which result in a considerable amount of polish development. Therefore, unlike flint, where use is predominantly identified based on polish characteristics, the use of quartz tools must be identified based on other evidence as well.

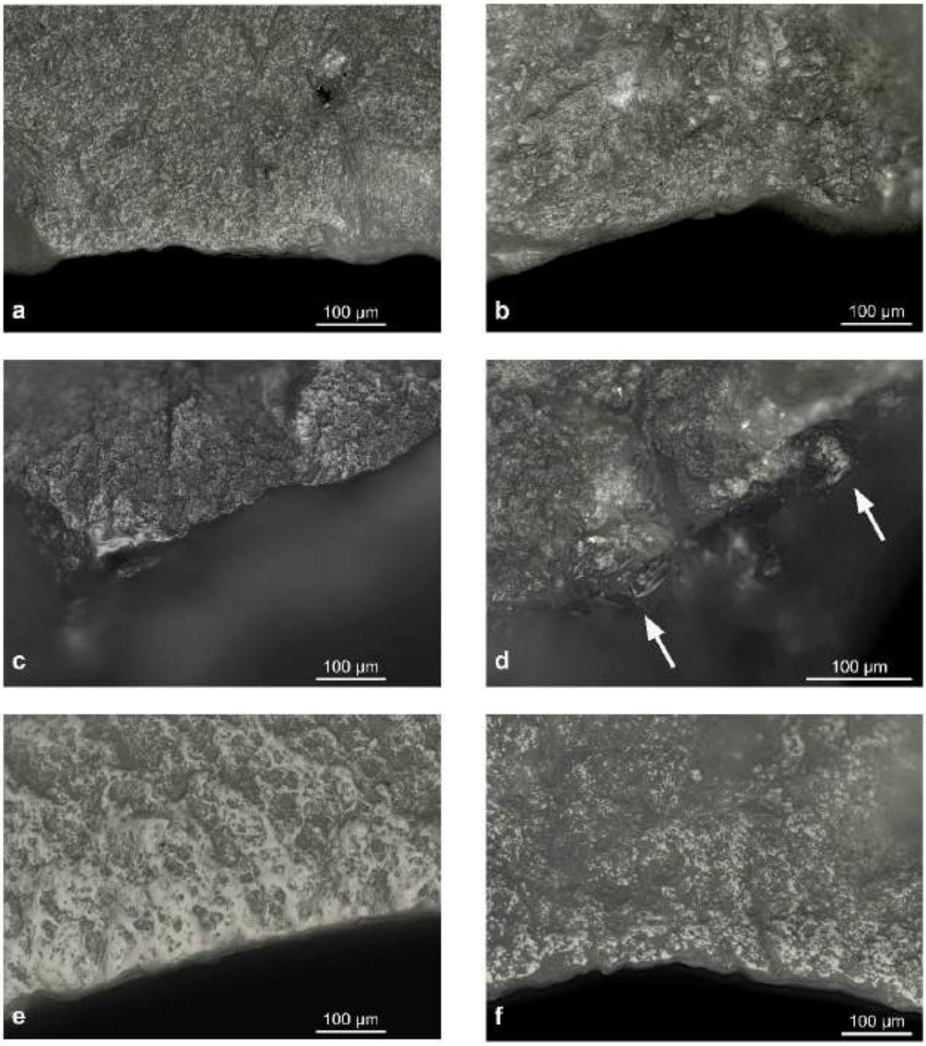

Figure 2.10: Comparison between polishes on experimental flint tools (a, c, e) and on the unflaked cortical exterior of experimental quartz tools (b, d, f). a-b) Fresh hide polish; c-d) Fresh bone polish; e-f) Fresh reed polish. a, b, c, e, f) magnification 200x; d) magnification 300x.

The main difference concerning polish distribution is represented by hide polish. On flint, hide polish tends to develop in a continuous band along the used edge, while on quartz, on small and isolated spots. However, on the cortical side of hide-cutting tools, polish distribution is more continuous (Fig. 2.11a). No differences in polish texture were noticed between flint and quartz (Fig. 2.11b). Hide polish displays a rough and greasy texture, while bone and reeds polishes are smooth and matt. Polish topography is mainly absent, especially on tools used to process fresh hide. A few spots of polish on the cortical exterior of the bone cutting tool have a domed topography. Pits and come tails, which are characteristics of bone as a contact material on flint (Keeley, 1980), were documented. Reeds polish topography is domed on both rocks (Fig. 2.11c).

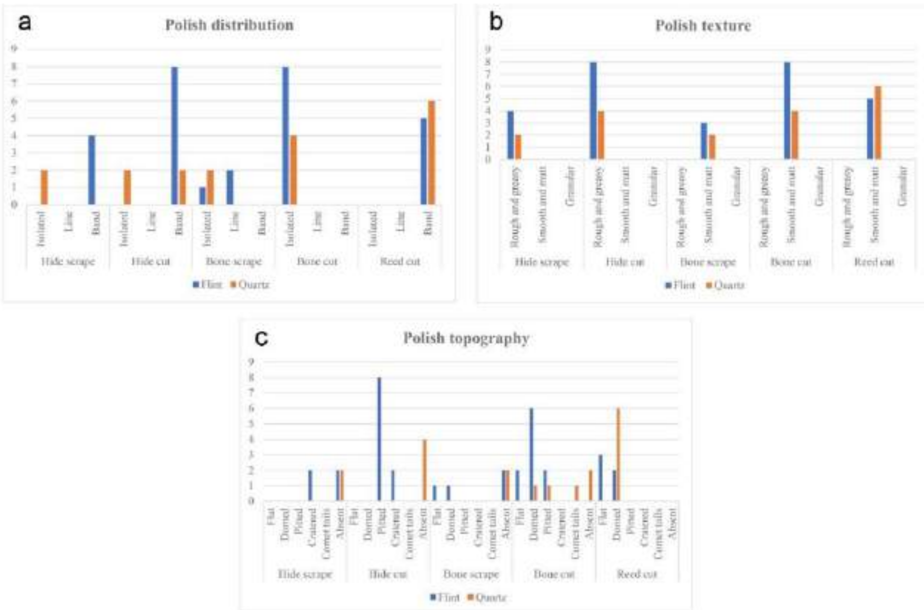

Figure 2.11: a) Column charts displaying polish distribution on experimental flint (blue) and quartz (orange) tools. On quartz hide-working tools the polish is mainly distributed in isolated spots, while on flint in a continuous band along the edge. Numbers represent the sum of locations on the tool (see section 2.4) where the polish characteristic was observed; b) Column charts displaying polish texture on experimental flint (blue) and quartz (orange) tools. Polish texture is consistent between flint and quartz tools. Numbers represent the sum of locations on the tool (see section 2.4) where the polish characteristic was observed; c) Column charts displaying polish topography on experimental flint (blue) and quartz (orange) tools. Polish topography is mainly absent on quartz tools except for the ones used to cut reeds. Polish from contact with reeds displays a domed topography. Numbers represent the sum of locations on the tool (see section 2.4) where the polish characteristic was observed.

Use-wear traces on quartz display distinctive characteristics, which are hard to interpret using only a flint comparative collection. By comparing the degree of edge-rounding and the amount of edge damage on quartz and flint tools, it may be possible to assess the hardness of the worked materials (cf. Semenov, 1964). But for a more precise interpretation of the contact materials based on quartz-specific wear traces (i.e. abrasion and striations), separate experiments are needed. Nevertheless, the results highlight the value of analysing also the cortical surface of quartz tools. Micro-polishes that develop here are comparable with the ones on flint tools and can aid interpretation.

Functionality

As pointed out in the results, tools made of different rocks varied in effectiveness in the same activities. The recognition of existing differences in tools' efficiency may have influenced prehistoric populations' choices in the selection of the raw materials for their tools. Flint and chert tools were overall the most effective. These rocks are easy to knap and produce implements with sharp edges easy to shape and maintain. Dolerite unretouched flakes were more effective than endscrapers. The presence of flakes with natural indented edges makes them particularly suited for cutting even though these edges became blunted soon when used to process hard materials. The degree of effectiveness of dolerite endscrapers was closely related to the shape of the functional edge. Scraper-heads were difficult to retouch and shape precisely because dolerite, like other tough and coarse-grained rocks, is prone to crushing rather than fracturing, making it difficult to control the direction of the removals (Wadley & Kempson, 2011). Because of that, dolerite may have been preferred for unmodified tools rather than retouched ones. That is the case of Sibudu Cave, where unretouched implements were mostly made of dolerite, while retouched tools with hornfels and quartz (Wadley & Kempson, 2011). Despite their small dimensions, quartz flakes and endscrapers were functional in all the activities. No differences in functionality between unretouched and retouched tools were observed. From good-quality blocks of quartz, it is possible to obtain tools with sharp and robust functional edges. Despite the obtuse angle of the scraper-heads, quartz endscrapers proved effective. This was because endscrapers remained sharp and functional due to the slow progression of wear damage on the working edge. A limitation in the use of quartz as raw materials for tools may lie in the small dimensions of the products due to its high fragmentation proneness during knapping (Tallavaara et al., 2010).

Limitations and archaeological visibility of traces

My experiments showed that use-wear traces, especially polishes, develop slower on non-flint rocks than on flint, especially on dolerite and quartz. Hence, archaeological non-flint tools briefly used may not display sufficiently developed use-wear to allow a functional interpretation.

That applies particularly to dolerite and tools made from brittle rocks in general, where the continuous crushing of the active edge affects the formation and recognition of use-wear traces since they are constantly removed during use and preserved only on little spots. Due to that, even artifacts with a long use life may display only light evidence of use or no wear traces (cf. Pedergnana & Ollé, 2017). For flint and chert, we can expect traces to accumulate through use unless they are intentionally removed by resharpening. A loss of wear traces can happen during the processing of hard contact materials. However, edge-damage rarely removes all the evidence of use. For quartz, the slow formation rate of wear traces may lead to a misidentification or misinterpretation of expedient tools.

In addition, my experiment showed that the edges of dolerite tools wore down rapidly, and resharpening sessions were needed to extend their use cycle. However, in areas where lithic raw materials were abundant, flakes may have been quickly abandoned and replaced when dull, especially if made of non-homogeneous rocks unsuited for retouching. That is the case, for instance in New South Wales, Australia, where knappable raw materials, like silcrete, quartz and quartzite, are locally available. Ethnographic accounts have shown that Australian Aboriginals prefer to replace their quartz tools when exhausted rather than invest time in rejuvenating them (Holdaway & Douglass, 2015). Even though resharpening tends to remove traces from previous use sessions, well-developed spots of use-wear may survive in between resharpening scars (e.g., Loebel, 2013). Conversely, discard of tools in an early stage of use means traces remain poorly developed. In addition, when used to process hard materials, dolerite and quartz tools displays small, localised areas with traces that can be easily missed during the analysis. As already stated in the literature (e.g., Clemente-Conte et al., 2015), the examination of wear-trace characteristics on single crystals and grains within the matrix requires higher magnifications compared to flint and possibly other analytical techniques such as SEM and digital microscopy. Thus, the analysis of heterogenous rocks is more time consuming than of flint. Furthermore, mechanical or chemical post-depositional alterations may remove or obliterate traces of use (Van Gijn, 1990). However, several studies showed that quartz appears more resistant to post-depositional modification than flint highlighting the potential of this material in contributing to our knowledge on activities of prehistoric people (Clemente-Conte et al., 2015; Lazuén et al., 2011; Venditti et al., 2016). Therefore, it is likely that the number of used non-flint artefacts is underestimated. The state of preservation of the material but also the mineralogical composition and mechanical properties of rocks featuring the lithic assemblage are all factors that could favour the identification of traces of use on knapped stone tools or explain their absence, and they should be considered by the analyst.

Conclusion

This experiment tested the possibility of using a flint reference collection to interpret use-wear on different rock types. Experimental wear traces on European flint tools were compared with wear traces on chert, dolerite and quartz. No major differences between flint and chert were observed. Use-wear on chert can be confidently interpreted using a reference collection of flint tools if the wear traces are developed enough. Even though I noted several differences in the distribution and characteristics of wear traces between dolerite and flint, a flint reference collection can allow a general interpretation of the use motion and hardness of the worked material. The precise interpretation of the contact material may be more problematic except for dolerite tools used to process siliceous plants. I observed strong similarities between the use polish on quartz cortical exterior and flint. However, the function of quartz tools cannot be inferred only using a flint reference collection. The degree of edge-rounding and abrasion and the frequency and distribution of edge-removals can help in the identification of the hardness of the contact material, but specific experiments are required for a more precise interpretation of the worked material.

Flint and chert tools performed best in all the various activities. The edges retained their functionality for a longer period of use, and wear traces developed sufficiently to allow a confident interpretation. Dolerite unretouched tools were more effective than retouched ones. Yet the edges of dolerite tools were the most fragile overall. They wore and crumbled, resulting in wear traces that were scattered and limited to isolated spots. Quartz tools were effective and wore slower compared to flint, chert and dolerite. However, the performance of quartz tools is influenced by the structure of the block. Internal discontinuities, micro-fractures or inclusion may lead to premature breaking of the working edge.

The direct comparison of use-wear on tools made from different rocks allowed me to observe how traces from the same contact material developed at different rates based on the tool's raw material. In addition, the observation of rock-specific mechanism of wear (e.g., continuous edge-crushing on dolerite) helped explain the limited presence, or absence of wear traces on used tools. The study shows that a partial overlap exists between the use-wear features on European flint and chert, dolerite, and quartz. These need to be considered in light of the different properties and characteristics of the rocks to achieve a correct functional interpretation.

Acknowledgements

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Chapter 3

Exploring South African compound adhesives

"A multi-analytical approach reveals flexible compound adhesive technology at Steenbokfontein Cave, Western Cape"

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Supplementary Information:

Abstract

Evidence of different compound resin-based adhesives is present in South Africa from at least 77,000 years ago. Ancient glue production is considered one of the oldest known highly complex technologies, requiring advanced technological and mental abilities. However, our current knowledge of adhesive materials, recipes, and uses in South Africa is limited by the lack of in-depth analysis and molecular characterization of residues. To deepen our knowledge of past adhesive technology, we performed a detailed multi-analytical analysis (use-wear, XRD, μ-CT, IR spectroscopy, GC-MS) of 30 Later Stone Age tools with adhesive remains from Steenbokfontein Cave, South Africa. At the site, tools made of various rocks were hafted with compound adhesives, and we identified three recipes: 1) resin/tar of *Widdringtonia* or *Podocarpus* species combined with hematite; 2) resin/tar of *Widdringtonia* or *Podocarpus* species mixed with hematite and another plant exudate; 3) resin/tar without hematite. The studied scrapers were used in hide-working activities, and the studied cutting tools were used to work animal and soft plant matters. All scrapers display evidence of intense resharpening and were discarded when no longer usable. The combination of different methods for residue analysis reveals the flexibility of adhesive technology at Steenbokfontein. Despite the consistent use of conifer resin/tar throughout the sequence, we observed that other ingredients were added or excluded independently of the tools' raw materials and functions. Our results highlight the long-lasting tradition of using adhesive material from conifer species but also the adaptability and flexibility of adhesive traditions. The systematic application of this multi-analytical approach to Pleistocene adhesives will be useful to better characterise adhesive traditions and enhance the debate on the technological, cognitive, and behavioural implications of this technology.

Introduction

Evidence for different adhesives, including compound adhesives, is present in South Africa from at least ~77,000 years ago (Rots et al., 2017; Wadley et al., 2009). Compound adhesives consist of multiple ingredients. The main ingredient is the tackifier that provides the stickiness like a resin or tar, and sometimes plasticisers are added to make the adhesive less brittle or more pliable. Examples of the latter are beeswax and fat. Other ingredients like ochre, sand, and fibres are added to increase strength, durability, and pliability (Langejans et al., 2022). The manufacture of compound adhesives requires considerable technical and cognitive skills, including an understanding of chemical reactions, the use of pyrotechnology, abstraction, recursion, and cognitive fluidity (Wadley, 2010; Wadley et al., 2009). The identification of differences in the composition of compound adhesives in the archaeological record has been viewed in relation to the various raw materials from which tools are made of and tools uses. This evidence highlights the versatility of prehistoric resin-based adhesives (Lombard, 2007; Wadley et al., 2015).

While there are several reports on adhesive remains from South African assemblages dated to the Middle Stone Age (MSA) and Later Stone Age (LSA), few of these performed chemical studies for secure identification. Adhesives have largely been studied through microscopy by documenting their distinctive morphologies and systematically mapping of their spatial distribution and association with use-wear traces (e.g., Gibson et al., 2004; Lombard, 2006; Lombard, 2007). However, several studies have emphasised the limitations of interpretation based solely on residues morphology and distribution patterns (Pedergnana, 2020; Pedergnana et al., 2016). Even when adhesive residues are correctly identified, optical microscopy alone cannot securely differentiate between plant exudates of different species (Soriano et al., 2015). Overall, the paucity of molecularly identified Stone Age adhesive residues limits our understanding of this technology. Without knowing the basic ingredients, additives, loading agents, and production methods of adhesives, current inferences on the complexity of compound adhesives are hard to validate.

The organic components of adhesives are occasionally identified with gas chromatography-mass spectrometry (GC-MS). To date, only five MSA residue samples were analysed by GC-MS: a quartz flake from Diepkloof Rock Shelter

(Charrié-Duhaut et al., 2013) and four segments, one from Rose Cottage Cave, and three from Sibudu Cave (Soriano et al., 2015) (Fig. 3.1). *Podocarpus* resin, possibly mixed with bone and quartz, was identified at Diepkloof, while only one of the segments from Sibudu provided evidence of a conifer resin (e.g., *Podocarpaceae* sp.) used to enable hafting. For the LSA, three artefacts with macroscopic residue from Elands Bay Cave (Charrié-Duhaut et al., 2016) and three microliths from the early LSA of Border Cave (Fig. 3.1) (Villa et al., 2012) were chemically analysed. GC-MS identified the residues as an adhesive, either resin or tar, made from species belonging to the *Podocarpaceae* family, likely *Podocarpus elongatus*. In the case of Elands Bay Cave, the adhesive was possibly mixed with organic and inorganic additives such as fat and quartz grains (Charrié-Duhaut et al., 2016). More adhesive residues from several South African LSA sites were analysed with GC-MS by Veall (2019), revealing the presence of compound adhesives produced from plant exudates, such as conifer resin and pitch and latex, and mixed with organic and inorganic additives such as fat, waxes, and crushed minerals. Overall, the sample size of analysed adhesives per site is limited, complicating a deep diachronic, regional, and technical understanding of adhesive technology.

Other methods of studying both the organic and inorganic fractions of adhesive residues, occasionally used in combination with GC-MS, include scanning electron microscopy (SEM), Raman, and infrared spectroscopy (Charrié-Duhaut et al., 2016; Villa et al., 2015; Wojcieszak & Wadley, 2018). Although GC-MS is more sensitive and capable of accurate detection of specific organic compounds, these other methods have the advantage of being non-invasive, relatively cheap, and quick (cf. Shillito et al., 2009). Despite the increasing popularity of chemical studies in residue analysis, the lack of a systematic molecular identification of alleged adhesive remains on South African tools, and particularly within assemblages, identified only by means of optical microscopy still represents a drawback in the field.

To gain more information on adhesive production and use during the South African Stone Age and enhance the discussion on the complexity of adhesive technology, we analysed a sample of 30 LSA artefacts with macro-residues from Steenbokfontein Cave, Western Cape (Fig. 3.1). Despite never being chemically analysed, based on residues' characteristics and distribution and the presence of two previously identified adhesive finds (Jerardino, 2001), the residues are interpreted as adhesive remains. To reconstruct the use-life of the stone tools and their residues and verify their nature, the artefacts were analysed with optical microscopy, spectrographic methods, and chemical analysis. This work represents one of the first comprehensive multi-analytical studies of a large sample of tools with potential adhesive remains dated to the LSA. The integration of optical descriptions and molecular data of use-wear and residues will help document the use of adhesives at the site and illuminate on raw material selection, recipe composition, and potential diachronic changes in adhesive technology. By analysing the Steenbokfontein Cave tools, we lay the groundwork for establishing the regional and geographical continuity of adhesive technology during the South African Stone Age.

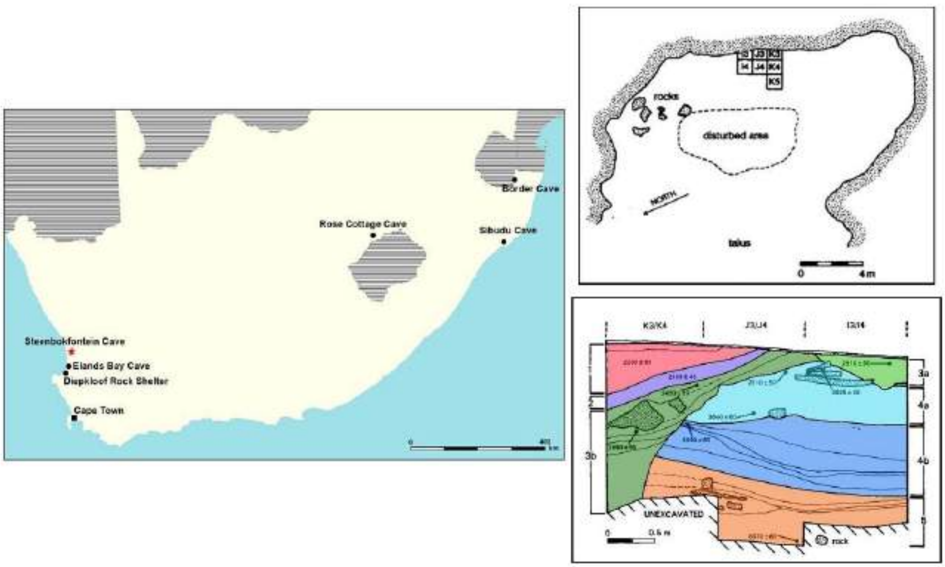

Figure 3.1: Location of Steenbokfontein Cave and other archaeological sites mentioned in the text. Site plan and stratigraphic section of the cave. From Jerardino and Swanepoel 1999, modified.

Materials and methods

Site introduction and materials

Steenbokfontein Cave is located about 200 km north of Cape Town and about 2.5 km east of the nearest shoreline on the west coast of South Africa (Fig. 3.1). Excavations at the site were undertaken between 1992 and 1997, and seven occupation layers were identified thus far. Radiocarbon analysis dates this sequence to between 2005 and 9530 cal BP (Jerardino, 2022). Despite the relatively small volume excavated from this coastal site (6.75 m^3), Steenbokfontein Cave has provided unique and key observations to understand the Holocene cultural sequence of the central west coast (Jerardino et al., 2013). Roughly 11,000 flaked stone artefacts were recovered, of which 368 are formal tools. The lithic technocomplex of Steenbokfontein Cave is characterised as a Wilton and microlithic final LSA assemblage, with scrapers being the most frequently identified retouched pieces (Jerardino, 2013; Lombard et al., 2012). Lithic raw materials are dominated by quartz and quartzite, which are ubiquitous locally, and exotic rocks such as silcrete, hornfels, and cryptocrystalline silica (CCS) are present in lower percentages (Jerardino, 2013). The highest percentages of exotic lithic raw materials are present in layers 4b (2σ: 4580-4155 cal BP, 10.4%) and 5 (2σ: 5480-9030 cal BP, 13.5%). This temporal trend is also reflected in several other sites within a 20 km radius (Jerardino, 2013; Jerardino et al., 2021) suggesting that mobility was increasingly restricted to the coast and its foreland in later occupations during the accumulation of layers 4a to 1 (2σ: 3990-2020 cal BP) (Jerardino et al., 2013). This coastal landscape includes sandstone outcrops and ravines 25 km south or inland where large shrubs and trees grow (Cartwright, 2013).

The Wilton technocomplex is one of several microlithic tool production sequences in the LSA. The microlithic tool production in the LSA started around 40,000 years ago, contrasting with the preceding MSA, during which larger stone artefacts were produced (Lombard et al., 2022; Lombard et al., 2012). Research shows that LSA lithic miniaturization during the Robberg dated to about 18,000-12,000 BP was the result of technological efficiency decisions with high adaptive payoffs, including bipolar bladelet production (Pargeter & Faith, 2020). This is likely to also have been the case for the Wilton technocomplex, but additional research must confirm this. Unfortunately and with few exceptions, the function of Wilton and post-Wilton microliths in southern Africa has received little attention when compared to much older lithic industries (e.g., Lombard, 2020). These few studies show that microliths were used for different purposes such as wood working (Binneman, 1983) or as insets in hunting composite tools (Lombard & Parsons, 2008). While these and many other microliths were hafted, thumbnail-sized and larger artefacts may not have needed this form of fixture and could have been handheld for hide-scraping.

Steenbokfontein Cave yielded two unique adhesive finds: a stone adze embedded in a large adhesive lump and a cigar-shaped resinous object. Both artefacts were found in layer 1, which roughly dates to 2200 cal BP (Jerardino, 2001). Additionally, macroscopic mastic residues or staining were observed on 30 retouched stone tools from all the stratigraphic units, but they were chemically characterised. Of these tools, two are from layer 5, 10 are from layer 4 (4a and 4b), 10 are from layer 3 (3a and 3b), two are from layer 2, and six are from layer 1 (Table SI1). The material is curated at the Department of Archaeology, University of Cape Town (South Africa). We collected morphometric data from all the tools and inspected them with a stereo microscope and a Dino-Lite Edge Digital Microscope (AM7915MZT) to describe the residues.

Of these 30 tools, we selected 13 for in-depth use-wear analysis and molecular identification of residues considering chronostratigraphy, tool morphology, and raw materials (Table 3.1). In-depth analyses were performed at Leiden University and Delft University of Technology (the Netherlands). The selection allows us to document possible diachronic changes in adhesive technology and directly link adhesives to tool technological aspects (e.g., tool type, tool use, and rocks material properties). All the selected artefacts display macro-residue or black staining clearly delimited to an area and always observed on both sides of the tools (Fig. 3.2).

Table 3.1: Overview of the 13 stone tools selected for in-depth analysis. CCS: crypto crystalline rock.

ID	Layer	Square	Tool type	Mastic	Mastic Stained	Raw material	Age cal BP	Analysis
SBF2	5	K3	Scraper	Yes		Silcrete	c. 5240	Traceology, µ-CT, GC-MS
SBF4	4b	I4	Scraper		Yes	Quartz	c. 4390	Traceology, XRD, FTIR
SBF5	4b	I3	Scraper (convex)	Yes		CCS	c. 4390	Traceology, XRD, µ-CT, GC-MS
SBF9	4b	J3	Retouched piece	Yes	Yes	Quartz	c. 4390	Traceology, XRD, FTIR
SBF10	4b	I4	Scraper		Yes	CCS	c. 3810	Traceology, XRD, FTIR

SBF14	3b	K5	Scraper (boat-shaped)	Yes		Quartz	c. 2770	Traceology, XRD, μ-CT, ATR, GC-MS
SBF15	3b	K3	Scraper (boat-shaped)		Yes	Quartz	c. 2770	Traceology, XRD, GC-MS
SBF23	3b	H3/I3	Adze	Yes	Yes	Silcrete	c. 2770	Traceology, XRD, GC-MS
SBF17	3a	I3	Scraper (boat-shaped)	Yes		Quartz	c. 2545	Traceology, XRD, GC-MS
SBF20	3a	I3	Scraper (boat-shaped, broken)	Yes		Quartz	c. 2545	Traceology, XRD, FTIR, GC-MS
SBF21	2	K3	Scraper	Yes	Yes	Quartz	c. 2340	Traceology, XRD, FTIR
SBF24b	1	K4	Scraper (boat-shaped)	Yes	Yes	Quartz	c. 2170	Traceology, XRD, FTIR
SBF27	1	K3	Scraper (boat-shaped)	Yes		CCS	c. 2170	Traceology, μ-CT, GC-MS

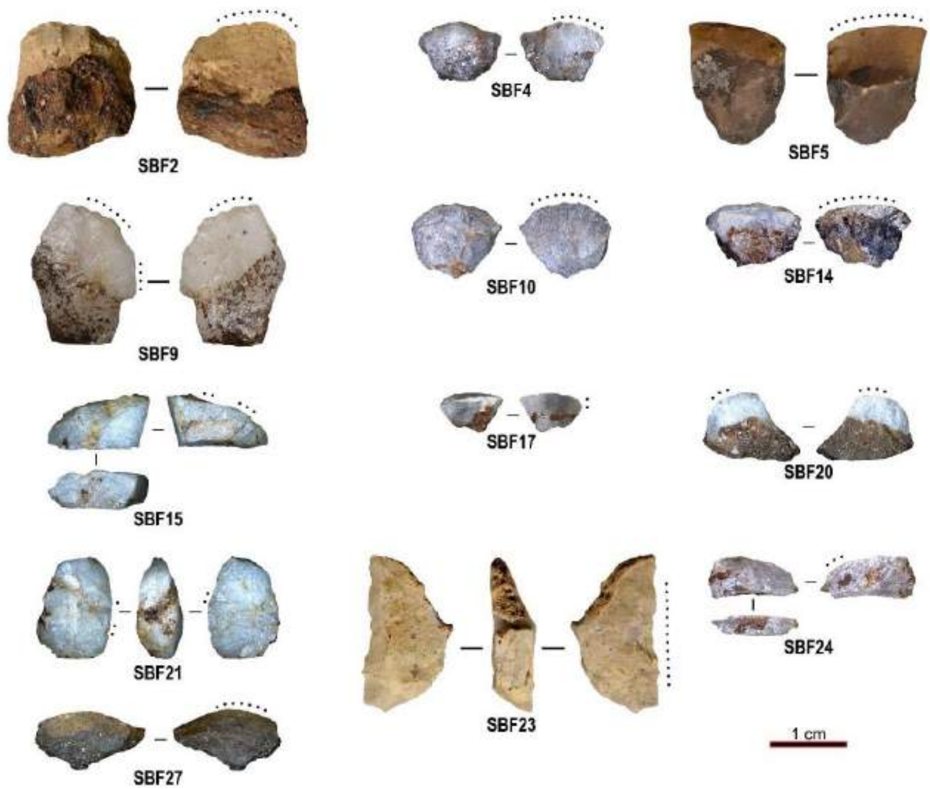

Figure 3.2: Tools with residues selected for in-depth analysis. The dotted line indicates the location of wear-traces.

Methods

As the first step of analysis, we examined all the tools with adhesive residues (N=30) with a stereomicroscope and a Dino-Lite Edge Digital Microscope (AM7915MZT) to describe the morphological features of residues. Thirteen tools were subsequently selected for in-depth non-destructive and destructive analyses (Table 3.1).

Non-destructive analyses consist of high-power optical microscopy for functional analysis, X-ray diffraction (XRD), X-ray micro-computed tomography (μ-CT), Fourier-Transform Infrared microspectroscopy (micro-FTIR) and attenuated total reflectance (ATR-FTIR) (SI, S1). Optical microscopy provides evidence on the use-life of the objects and their residues (Van Gijn, 2010). XRD and μ-CT provide information on the inorganic components of the adhesive mixtures. XRD was performed on 11 tools to verify the presence of additives, such as ochre (e.g., Rosso et al., 2016). We excluded SBF2 and SBF27 because the residues are covered with soil particles and contaminations, which would prohibit confident identification of additives. Four tools displaying millimetres-thick residue were selected for µ-CT to analyse the internal structure of the adhesives and confirm the presence of additives (cf. Niekus et al., 2019). Micro-FTIR and ATR-FTIR were used complementarily to identify organic components in the adhesives (Chen et al., 2022; Helwig et al., 2014). Six tools, which were not sampled for GC-MS, were analysed with micro-FTIR in reflectance mode to gain information on the nature of the residues. Additionally, ATR was performed on a residue sample removed from SBF14 to compare the results of ATR with those obtained in reflectance mode. Excluding ATR, these analyses are non-invasive and do not require destructive sampling. Due to the risk of accidental damage to the artefact, ATR was not used for *in-situ* residue characterisation.

Destructive analysis consists of GC-MS (SI, S1). Despite its destructive nature, GC-MS is the most precise method to characterise unknown organic residues in archaeological samples (Langejans et al., 2022). GC-MS allows the identification of material-specific biomarkers which are used to fingerprint unknown mixtures (Evershed, 2008). Eight tools were analysed for GC-MS following previously published protocols (Regert et al., 2006), including two samples (SBF14 and SBF20) that were also analysed with ATR and micro-FTIR respectively. This will help to verify the level of accuracy of spectroscopy results.

Results

Collection overview: typology, raw materials and morphometrics

Most of the analysed tools (N=24) are typologically classified as scrapers. Two tools are classified as adzes, two as multipurpose retouched tools, and two as utilised flakes. Of the 24 scrapers, 10 are classified as boat-shaped scrapers, eight as convex scrapers, five as generic scraper, and one as a backed scraper. Twenty-three tools are made of quartz, four are made of CCS, and three are made of silcrete (Table SI1).

Eighteen tools are complete, while 12 display at least one fracture at one of the extremities. Of the 12 tools with fractures, three are distal fragments, with only the retouched edge preserved (Table SI2). These three fragmented tools may have been broken during use or resharpening, but the presence of residues also on the proximal fracture surface (Fig. 3.2, SBF24) may indicate that they were hafted and used when already fragmented. Complete quartz tools (N=12) are overall smaller than CCS and silcrete tools (N=6). The average length/width ratio for quartz tools is 0.89 mm, and the average length/width ratio for non-quartz tools is 1.13 mm. We performed non-parametric Mann-Whitney, and the difference in size is not statistically significant (U=28.00, p=0.49).

Overview of archaeological residues

Considering the whole sample of tools (N=30), we identified four groups of residues by optical microscopy (Table 3.2) (Fig. 3.3). Group 1 is the most common and includes residues documented on 13 tools (Fig. 3.3.1). Group 2 (Fig. 3.3.2) includes residues on nine objects; group 3 (Fig. 3.3.3) and 4 (Fig. 3.3.4) include residues on three artefacts each. The differentiation of the studied residues into these groups based on their qualitative and morphological characteristics is not strict, and some residues may display characteristics shared with the other groups. These residues are interpreted as organic adhesives, likely a tar or resin or a combination of both, which could have been sourced from trees and other vegetation growing in ravines and outcrops 25 km south or inland from Steenbokfontein Cave (see Cartwright, 2013).

Table 3.2: Qualitative descriptions of residue groups observed on Steenbokfontein tools. Note that residues in group 3 can be assigned to group 1 or 2 based solely on residue's morphological characteristics. Asterisk (*) indicates that those residues were sampled for chemical analysis.

Residue group	Nr of occurrence	Qualitative description
1	13 (SBF3, SBF4, SBF5*, SBF6b, SBF7, SBF8, SBF9, SBF12, SBF15*, SBF21, SBF24b, SBF25, SBF28)	The colour of the residue ranges from black to brown. The residue is usually smooth and matte with cracks on the surface. The limits are sharp and straight, and the residue mostly displays angular terminations. The residue is opaque both when observed in normal and cross-polarised light. Occasionally, thin, flat, orange, semi-translucent/translucent residues are associated with this group. The residue crumbles into small angular fragments.
2	9 (SBF6a, SBF14*, SBF16,	The colour of the residue ranges from brown to orange with a shiny, greasy appearance. The residue is smooth and matte with a rounded shape. When the residue is very thin, it is flat

	SBF17*, SBF18, SBF19, SBF23*, SBF24a, SBF26)	and angular. Cracks are sometimes present on the surface. The residue is opaque and polished when observed in normal light, while semi-translucent spots are visible in cross-polarised light. Occasionally, dark angular inclusions and ochre grains are visible in the residue in cross-polarised light. The residue crumbles into small angular fragments. Thicker lumps of residue are brown, rounded, cracked, and weathered. They are opaque in normal light with some smooth orange inclusions that are semi-translucent in cross-polarised light.
3	3 (SBF2*, SBF20*, SBF27*)	The residue displays on top contaminations from the soil such as sediment grains, charcoal/charred wood, shell fragments, rootlets, etc. The residues underneath the contaminations can either be assigned to groups 1 (SBF2) or 2 (SBF20, SBF27).
4	3 (SBF10, SBF11, SBF22)	The colour of the residue ranges from reddish to light orange with a granular texture. Edges may be either straight and angular or more gradual. The residue is mostly opaque in normal light and opaque with semi-translucent inclusions or semi-translucent in cross-polarised light.

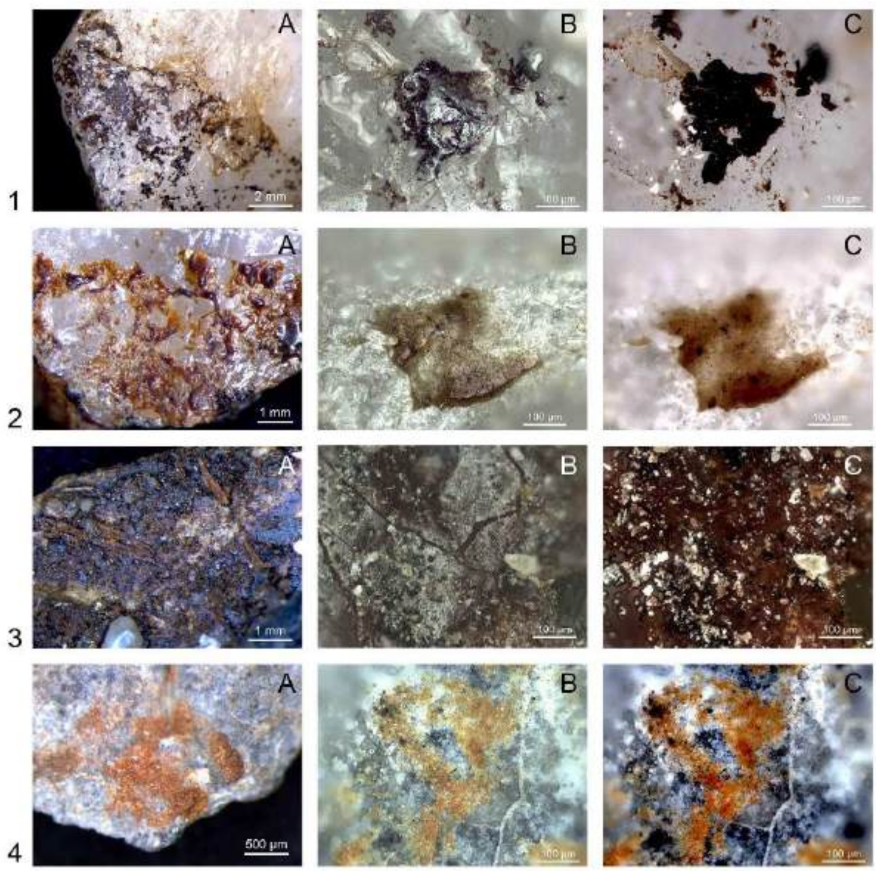

Figure 3.3: Different residue groups identified by optical microscopy. A) General view of the residues; B) View of the residues in bright field illumination (magnification 200x); C) View of the residues in cross-polarised light (magnification 200x).

Group 1 residue was recovered primarily in layer 4b (50%), with lower amounts in layer 1 (22%), layer 3b (14%), layer 2 (7%), and layer 5 (7%). Group 2 residue is mainly present in layer 3a (45%), and it is found in lower amount in layers 1 (22%), 3b (22%), and 4b (11%). This distribution reflects that group 1 is predominant in older layers and that in time, its use was superseded by group 2 residues (Fig. 3.4). This change in adhesive technology may be linked to different production techniques or raw material exploitation.

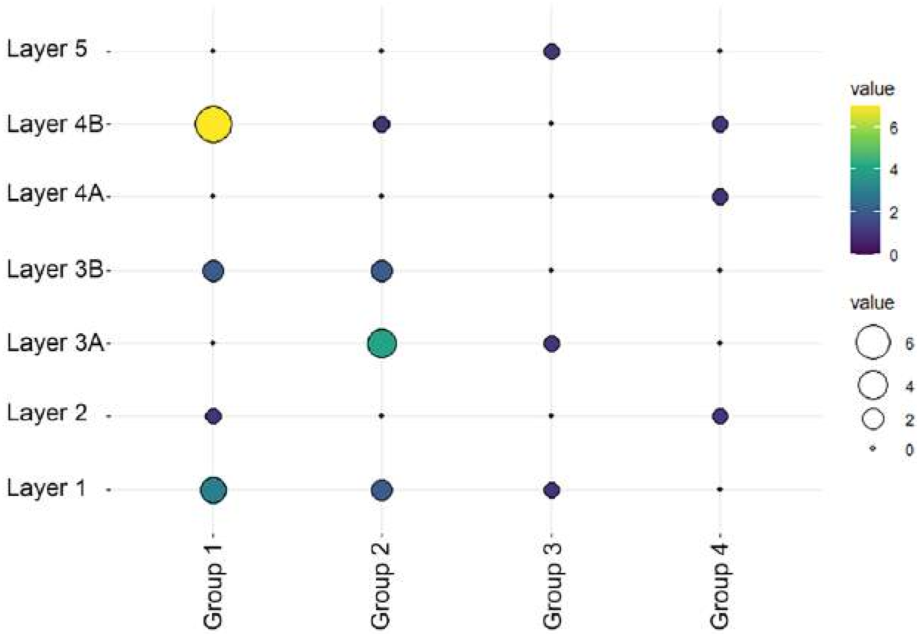

Figure 3.4: Ballon plot showing residue types frequency across the different stratigraphic layers.

The residues on two artefacts, SBF1 and SBF13, were not inserted in any of the groups since their distribution and morphological and surface characteristics seem to suggest a post-depositional origin. Residues on both tools are black, granular, cracked, and randomly distributed on the dorsal and ventral surfaces as well as in the fractures. They lack any other characteristics observed in the previously mentioned residues such as the greasiness, the presence of smooth orange semi-translucent inclusion, or the association with thin, flat, semi-translucent orange stains.

In-depth analyses

Use-wear traces All the tools analysed (N=13) display traces of use (Table 3.3).

Table 3.3: Overview of the use-wear traces observed on the Steenbokfontein tools.

ID	Macrowear	Microwear	Interpretation
SBF2	-Intense resharpening	-Light edge-rounding -Band of greasy, rough, bright polish	-Scraping hide
SBF4		-Isolated edge-removals -Abrasion -Longitudinal striations	-Cutting medium hard material
SBF5	-Resharpening -Edge-rounding	-Edge-rounding - Band of greasy, rough, bright polish with transverse directionality	-Scraping hide
SBF9		-Edge-removals with longitudinal orientation -Abrasion -Longitudinal striations	-Cutting medium hard material
SBF10	-Resharpening -Edge-rounding -Fire alteration	-Edge-rounding -Band of greasy, rough, dull polish with oblique directionality	-Scraping hide
SBF14	-Some edge-damage	-Abrasion in combination with polish and longitudinal striations	-Cutting medium hard abrasive material
SBF15	-Intense resharpening -Proximal fracture	-Some crystals are rounded -Rough and greasy polish on the crystals with longitudinal directionality	-Longitudinal motion -Soft material
SBF17		-Very few spots of domed, smooth polish with longitudinal directionality	-Motion unclear -Maybe soft plant material but minimal evidence -Likely part of a composite tool
SBF20	-Snap lateral fracture	-Light edge-rounding -Isolated edge removals, some with longitudinal orientation -Domed, smooth, 'fluid' polish with diagonal striations	-Longitudinal motion -Possibly plant material
SBF21	-Few edge-damage	-Light edge-rounding -Light abrasion -Isolated spots of domed, smooth, 'fluid' polish	-Motion unclear -Possibly plant material
SBF23	-Overlapping edge-removals with step terminations	-Light edge-rounding on some protruding crystal -Isolated spots of rough and greasy polish	-Motion unclear -Animal contact material -Edge-damage related to a different use

SBF24	-Resharpening -Proximal fracture	-Light edge-rounding -Medium abrasion -Rough polish	-Scraping hide (dry?)
SBF27	-Intense resharpening	-Light edge-rounding -Band of greasy rough dull polish with transverse directionality -Metal traces à aluminium foil	-Scraping hide

At least five tools were used with a transverse motion in scraping activities, and five were with a longitudinal motion (Fig. 3.5). For three tools it was not possible to infer the use-motion. The hardness of the contact materials ranged from soft to medium, with eight tools used on soft material, two on soft/medium material, and three on medium hard material. On eight tools, microwear traces allowed a better understanding of the worked materials. For all the tools used in scraping activities (N=5), hide was identified as the contact material. These tools display light to medium developed edge-rounding and a continuous band of polish along the edge with oblique or transverse directionality (Fig. 3.5A). One tool (SBF24) displays medium developed edge-rounding and medium developed abrasion of the active edge, suggesting contact with a soft abrasive material, such as dry hide or hide with additives (Fig. 3.5B). All scrapers (N=5) display evidence of resharpening of the active edge. Resharpening was identified by the presence of small, overlapping stepped or hinged terminating scars on the dorsal face of the tools and incipient cracks (Fig. 3.5C, D) (cf. Aleo et al., 2021). Furthermore, one scraper (SBF5) shows on the ventral lateral edge at the haft limit large scars that may be related to de-hafting (Fig. 3.5E) (cf. Rots & Williamson, 2004). One tool (SBF23) was used to work unspecified animal material due to the rounding of some crystals and isolated spots of rough and greasy polish (Fig. 3.5F) (cf. Van Gijn, 1990). This tool also displays on the ventral face close, overlapping, step terminating edge-removals with no orientation, likely linked to a different use (Fig. 3.5G). Two quartz tools (SBF20 and SBF21) were likely used on plant. Both tools show isolated edge-removal, some with longitudinal orientation, in combination with a domed, smooth, and almost ‘fluid’ polish (Fig. 3.5H) (cf. Aleo, 2022). Lastly, SBF17 displays minimal traces of use likely related to contact with a soft plant, but the available evidence is not enough to reliably infer the contact material.

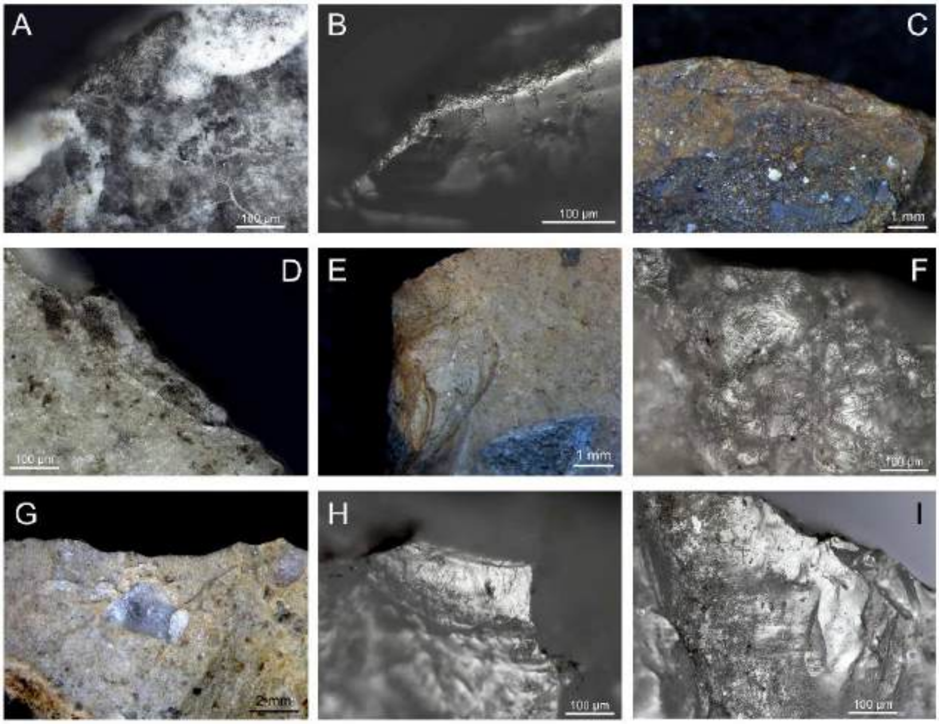

Figure 3.5: Selection of use-wear traces documented on the archaeological tools. A) Edge-rounding and continuous band of polish with diagonal directionality from scraping hide (200x); B) Edge-rounding and abrasion from scraping abrasive soft material (300x); C) Overlapping hinge/step terminating fractures from resharpening (16x); D) Incipient crack from resharpening and band of polish from contact with hide (200x); E) Large edge-removal on the lateral edge of SBF5 possibly from de-hafting (16x); F) Rounding and greasy polish from contact with soft animal materials (200x); G) Overlapping scars with step terminations on the cutting edge of SBF23; H) Domed, smooth and 'fluid' polish from contact with soft plants (200x); I) Abrasion and longitudinal striations from cutting a medium hard material (200x).

XRD results (N=11) allow the identification of additives mixed with the adhesive (Table SI3). Hematite (Fe_2O_3) is identified in nine tools, and magnetite (Fe_3O_4) is identified in one. Magnetite is present in a sample showing severe thermal damage (SBF10). Experimental work has demonstrated that hematite reduces to magnetite when heated (Lanier et al., 2009). Hematite signals can come from the minerals the rocks are made of, the burial environment, or the hafting adhesive. Since none of the XRD patterns collected on the rock substrate display hematite contribution (Fig. 3.6A), we conclude that it was intentionally added to the adhesive mixture. In addition to hematite, the XRD patterns of residue spots of SBF14 display peaks that match with n-paraffin (Fig. 3.6A). Paraffin wax is detected on both faces of the tool but not on the rock substrate, reinforcing the use of a waxy component, such as beeswax, in the adhesive mixture. However, the XRD pattern of paraffine wax only partially overlaps with the beeswax reference. Hence, the detection of beeswax in SBF14 is likely a misinterpretation. The spectrum also shows the detection of amorphous contribution around 17˚, corroborating the organic nature of the residue. Other crystal phases (calcite and halite), which dominate the XRD patterns, are related to the burial environment. Calcite ($CaCO_3$) is a common constituent of archaeological sediment, and halite (NaCl) may relate to soil salinity (El-Ghareb, 2017; Weiner, 2010).

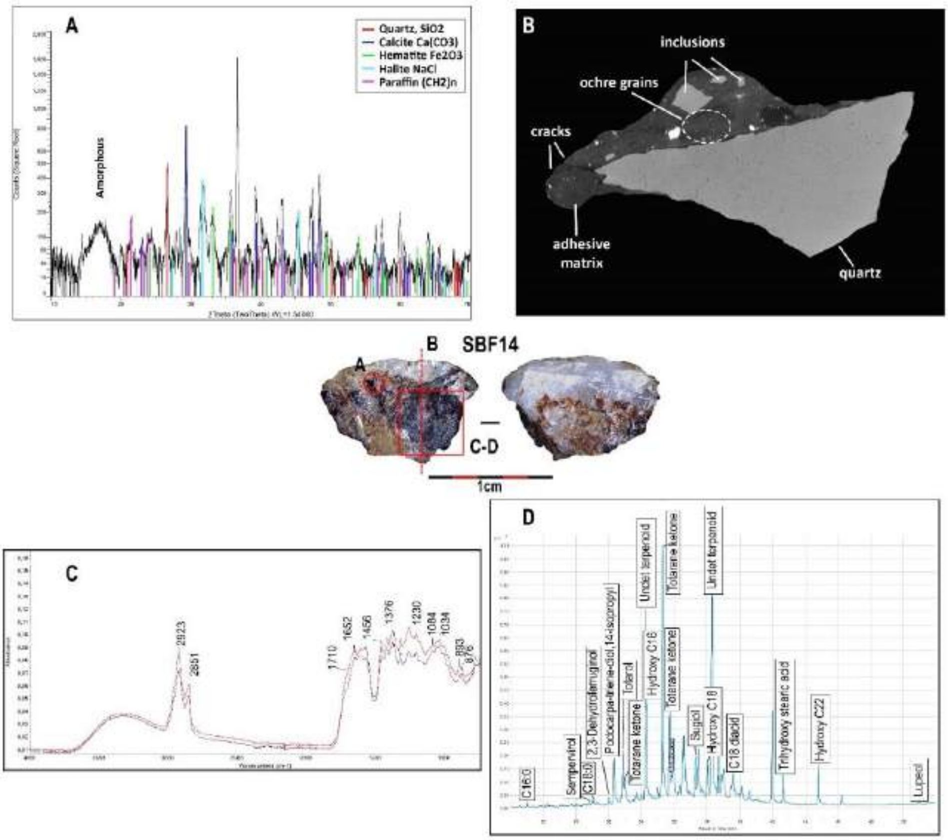

Figure 3.6: Summary of results of in-depth analysis on SBF14 residue. A) XRD pattern showing the presence of hematite Fe_2O_3 and paraffine wax $(CH_2)n$ in the residue spots; B) µ -CT scan of the residue showing iron-oxide grains within the adhesives; C) ATR spectra of SBF14. Only peaks assigned to molecular vibrations of adhesives are labelled; D) Partial chromatogram showing terpenoids relevant for the residue molecular identification.

µ-CT scan On the µ-CT scans, the adhesive appears as an amorphous grey matrix with several inclusions that more or less attenuate the X-ray. Since X-ray attenuation is influenced by the concentration and the atomic number of chemical elements, different features can be identified. Cracks that cross through the adhesive and voids, which are the darkest elements, appear in all the scanned residues. Adhesive residues on SBF2 and SBF27 contain several inclusions with an elongated rounded shape interpreted as sediment grains. Some of the inclusions on SBF2 are of biological origin (shells), and one fragment of a biological organism (carbonised plant) is also visible. These inclusions are distributed mostly on top of the residue, so they are likely post-depositional. Both tools display bubbles/voids at the interface between the adhesive and the stone. The adhesive on SBF14 also contains several inclusions, but they are embedded in the matrix. Big angular inclusions are likely quartz fragments since they present the same X-ray attenuation and texture as the quartz raw material of the tool. Fine-grained rounded contaminants evenly distributed within the matrix and characterised by a high X-ray attenuation coefficient are likely iron oxide rich grains (ochre) mixed into the adhesive when it was in a molten state. The residue is no longer fully adhering to the tools and cracks are visible in several locations (Fig. 3.6B). The residue on SBF4 is very thin. Therefore, no information on its internal structure can be drawn from the scans.

ATR The ATR spectra of SBF14 (Fig. 3.6C, Table SI4) indicate more strongly a gymnosperm extractive, such as a ‘cupressaceous resin’ (see Tappert et al., 2011). The label ‘cupressaceous resin’ includes resins originating from conifers of the *Araucariaceae* or *Podocarpaceae* families. The spectra display a broad band around 3300 cm^{-1} attributed to O-H stretching vibrations (ν(O-H)) and a doublet of peaks at around 2920 and 2850 cm^{-1} that correspond to stretching vibrations of the methylene group (νCH2). Typical bands of resins (cf. Martín Ramos et al., 2018) are detected at 1652 cm^{-1} due to ν(C=C) vibrations, at 1230 cm^{-1} associated with δ(C-H) vibrations and at 1710 cm^{-1} strong carbonyl (C=O) stretch. The shoulder at 1710 cm^{-1} indicates more strongly abietic acid resins than phenol or ketone group resins. However, the region 1500-700 cm^{-1} shows a stronger resemblance to ketone group resins of trees of the angiosperm clade rather than phenol group resins (Martín Ramos et al., 2018).

Micro-FTIR The results of the reflectance micro-FTIR (Tables SI5-10) are not as clear as the ATR results due to the residue’s surface morphology, size, and the significant interference from the underlying siliceous substrate (Monnier et al., 2017; Prinsloo et al., 2014). For all the analysed tools, there is evidence for a tree extractive, but the clade or family of the tree is unclear. All the spectra except for SBF20 and SBF24 display a doublet of peaks at around 2920 and 2850 cm^{-1} corresponding to stretching vibrations of the methylene group (vCH2) as observed in SBF14. Other typical bands of plant extractives identified include a band around 1650 cm^{-1} due to ν(C=C) vibrations and the one around 1500 cm^{-1} due to ν(C=C) vibrations typical of phenolic resins. In addition to those bands, the micro-FTIR spectra of SBF21 shows a peak at 1450 cm^{-1} and a strong peak at 885 cm^{-1}, attributed to the out-of-plane C-H bending motions in terminal methylene groups, which seem indicative of 'cupressaceous resins' (cf. Tappert et al., 2011).

<u>GC-MS</u> Two samples (SBF5 and SBF15) analysed by GC-MS contain no evidence for archaeological lipids, with only trace amounts of palmitic acid preserved. The remaining six contain evidence for adhesives (Table SI11). Identified molecules include saturated fatty acids, hydroxy fatty acids, carboxylic acids, phenolic compounds, diterpenoids, and pentacyclic triterpenoids (Fig. 3.6D).

Saturated fatty acids range from $C_{7:0}$-$C_{22:0}$, with a predominance of long-chain even-numbered molecules. This wide range of even- and odd-numbered molecules is suggestive of the use of animal and plant products (Pollard & Heron, 2008). Hydroxy fatty acids are both short- (C_6 and C_7) and long-chain (C_{16}, C_{18}, and C_{22}). Dihydroxy fatty acids are less common and include C_8 and C_{10}. The only trihydroxy fatty acid identified is C_{18}, but it is present in each sample. Carboxylic and dicarboxylic acids are primarily short-chain, although two long-chain dicarboxylic acids with C_{16} and C_{18} were consistently identified. Both the hydroxy and carboxylic acids are formed from the degradation of plant biopolymers (e.g., Bernards, 2002; Gandini et al., 2006; Kolattukudy, 2001). Most commonly, they are linked to suberin, which is identified in bark (Kolattukudy, 2001), and their presence is used to suggest the formation of a tar (Ribechini et al., 2011). However, these molecules also form from cutin, which acts as a waxy substance covering leaves and fruits (Kolattukudy, 2001), so the adhesive may include material from multiple plant parts. Isovanillic acid was identified in one residue (SBF23), and this polyphenol may also be attributed to the degradation of plant biopolymers (Bernards, 2002; Kolattukudy, 2001).

The most prominent class of lipids identified are diterpenoids. The same set was identified in all six samples: sempervirol, 2,3-dehydroferruginol, 14-

isopropylpodocarpa-8,11,13-triene-7,13-diol, totarol, and sugiol; dehydrototarol was identified in one adhesive (SBF17). In addition, a series of totarane ketones was identified. These diterpenoids are identified in a limited set of plant families: *Cupressaceae* and *Podocarpaceae* (Cox et al., 2007; Otto & Wilde, 2001), which both have species native to South Africa (Palgrave, 2002). *Cupressaceae* includes different species of *Widdringtonia*, and *Podocarpaceae* includes different species of *Afrocarpus* and *Podocarpus*. Resin may be recovered from the bark of *Widdringtonia* trees as well as from the leaves of *Afrocarpus* and *Podocarpus* trees (Page, 1990a, 1990b). Chemically, these are highly similar (Cox et al., 2007), so it is unclear which tree species were exploited for their adhesive properties, and it is possible that both were utilised separately or in tandem. Phenolic and aromatic compounds, which form from the degradation of suberin due to intense heating (Robinson, et al., 1987) as in tar production, are absent in most of our samples. α,ω–Dicarboxylic acids, which also suggest tar making (Ribechini et al., 2011; Villa et al., 2012), are conversely consistently identified. The absence of phenolics and aromatics may relate to preservation biases and not exclusively to the use of resin over tar. Therefore, despite some indications in favour of tar, we cannot rule out the use of resin.

In addition, three adhesives (SBF2, SBF14, and SBF17) contain pentacyclic terpenoids. SBF14 and SBF17 contain lupeol, and SBF2 contains α-lupane and lupa-2,20(29)-diene, which form from the degradation of lupeol. Several plant species native to South Africa contain lupeol (Mavundza et al., 2022; Poumale et al., 2008; Sunita & Abhishek, 2008). Among these, *Euphorbia* is renowned for its latex's adhesive properties (Mwine et al., 2013). However, in these plant species, lupeol is identified alongside other biomolecules, which are absent from the Steenbokfontein samples, deterring a conclusive identification. Nonetheless, it is clear that in these three adhesives, an additional material was combined with the *Widdringtonia* or *Podocarpus* resin/tar as these do not contain any pentacyclic terpenoids.

Discussion

Function and hafting methods

We documented use-wear traces in different stages of development on all the analysed tools (N=13). Despite 11 of the analysed tools being typologically classified as scrapers, only five were actually used in scraping activities and particularly for hide-working. Although Wilton scrapers are often assumed to be hide-working tools, functional studies have shown that they were involved in other craft activities, including working wood and bone (Forssman et al., 2018). The scraping tools we analysed (N=5) were consistently used on hide; however, our analysis considers only a small number of artefacts, and different uses may emerge from the use-wear study of a larger number of scrapers. Five other tools were likely used to work animal and plant materials with longitudinal motions. Even though we did not observe diagnostic wear features, we cannot rule out that some of the tools with evidence for contact with animal material were used as lateral barb projections on hunting weapons (de la Peña et al., 2018; Rots, 2016) and not as hide scrapers. Based on the longitudinal directionality of the micro-traces (abrasion and striations) on these tools, we can however exclude their use as transverse arrowheads. Experimental work has shown that in transverse end-hafted arrowheads, traces are perpendicular to the cutting edge (de la Peña et al., 2018).

All the scraping tools show clear evidence of resharpening of the working edge. Although end-scrapers are normally subjected to several resharpening episodes during their use-life (Aleo et al., 2021; Blades, 2003), at Steenbokfontein this may relate to their use as hafted tools. The manufacturing of hafted tools requires more technological investment; therefore, they are often heavily curated and maintained tools (Rots, 2010).

The distribution of residues on most of the analysed scraping tools suggests that they were not inserted in a groove in the haft and then fixed with adhesive. On the contrary, they were likely inserted in an adhesive lump and side-mounted to the handle of a wooden or bone haft with variable inclinations like the specimens from Boomplaas Cave and Plettenberg Bay (Deacon & Deacon, 1980; see also Porraz & Guillemard, 2019). The cutting tools were likely mounted with adhesive to the haft in a parallel lateral position. This hafting method is confirmed by one quartz microlith (SBF16) (Fig. 3.7). The tool is set in a lump of adhesive with a concave-shaped base. That shape is the result of the adhesive being folded around a wooden shaft, as demonstrated by the presence of wood residues and wood impressions on the inner surface of the adhesive (Fig. 3.7B).

The proximal extremity of the quartz flake does not protrude from the adhesive, confirming that the flake was not in contact or inserted into a socket in the handle.

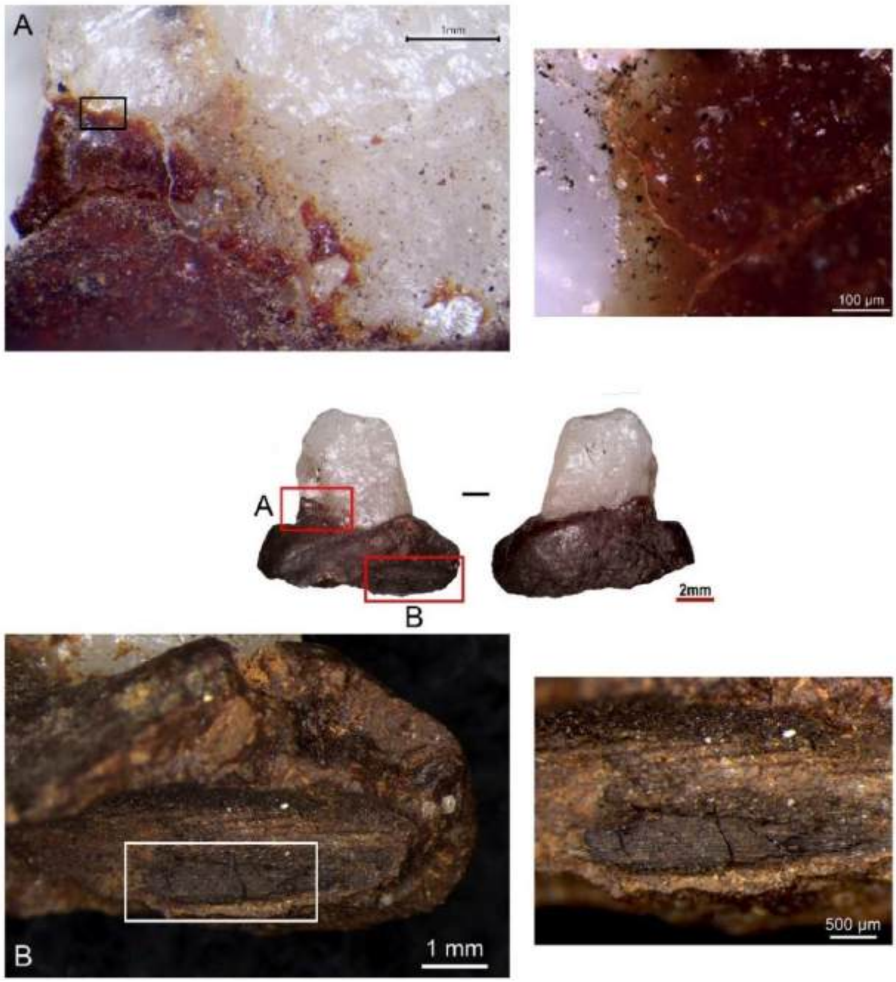

Figure 3.7: Quartz microlith set in a large lump of adhesive (SBF16). Wood impressions and wood residues are visible in the inner surface of the adhesive. A) Overview of the residue (30x) and close-up of the residue in cross-polarised light (200x); B) Wood impression and wood residues on the inner surface of the residue (16x) and close-up of a preserved wood fragment from the shaft (32x).

Two of the three artefacts with the adhesive covered in soil particles and contaminants were subjected to µ-CT scanning. Both are characterised by the presence of bubbles and voids at the interface between the residue and the stone. Bubbles may have formed during the de-hafting process. Based on ethnographic accounts (Sahle, 2019), exhausted or broken hafted tools can be placed near the fire to soften the adhesives and facilitate de-hafting. Small bubbles in the adhesive usually form during this process (Y. Shale, personal communication 2023). Wadley (2010) also reported the formation of air-filled hollows under the adhesive surface when the adhesive is heated too rapidly or placed too close to the fire. The tools were then probably discarded while the adhesive was still malleable, and sediment particles adhered to it.

Evaluating the effectiveness of analytical methods for adhesive residues analysis

We used optical microscopy to make the first interpretation of residues on stone tools. Based on their location, distribution, and appearance, we interpreted all residues as potential adhesive remains except in two cases (SBF1 and SBF13). Visually, we identified different groups based on the residues' morphological features (see Table 3.2). Most residues (N=22) are distributed between groups 1 and 2, which are visually distinguishable and vary in concentration in the archaeological units. Group 1 residues are more abundant in older layers, and their number gradually decreases in younger layers in favour of group 2 residues. Our initial interpretation was that this is linked to a change in adhesive technology over time. Subsequent spectroscopic and chemical analyses on a sample of tools (N=13) confirmed our interpretation as adhesive remains. But the GC-MS results disputed all the other inferences made during residue morphological description based mainly on the colour and surface attributes. Two residue samples from group 1 do not preserve archaeological lipids, but the results of the sample from SBF2, which can be assigned to group 1, show that it is molecularly similar to group 2 residues. Therefore, the distinction of residues in groups based on morphological attributes does not reflect a difference in the organic components of adhesive mixtures or technology. Black opaque residues were likely exposed to different depositional environments in the cave that affected their surface qualities and, in some cases, the preservation of organic molecules. These results demonstrate that residue analysis based solely on morphological attributes can form misleading interpretations.

We analysed the inorganic fraction of the adhesive mixture by XRD. XRD proved to be a useful tool for the identification of crystalline additives in the adhesive, such as hematite. Hematite was identified in all the analysed residues but one. μ-CT also provided evidence supporting the use of hematite as an additive corroborating XRD results. The μ-CT allowed us to virtually section the residue on the tools to evaluate the presence and distribution of mineral particles within the adhesive. Iron-rich particles evenly mixed in the adhesive can be seen in SBF14 images of the sectioned residue, suggesting they were intentionally mixed into the adhesive. Similar conclusions can be drawn for the other tools as well.

We applied IR spectroscopy (micro-FTIR and ATR) and GC-MS to identify the organic fraction of the residues. FTIR microspectroscopy effectively identified the organic nature of residues. All the residue samples analysed by micro-FTIR and ATR (N=7) are of plant origin. The FTIR results suggest a tree resin/tar, but the clade or family of the tree of origin is unclear for most of the samples. The ATR results are clearer than those obtained in reflectance mode (micro-FTIR). The ATR spectra of SBF14 strongly resemble the spectra of various extractives from gymnosperm conifer trees (cf. Tappert et al., 2011), as subsequently confirmed by GC-MS. However, residue identification based on spectroscopy alone is challenging mainly due to limitations posed by degradation, natural or anthropic, of the organic component of the adhesives, absence of extensive reference libraries, and weak/noisy reflectance spectra (Monnier et al., 2017). Nonetheless, IR spectroscopy is a powerful pre-screening method that allows the selection of promising samples for destructive GC-MS and can help narrow down the range of options for identifying unknown organic residues. The precise identification of organic compounds in the adhesive mixture was achieved with GC-MS. For all the residue samples analysed by GC-MS with preserved residues (N=6), the primary ingredient of the adhesive comes from tree extractives (resin or tar) of the *Cupressaceae* and/or *Podocarpaceae* families. Our results do not clearly point towards heated resin or tar production.

Adhesive materials and additives at Steenbokfontein

The combination of GC-MS, XRD, and μ-CT allowed the identification of at least two different compound adhesive recipes at the site: *Widdringtonia* or *Podocarpus* resin/tar mixed with hematite and *Widdringtonia* or *Podocarpus* resin/tar mixed with a different tree extractive containing pentacyclic terpenoids (*Euphorbia* latex?) and hematite (Table 3.4). Moreover, micro-FTIR for SBF9 indicates that this residue is a tree extractive, but no mineral additives were detected in the XRD pattern of the measured residue spots. Therefore, it is likely that resin/tar or a mixture of plant extractives were also used as hafting adhesive without mineral additives.

Table 3.4: Overview of the different adhesive recipes identified at Steenbokfontein Cave considering the tool's raw material and stratigraphic position. CCS: crypto crystalline rock. The dash symbol (-) indicates that the compound was not detected. The slash symbol (/) indicates that the analysis was not performed. SBF5 and SBF15 (in grey) do not contain evidence of organic residues.

ID	Layer	Tool raw material	Organic fraction (ATR, micro-FTIR)	Organic fraction (GC-MS)	Inorganic fraction (XRD)	Use
SBF2	5	Silcrete	/	-*Widdringtonia* or *Podocarpus* sp. resin/tar -Plant exudate containing pentacyclic terpenoids	/	Hide-scraping
SBF4	4b	Quartz	Tree extractive	/	Hematite Fe_2O_3	Cutting medium hard material
SBF5	4b	CCS	/	-	Hematite Fe_2O_3	Hide-scraping
SBF9	4b	Quartz	Tree extractive	/	-	Cutting medium hard material
SBF10	4a	CCS	Tree extractive	/	Magnetite Fe_3O_4	Hide-scraping
SBF14	3b	Quartz	Tree extractive (*Cupressaceae*)	-*Widdringtonia* or *Podocarpus* sp. resin/tar -Plant exudate containing pentacyclic terpenoids	Hematite Fe_2O_3	Cutting medium hard material
SBF15	3b	Quartz	/	-	Hematite Fe_2O_3	Cutting soft material
SBF23	3b	Silcrete	/	*Widdringtonia* or *Podocarpus* sp. resin/tar	Hematite Fe_2O_3	Working soft animal material
SBF17	3a	Quartz	/	-*Widdringtonia* or *Podocarpus* sp. resin/tar -Plant exudate containing pentacyclic terpenoids	Hematite Fe_2O_3	Likely soft plant material

SBF20	3a	Quartz	Tree extractive	*Widdringtonia* or *Podocarpus* sp. resin/tar	Hematite Fe_2O_3	Cutting siliceous plant
SBF21	2	Quartz	Tree extractive (*Cupressaceae*)	/	Hematite Fe_2O_3	Likely siliceous plant
SBF24	1	Quartz	Tree extractive	/	Hematite Fe_2O_3	Hide-scraping (dry?)
SBF27	1	CCS	/	*Widdringtonia* or *Podocarpus* sp. resin/tar	/	Hide-scraping

Powdered hematite and another plant exudate were added as additives to the main tackifier to enhance the adhesive's material properties, such as tackiness, elasticity, and plasticity (Langejans et al., 2022). Ochre is a common ingredient in South African compound adhesives (e.g., Gibson et al., 2004; Lombard, 2007; Wojcieszak & Wadley, 2018), and, as several studies demonstrated, it functions to increase the strength and improve the workability of the adhesive and reduce the curing time and hygroscopicity of the adhesive (Kozowyk et al., 2016; Wadley, 2005; Zipkin et al., 2014). Based on the GC-MS results, another possible additive could have been animal fat. SBF2, SBF14, SBF23, and SBF27 display an odd-numbered fatty acid ($C_{15:0}$), which is typically associated with ruminant animals (Helwig et al., 2014; Regert, 2011). Although animal fat is sometimes reported as an ingredient of adhesives in South Africa (e.g., Charrié-Duhaut et al., 2016; Lombard, 2006), this is unlikely to be the case. $C_{15:0}$ is documented in tools displaying wear traces of contact with animal materials, and it is absent in the ones (SBF20 and SBF27) used to work plants. Therefore, the presence of animal fat should be seen as contamination linked to tool use and not as an ingredient intentionally mixed into the adhesive.

Behavioural aspects and implications linked to adhesive technology

The presence of adhesives in the archaeological record is often seen as a proxy for technological complexity. The manufacturing of a completely new material through the distillation of bark or leaves (tar) or the mixing of several organic and inorganic ingredients (compound adhesives) requires advanced cognitive abilities, considerable technical skills, control of fire, and an understanding of material properties (Niekus et al., 2019; Schmidt et al., 2022; Wadley et al., 2009). Adhesives can have a wide range of uses, and adhesive mixtures can be altered by adding and manipulating ingredients to achieve various desired products suitable for different applications. At Elands Bay Cave, for instance, animal fat was likely added to the adhesive mixture used to seal the perforation of an ostrich eggshell flask, but it is absent on the two other samples of hafting adhesives (Charrié-Duhaut et al., 2016). The deliberate addition of animal fat here likely acted on the properties of the adhesive to accommodate a different type of use. Therefore, characterising adhesive traditions and documenting the variations in adhesive recipes and uses is fundamental for discussing: i) the level of understanding of natural material properties of prehistoric populations, ii) their level of technical expertise, and iii) technological flexibility and technological innovations. This is particularly relevant when considering adhesives dated to the Middle Pleistocene and the role played by this technology in the ongoing debate about the cognitive abilities of Neandertals and early modern humans.

Recipes in relation to tool materials and function

At Steenbokfontein, we identified several multi-component adhesives made of resin/tar and mixed with powdered hematite and, in some cases, another plant exudate. Adhesives were primarily used at this site for mounting stone tools to handles, with the only exception of a cigar-shaped mastic tool with a still unclear function (Jerardino, 2001). It has been argued that the variability in adhesive types may be influenced by the object's raw material or use. At Sibudu for instance, ochre stains are common on segments made of dolerite and hornfels and are notably less frequent on quartz and crystal-quartz segments (Lombard, 2007). According to Lombard (2007), this discrepancy relates to differences in the raw material, such as roughness, grain size, and porosity. Experimental work also showed that glue performance varies according to different substrates (Tydgadt & Rots, 2022); therefore, it is conceivable that the composition of mixtures varied according to the object's raw material. Furthermore, brittle adhesives may have been preferred for certain activities, e.g., shooting, while robust adhesives were preferred for repetitive tasks, e.g., scraping and cutting, suggesting that the selection of adhesives was task-oriented (Wadley et al., 2009). However, in our sample, coniferous ochre-loaded adhesives were used both on quartz and non-quartz tools, and these tools were used for diverse activities, showing no correlations between tasks and adhesive recipes.

Diachronic view on recipes

This consistent adhesive recipe is also observed diachronically at Steenbokfontein Cave despite important changes in subsistence and settlement patterns in the Western Cape since about 3500 BP. Settlements shifted from caves and shelters to large open-air sites along the coast, mobility drastically decreased and was limited to the costal margin and foreland, and the diet became more marine oriented soon after about 3200 BP (Jerardino et al., 2013). The changes in mobility in Steenbokfontein Cave and other local sites are reflected in the frequency of exotic raw materials, which decreases after the deposition of layer 4a (2σ: 3990-3245 cal BP) in favour of local ones (Jerardino, 2013). However, the observed changes in mobility did not affect the procurement of the primary ingredient for adhesive production.

It is also important to observe that while conifer resins/tar is consistently exploited throughout the sequence, the use of additives seems more flexible and less standardised than previously argued. A recent ethnographic work (Fajardo et al., 2024) showed that traditional adhesives exhibit adaptability in materials, productions, and behaviours. Ingredients can be replaced, left out, and mixed based on their availability (Fajardo et al., 2024). The same flexibility in adhesive technology also emerges from the analysis of Steenbokfontein Cave adhesives. Despite the near consistent use of coniferous resin or tar as the main component of their mixtures as well as ochre, prehistoric glue makers mixed and substituted additional ingredients to achieve the final product. This variation shows that adhesives were not material-specific or task-oriented but were likely made of what was available. Another potential explanation for the variability in recipes is that the different adhesive types were produced by different makers or groups who inhabited the cave with slightly different adhesive traditions. This hypothesis could find support from previous interpretations of Steenbokfontein Cave as an aggregation place for different human groups since the mid-Holocene (Jerardino et al., 2013).

The continuity of the use of coniferous resin adhesives in South Africa is documented in different sites from the west to the east coast and dates to at least ~65,000 years ago (Soriano et al., 2015). Charcoal and pollen of *Podocarpus/Afrocarpus* trees are documented in Elands Bay Cave (Cartwright et al., 2016), Diepkloof Rock Shelter (Cartwright, 2013), Sibudu Cave (Zwane

& Bamford, 2021), and Border Cave area (Scott et al., 2023) indicating the availability of these species in the environment. Furthermore, the abundance of *Podocarpus* sp. charcoal in some archaeological layers or sites where adhesives were found has been seen in relation to its role in adhesive production (Cartwright et al., 2016). Despite the availability of other natural sources of adhesives (see Schmidt et al., 2022), most molecular studies of South African Stone Age adhesives indicate the use of an extractive of the *Podocarpus* genus. The preferential use of *Podocarpus* for adhesives by hunter-gatherers groups is seen as a long lasting tradition and adaptation that transcends changes in technology and has been compared to the preferential exploitation of birch tar in western Europe (Charrié-Duhaut et al., 2016, p. 302). In a recent study, Schmidt and colleagues (2022) argued that *Podocarpus* tar was preferentially selected over other substances, including *Widdringtonia* resin, for its superior mechanical properties, similar to birch bark tar (Kozowyk & Poulis, 2019). While this may be the case, it is for now impossible to molecularly distinguish between *Widdringtonia* and *Podocarpus* extractives. In this regard, modern botanical and archaeobotanical evidence may help narrow down the potential sources of adhesives. In the case of Steenbokfontein, *Podocarpus elongatus* is found today within a ~12 km radius of the cave (Lombard, 2023) and is present in the archaeological deposits of nearby Diepkloof and Elands Bay Cave (Cartwright et al., 2016; Cartwright, 2013). Therefore, it was locally available in the Steenbokfontein landscape. On the other hand, *Widdringtonia cedarbergensis* currently grows within a ~40 km radius of the cave (Lombard, 2023). Based on this evidence, it is possible that *Podocarpus* was the source of adhesive at Steenbokfontein although based on the molecular signature of our sample, we cannot rule out *Widdringtonia*. Also, importantly, experiments show significant differential preservation of natural adhesives and particularly between tars and exudates (resins and gums) (Kozowyk et al., 2020), so it is possible that additional materials were used, but their signature is not preserved.

The systematic application of optical microscopy, spectroscopy, and chemical analysis on a relatively large sample of tools with adhesive residues from all the stratigraphic units allowed us to characterise adhesive technology at Steenbokfontein Cave. Adhesive recipes and use transcend changes in technology, raw materials exploitation, and also subsistence on the west coast of South Africa since the mid-Holocene. Throughout the entire sequence, adhesive technology involved the systematic exploitation of conifer resin/tar combined with organic and inorganic additives. However, its use was part of a flexible strategy in which different ingredients were mixed to achieve the final product, independent of the tool raw material and function. Such behaviour is difficult to document at other LSA sites where (chemical) analyses were mostly performed on a single or a few objects among those of interest (e.g., Charrié-Duhaut et al., 2013). For now, this limits the ability to discuss diachronic and regional trends in adhesive production and use and compare between sites. When viewed in light of the wider history of adhesives, applying a multi-disciplinary approach to studying Middle Pleistocene adhesive residues can help capture nuances in adhesive technology. Recent studies on adhesives used by Neanderthals revealed that they used and combined various organic and inorganic sources (Degano et al., 2019; Schmidt et al., 2024b) to haft different tool types, suggesting the flexibility of their adhesive technology in terms of resources exploited and additives. However, most of the studies focusing on adhesives dating to the Middle Pleistocene lack a comprehensive analytical methodology encompassing use-wear analysis, experiments, and molecular characterization of organic and inorganic adhesive ingredients. Hopefully, in the future, the re-examination of old collections and the systematic application of molecular and spectrographic techniques for residue identification will allow a better understanding of adhesive technology in the deep past.

Conclusion

We presented the results of a multidisciplinary study on 30 LSA tools with adhesive residues from Steenbokfontein Cave, South Africa. We combined optical microscopy, μ-CT and XRD, IR spectroscopy, and chemical methods for use-wear and residue molecular identification to reconstruct the use-life of these tools and their residues and explore adhesive use at the site. Use-wear analysis shows that the tools were used as hafted scrapers for hide-working and as elements of composite tools for cutting animal and plant matters, although other functions, such as use as barb-projections, cannot be excluded. Tools were side mounted on the shaft using compound adhesives made of extractives (resin or tar) of *Widdringtonia* or *Podocarpaceae* species and powdered hematite or by adding a third unidentified plant ingredient, possibly *Euphorbia* latex. There is also an indication of a third recipe that did not include mineral additives.

Throughout the whole sequence, conifer resin/tar is exploited as the main adhesive material, while organic and inorganic additives were added or left out without any observable correlation to the tool's raw material and function. Moreover, we did not observe a chronological trend in the use of the different mixtures. From our results emerge that adhesive technology at Steenbokfontein was flexible in terms of additives and ingredients.

The application of methods for molecular identification of Steenbokfontein Cave residues, in combination with microscopic observation and morphological description, led to a better reconstruction of artefacts' use-life and increased our knowledge and understanding of adhesive technology during the Later Stone Age. Our detailed analysis enables us to discuss adhesive technology in relation to tool's function and highlight the absence of diachronic changes in the production and use of compound adhesives at the site. Nonetheless, we documented a flexible use of adhesive ingredients that challenged previous interpretations of glue recipes designed according to the tool's material properties and use. The systematic application of this approach to other South African archaeological assemblages will allow us to better understand past adhesive technologies and pinpoint the continuity or breaks of traditions in the manufacturing and use of adhesives. Similarly, applying this approach to Pleistocene adhesives ought to generate new insights into different adhesive traditions and allow us to evaluate possible technological flexibility and variability in the deep past and discuss it in light of the technological and cognitive abilities of different human populations.

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Chapter 4

European Palaeolithic adhesives: a case study from Spain

"Persistent use of ochre and plant extracts in Palaeolithic adhesive technology at Morin Cave, Spain"

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Abstract

Adhesives have become a crucial subject in the discussion surrounding the cognitive and technological capabilities of Neanderthals and modern humans. Evidence of adhesives identified so far through spectrometry and/or chemical analyses showed that Neanderthals primarily used birch bark tar adhesives and, to a lesser extent, bitumen and conifer resin. Conversely, modern humans appear to have exploited a slightly wider range of raw materials, including plant resins and gums, tars, beeswax, and bitumen, occasionally mixed with inorganic additives. To expand our understanding of prehistoric adhesive technology and compare Neanderthal and modern human adhesives, we conducted a comprehensive study of the residues found on the entire stone tool collection of Morín Cave in northern Spain. This involved a multi-analytical approach for residue analysis, utilizing optical and scanning microscopy, spectrometry, and chemical methods. We were able to identify adhesive residues on five tools, chrono-typologically attributed to the Châtelperronian (N=2), Protoaurignacian (N=1), Early Aurignacian (N=1), and Gravettian (N=1). Plant resins or tars, including the extractive of a species of the *Cupressaceae* family, were used as hafting material. Additionally, the residue signal on one tool is indicative of animal or plant wax or bitumen. In all cases but one, crushed ochre was added to the mixture, highlighting the continuity of this practice from the Châtelperronian to the Gravettian. The Raman spectra of three residues assigned to the early stages of the Upper Palaeolithic display strong similarities, suggesting the homogeneity of adhesive traditions between the Châtelperronian and the Aurignacian. This homogeneity may be the result of environmental constraints or interactions between diverse human groups who possibly coexisted in northern Iberia for about 1,000 years between the end of the Middle and the beginning of the Upper Palaeolithic.

Introduction

In the last decades, prehistoric adhesives have attracted increasing scientific attention. Due to the antiquity of this technology, the oldest adhesive dates to ~190 ka BP (Mazza et al., 2006), and the set of cognitive and technical skills associated with adhesives production and use (Wadley et al., 2009; Wragg Sykes, 2015), adhesive technology rapidly became a key component of the debate on Neandertals and anatomically modern humans (AMH) cognitive and technological complexity (Fajardo et al., 2023; Kozowyk et al., 2023b; Roebroeks & Soressi, 2016; Schmidt et al., 2023). In western and central Europe, Neanderthal adhesives that have been chemically and spectrometrically identified (Table 4.1) consist almost exclusively of birch bark tar (Koller et al., 2001; Mazza et al., 2006; Niekus et al., 2019), except adhesives from Fosellone and Sant'Agostino Caves (Italy) where conifer resin and occasionally a mixture of resin and beeswax were used (Degano et al., 2019). In eastern Europe and the Levant, natural bitumen was exploited by Neandertals as hafting adhesives as identified at Mezmaiskaya Cave and Saradj-Chuko Grotto (Russia), Gura Cheii-Râşnov Cave (Romania), Umm el Tlel and Hummal (Syria) (Boëda et al., 2008b; Cârciumaru et al., 2012; Doronicheva et al., 2022; Hauck et al., 2013) A goethite-loaded bitumen adhesive was also recently identified at Le Moustier (France) (Schmidt et al., 2024b).

Table 4.1: Chemically and spectrometrically identified adhesives from European Pleistocene sites with Neanderthals and AMH occupations.

Site	Age	Adhesive	Additives	Tool type	Methods	Reference
Campitello Quarry (IT)	*H. Neanderthalensis* >190 ka	Birch bark tar	-	2 flakes	SEM-EDX, FTIR, GC-MS	(Mazza et al., 2006)
Saradj-Chuko Grotto (RU)	*H. Neanderthalensis* 92-41 ka	Bitumen	-	1 side-scraper 1 point 2 convergent scrapers	SEM-EDX, FTIR, Raman	(Doronicheva et al., 2022)
Hummal (SY)	*H. Neanderthalensis* 80-50 ka	Bitumen	-	1 Levallois points 1 Mousterian point 1 Levallois flake	SEM-EDX, FTIR, confocal Raman, GC-MS	(Hauck et al., 2013; Monnier et al., 2013)

Umm el Tlel (SY)	*H. Neanderthalensis* ~71 ka	Bitumen	-	11 Levallois artefacts	GC-MS	(Boëda et al., 2008a; Boëda et al., 2008b)
Mezmaiskaya Cave (RU)	*H. Neanderthalensis* ~70-40 ka	Bitumen	-	1 convergent scraper	SEM-EDX, FTIR, Raman	(Doronicheva et al., 2022)
Le Moustier (FR)	*H. Neanderthalensis* (?) 56-40 ka	Bitumen	Goethite	1 end-notched flake 1 retouched Levallois blade	SEM-EDX, ATR-FTIR, Raman	(Schmidt et al., 2024b)
Fosellone Cave (IT)	*H. Neanderthalensis* 55-40 ka	Conifer resin	Beeswax	3 scrapers 1 flake	GC-MS	(Degano et al., 2019)
Zandmotor beach (NL)	*H. Neanderthalensis* ~50 ka	Birch bark tar	-	1 flake	THM-Py-GC-MS	(Niekus et al., 2019)
Königsaue (DE)	*H. Neanderthalensis* >43 ka and >48 ka	Birch bark tar	-	Free lumps	GC-MS	(Koller et al., 2001)
Sant'Agostino Cave (IT)	*H. Neanderthalensis* ~43 ka	Conifer resin	-	5 scrapers 1 Levallois flake	GC-MS	(Degano et al., 2019)
Gura Cheii-Râşnov Cave (RO)	*H. Neanderthalensis* Undefined Mousterian	Bitumen	-	1 retouched flake	FTIR, EDXRF, GC-MS, ICP-AES	(Cârciumaru et al., 2012)
Grotta del Cavallo (IT)	*H. Sapiens* ~45-39 ka	Plant/tree gum	Beeswax Ochre	6 backed points	SEM-EDX, FTIR	(Sano et al., 2019)
Morín Cave (ES)	*H. Sapiens* >36.5 ka and ~24 ka	Natural resin	Ochre Possibly burnt bone	1 backed blade 1 partially backed blade 1 burin 1 truncation	SEM-EDX, FTIR, Raman	(Bradtmöller et al., 2016)
Hinxton (UK)	*H. Sapiens* ~15-12 ka cal BP	Conifer tar, maybe resin	-	1 backed blade	THM-Py-GC-MS	(Langejans et al., 2023)
Bergkamen (DE)	*H. Sapiens* ~13 ka	Beeswax	-	1 barbed point	FTIR, Py-GC-MS	(Baales et al., 2017)
Maasvlakte 2 (NL)	*H. Sapiens* ~13 ka cal BP	Birch bark tar	-	1 barbed point	GC-MS	(Aleo et al., 2023)

Gura Cheii-Râşnov Cave (RO)	*H. Sapiens* Undefined Upper Palaeolithic	Bitumen	-	1 backed blade	FTIR, EDXRF, GC-MS, ICP-AES	(Cârciumaru et al., 2012)

From the few Upper Palaeolithic adhesives identified through chemical and spectroscopic analyses (Table 4.1) emerges that AMH exploited a slightly wider range of materials than Neanderthals, including birch and conifer tar, resin, beeswax, bitumen, and iron oxides and possibly burnt bone as additives (Aleo et al., 2023; Baales et al., 2017; Bradtmöller et al., 2016; Cârciumaru et al., 2012; Sano et al., 2019). Ochre has been documented in a few Upper Palaeolithic adhesive mixtures (Bradtmöller et al., 2016; Sano et al., 2019), and its use is well-known in Middle and Later Stone Age African adhesive technology (e.g., Gibson et al., 2004; Lombard, 2006; Rots et al., 2011). Conversely, despite evidence for the use of different iron oxides by Neandertals (e.g., Dayet et al., 2014; Dayet et al., 2019; Roebroeks et al., 2012), direct evidence for the use of ochre as an adhesive component has been found once, but its attribution to Neanderthals is uncertain (Schmidt et al., 2024b).

The data highlights differences in adhesive technology between Neandertals and AMH, these appear mainly related to the exploitation of different organic and mineral resources for adhesives production. These differences may be related to diverse natural resources available locally or represent cultural and technological traditions. Nothing can be said about differences in production methods during the Palaeolithic since it is yet unknown how adhesives were produced although some methods have been proposed (Kozowyk et al., 2017; Schmidt et al., 2019; Schmidt et al., 2023). To further explore Neanderthal and AMH adhesive technologies and document differences, similarities, and/or continuity in adhesive traditions in the deep past, we analysed the lithic assemblage of Morín Cave. Our research aim is to determine if both Neanderthals and AMH produced adhesives at the site; the type and function of stone tools they hafted; if they made use of different or similar materials; and to assess to what extent their adhesive traditions differ.

Morín Cave is located in northern Iberia, a crucial area to study the Middle to Upper Palaeolithic transition (Fig. 4.1). This area preserves numerous archaeological sites with long stratigraphic sequences including Mousterian, Châtelperronian, Aurignacian, and Gravettian layers such as El Castillo, La Viña, and Morín, which have allowed archaeologists to characterise Neanderthal and early AMH technology, behaviour, and subsistence (e.g., Bernaldo de Quirós et al., 2015; Bradtmöller, 2015; d'Errico et al., 2016; Rasilla et al., 2020; Yravedra Sainz de los Terreros, 2013). Moreover, many key sites of this region have been recently reviewed and redated providing a chronological and environmental framework in which the results of the analysis of Morín Cave can be discussed and integrated (Fernández-García et al., 2023; Jones et al., 2019; Marín-Arroyo et al., 2018). We studied Morín stone tools with a holistic approach integrating microscopic examination of use-wear and residues and chemical and spectrometric techniques for residue characterization. By studying material from Morín, we document Neanderthal and AMHs' use of raw materials and additives for adhesive production. We discuss these in light of raw material availability and technological choices. The results will broaden our knowledge of adhesive technology and enhance the discussion on Neanderthal and AMH technology and behavioural complexity.

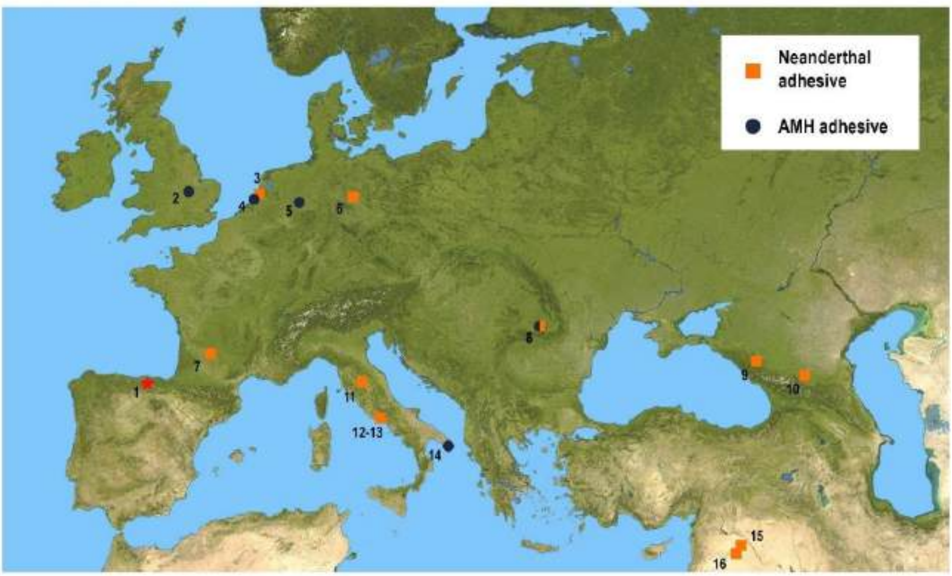

Figure 4.1: Location of Morín Cave (Spain) and other sites cited throughout the paper. 1) Morín Cave (Spain); 2) Hinxton (United Kingdom); 3) Zandmotor (Netherlands); 4) Maasvlakte 2 (Netherlands); 5) Bergkamen (Germany); 6) Königsaue (Germany); 7) Le Moustier (France); 8) Gura Cheii-Râşnov Cave (Romania); 9) Mezmaiskaya Cave (Russia); 10) Saradj-Chuko Grotto (Russia); 11) Campitello Quarry (Italy); 12) Fosellone Cave (Italy); 13) Sant'Agostino Cave (Italy); 14) Grotta del Cavallo (Italy); 15) Hummal (Syria); 16) Umm el Tlel (Syria). Base map via Pixabay.

Materials and Methods

The site and studied assemblage

Morín Cave is located in Villanueva de Villaescusa (Cantabria), 6 kilometres away off the modern coastline. The cave was discovered in the early 1900s, and several excavation campaigns have since taken place (Carballo, 1923; Echegaray et al., 1971; Obermaier, 1916). The main archaeological exploration was between 1966-1969, when a long stratigraphy consisting of 22 layers was exposed. The sequence includes one Azilian (layer 1), one Magdalenian (layer 2), one Solutrean (layer 3), two Gravettian (layers 4 and 5b), one evolved and two early Aurignacian (layers 5a, 6, and 7), two Protoaurignacian (layers 8 and 9), one Châtelperronian (layer 10), and eight Mousterian (layers 11-17 and 22) layers (Maíllo-Fernández et al., 2014). The chrono-cultural sequence is mostly based on the lithic assemblage, as several dating attempts provided unsatisfactory results (Maíllo-Fernández et al., 2014; Marín-Arroyo et al., 2018).

In 2016, Bradtmöller and colleagues reported the presence of potential adhesive remains on three Gravettian and one Aurignacian tools (Bradtmöller et al., 2016). The residues were analysed using scanning electron microscopy-energy-dispersive X-ray spectrometry (SEM-EDX), Fourier transform infrared spectroscopy (FTIR), and Raman spectroscopy and interpreted as adhesives made of natural resin mixed with ochre and possibly burnt bones (Bradtmöller et al., 2016). With the aim of chemically characterising these residues and identifying more adhesives, we inspected the entire lithic collection of Morín Cave (N=23,796) covering the entire sequence of the cave.

The material is stored and curated at the Museo de Prehistoria y Arqueología de Cantabria (MUPAC), Santander (ES). All the lithic tools with at least one usable edge were macroscopically analysed with a stereo microscope to detect potential adhesive residues. We excluded tools with unknown stratigraphic position or from mixed layers, blocks, cores, and Mousterian material smaller than 1 cm. After this first examination, we selected 62 tools with ochre stains or black adhering residues for a more detailed investigation using a metallographic microscope. Then, we selected 31 tools with potential adhesives based on the location, distribution, appearance, and surface quality of the residues. Three of the four tools with adhesive remains identified in 2016 were reanalysed for use-wear and GC-MS (MOR1, MOR5, and MOR6). The residue on the selected tools can be divided into two groups: residue spots <1 mm, barely visible without a stereomicroscope and thus too small to be sampled for chemical analysis (N=12), and those larger and thicker than 1mm, large enough to be sampled with a scalpel blade or cotton swabs (N=19). The former, excluding MOR6 and MOR59, were analysed with SEM-EDX and micro-Raman to confirm their organic nature and gain insights into their molecular structure; the latter were analysed with GC-MS for their molecular characterization (Table 4.2). We excluded the other tools because the residues were randomly distributed on the surfaces or were identified as mineral deposits and manganese stains. Black manganese deposits appear as diffuse spots or dendritic patterns (Randolph-Quinney et al., 2016). To verify the accuracy of our interpretation of mineral deposits based on optical microscopy, we selected four tools with manganese/post-depositional stains for GC-MS analysis. Therefore, the final number of analysed tools is 35 (Table 4.2).

Table 4.2: Overview of the tools with potential adhesive remains and the analyses performed. Tools with asterisk (*) display mineral/manganese deposits.

ID	Layer	Cultural affiliation	Tool type	Residue location	Analyses
MOR1	5a	Gravettian	Retouched blade (proximal-mesial fragment)	Dorsal and ventral face	Microscopy, GC-MS
MOR2	9	Protoaurignacian	Endscraper	Dorsal and ventral	Microscopy, SEM-EDX, micro-Raman
MOR4	7	Early Aurignacian	Dufour bladelet (proximal fragment)	Dorsal, ventral, back	Microscopy, SEM-EDX, micro-Raman
MOR5	5a	Gravettian	Truncation	Dorsal, ventral, back	Microscopy, GC-MS
MOR6	4	Gravettian	Retouched blade (proximal fragment)	Dordal and ventral	Microscopy. Most of the residue removed in 2016
MOR9	10	Châtelperronian	Châtelperronian point	Dorsal, ventral, back	Microscopy, SEM-EDX, micro-Raman
MOR11	10	Châtelperronian	Châtelperronian point	Dorsal, ventral, back	Microscopy, ATR-FTIR, GC-MS
MOR12	5a	Gravettian	Truncation	Back and dorsal	Microscopy, SEM-EDX

MOR13	6	Early Aurignacian	Backed blade (proximal fragment)	Dorsal, ventral, back	Microscopy, SEM-EDX, micro-Raman
MOR14	5a	Gravettian	Backed bladelet	Dorsal, ventral, back	Microscopy, GC-MS
MOR17	9	Protoaurignacian	Endscraper	Dorsal and ventral	Microscopy, SEM-EDX
MOR22	Trench 9	Upper Palaeolithic	Blade (proximal-mesial fragment)	Ventral	Microscopy, SEM-EDX, micro-Raman
MOR24	10	Châtelperronian	Blade (proximal fragment)	Dorsal and ventral	Microscopy, GC-MS
MOR25	10	Châtelperronian	Bladelet (natural back)	Dorsal, ventral, back	Microscopy, GC-MS
MOR28	10	Châtelperronian	Blade (mesial fragment)	Dorsal and ventral	Microscopy, GC-MS
MOR32	7	Early Aurignacian	Endscraper	Dorsal and ventral	Microscopy, GC-MS
MOR35	2	Magdalenian	Backed blade (mesial fragment)	Dorsal, ventral, back	Microscopy, SEM-EDX
MOR40	8a	Protoaurignacian	Blade (mesial fragment)	Dorsal and ventral	Microscopy, ATR-FTIR, GC-MS
MOR41	10?	Châtelperronian	Flake (natural back)	Dorsal, ventral, back	Microscopy, GC-MS
MOR42	9	Protoaurignacian	Retouched flake	Dorsal	Microscopy, SEM-EDX
MOR43	10	Châtelperronian	Blade (mesial fragment)	Dorsal and ventral	Microscopy, ATR-FTIR, GC-MS
MOR45	7	Early Aurignacian	Flake (natural back)	Dorsal and ventral proximal	Microscopy, GC-MS
MOR47	8	Protoaurignacian	Endscraper	Dorsal and ventral	Microscopy, ATR-FTIR, GC-MS
MOR49	8	Protoaurignacian	Bladelet with convergent edges	Dorsal	Microscopy, , GC-MS
MOR50	8	Protoaurignacian	Bladelet (mesial-distal fragment)	Dorsal and ventral	Microscopy, SEM-EDX

MOR57	13	Mousterian	Flake	Ventral left edge	Microscopy, GC-MS
MOR58	13	Mousterian	Laminar flake (distal-mesial fragment)	Ventral, dorsal right edge	Microscopy, GC-MS
MOR59	16	Mousterian	Levallois point	Ventral	Microscopy
MOR60	16	Mousterian	Discoid point	Dorsal and ventral	Microscopy, GC-MS
MOR61	Aurignacian	Aurignacian	Retouched flake	Dorsal	Microscopy, GC-MS
MOR62	1	Azilian	Bladelet (mesial fragment)	Dorsal	Microscopy, GC-MS
MOR26*	10	Châtelperronian	Blade (fragment)	Dorsal and ventral	Microscopy, GC-MS
MOR38*	8a	Protoaurignacian	Flake	Dorsal	Microscopy, ATR-FTIR, GC-MS
MOR44*	10	Châtelperronian	Flake	Dorsal (mostly) and ventral	Microscopy, ATR-FTIR, GC-MS
MOR46*	8	Protoaurignacian	Blade (fragment)	Dorsal and ventral	Microscopy, ATR-FTIR, GC-MS

The studied assemblage consists mostly of complete and fragmented blades and bladelets, some retouched, and points (N=24) (Table 4.2). These tool types are often assumed to have been used as projectiles or elements of composite tools and therefore hafted. Thus, the staining on these tools can potentially represent adhesive remains used to attach them to the haft. However, the sample also features the so-called 'domestic tool types' (N=11), such as endscrapers and retouched flakes (Table 4.2), for which hafting it is not an absolute precondition and may be therefore overlooked.

Methods of analysis

The tools and their residues were analysed with a multi-analytical approach including optical and scanning electron microscopy, spectrometry (ATR-FTIR and micro-Raman), and chemical analysis (GC-MS).

Optical microscopy

Use-wear traces and residues were studied through optical microscopy following established protocols (Coppe & Rots, 2017; Fullagar, 2006; Langejans & Lombard, 2015; Van Gijn, 2010). We employed a Leica M80 stereomicroscope (magnification 7.5x-60x) for documenting the presence of residues on the surfaces of tools and mapping their distribution and for macrowear analysis (low-power approach, Semenov, 1964; Tringham et al., 1974). A Leica DM2500 M metallographic microscope (magnification 50x-500x) was employed for residues morphological description and microwear analysis (high-power approach, Keeley, 1980). Pictures of wear traces and residues were taken using a Leica DFC425 C digital camera.

Microscopic residues (<1 mm)

Ten tools displaying microscopic residues were analysed with scanning electron microscopy-energy dispersive X-ray spectrometry (SEM-EDX) to provide elemental information allowing the verification of the organic nature of the residues (Table 4.2). A Variable Pressure Scanning Electron Microscope (VPSEM) ZEISS EVO MA15 with an integrated EDX system was used for the analysis. We measured at least two residue spots for each archaeological tool, and for tools with residues displaying a strong C peak, a measurement of the rock substrate was also collected to cross-check the results. Based on the results of SEM-EDX, four tools bearing organic residues and one with mineral deposits were further analysed with micro-Raman (Table 4.2). We carried out *in situ* Raman analysis using a BWTEK Raman device, composed of a BWTEK BRM-OEM-785 diode laser (785 nm and 532 nm), a BWTEK BAC100-785E Raman head, and a BWTEK Prime T BTC661E-785CUST spectrometer with a Hamamatsu CCD (S10141-1107S, 2048 pixels) detector. The equipment covers a spectral range in Raman displacement of 80–3600 cm^{-1}, with a spectral resolution of 4 cm^{-1} measured as FWHM. Spectra were acquired using commercial software provided by BWTEK. The maximum laser power was 10 mW on the sample surface to prevent thermal degradation of the materials, which is normally evidenced by a drastic increase in the Raman spectrum background. The mean integration time ranged from 2 to 60 s, and 5 accumulations were performed in each run. The Raman head is equipped with a CCD camera for imaging and an accessory to incorporate 20x and 50x microscope objectives that allow us to choose the focus point of the excitation laser on the sample. A backscattering geometry was adopted to detect the optical signal. We compared the results with reference Raman spectra of experimental birch bark tar and pine resin in a pristine and degraded state, and previously published data (Kozowyk et al., 2020).

Macroscopic residues (>1 mm)

Twenty-three tools displaying macroscopic residues, including 19 with potential adhesive remains and four with mineral deposits identified through optical microscopy, were sampled for molecular analysis (Table 4.2). The latter were included to validate our interpretation of residue as inorganic based on the morphology. Residue samples were removed from each object with a sterile scalpel blade (N=15), with a cleaned cotton swab dipped in methanol (N=8), or by submerging the tool in the same solvent and sonicating for 20 minutes at 30˚C (N=1). The residue on MOR38 was sampled using both a scalpel blade and a cotton swab to compare results of the two extraction methods. From these, we randomly selected seven residue samples for attenuated total reflectance (ATR) analysis (Table 4.2) to verify the results. For the ATR analysis, we used a Perkin Elmer Spectrum 100 FT-IR Spectrometer with a diamond crystal. The software for equipment control and data evaluation was the Spectrum IR. The measurements were performed with a resolution of 4 cm-1, 16 accumulations per measurement, and a wavelength range of 4000-600 cm-1. All samples were then analysed by gas chromatography-mass spectrometry (GC-MS). Lipids were solvent extracted and analysed following previously published methods (Regert et al., 2006) and a modified version of the protocol presented in Birkemeyer et al. (2016) for samples extracted with cotton swabs. The analyses were performed on an Agilent 7890B GC with a split/splitless inlet, coupled with an Agilent 5977B EI MSD, an FID, and a splitter with corresponding EPC pressure control. The GC was fitted with a nonpolar Agilent J&W DB5 MS column (30 m × 0.25 mm i.d.; 0.25 μm film thickness). The chromatograms and resulting mass spectra were interpreted using the National Institute of Standards and Technology (NIST) library (Version 2.2).

Results

Post-depositional traces and modern contamination

Old and modern alterations are visible on all tools. The former consists of patination and surface modifications (Fig. 4.2a) due to taphonomic processes and the depositional environment. Modern day contaminations mostly derive from handling, storing and curating the material and consists of modern fractures and edge-damage (Fig. 4.2b, c), ink and varnish from labelling (Fig. 4.2d, e, f), modern glue staining (Fig. 4.2g), and pencil and pen marks (Fig. 4.2h, i). On almost all tools, and especially the retouched ones, we observed evidence of pencil. On formal tools, the pencil was used to draw the retouch negatives, hindering the observation of potential wear traces and contaminating the residue. Modern day pencil lead is a mixture of graphite, clay, and additives like fats (David et al., 2017) and this must be considered when interpreting the results of residue analysis. Similarly, transparent varnish, used to coat the labelling on the tools, hindered the observation of use-wear and residues on some tools, especially small ones such as bladelets and tool fragments.

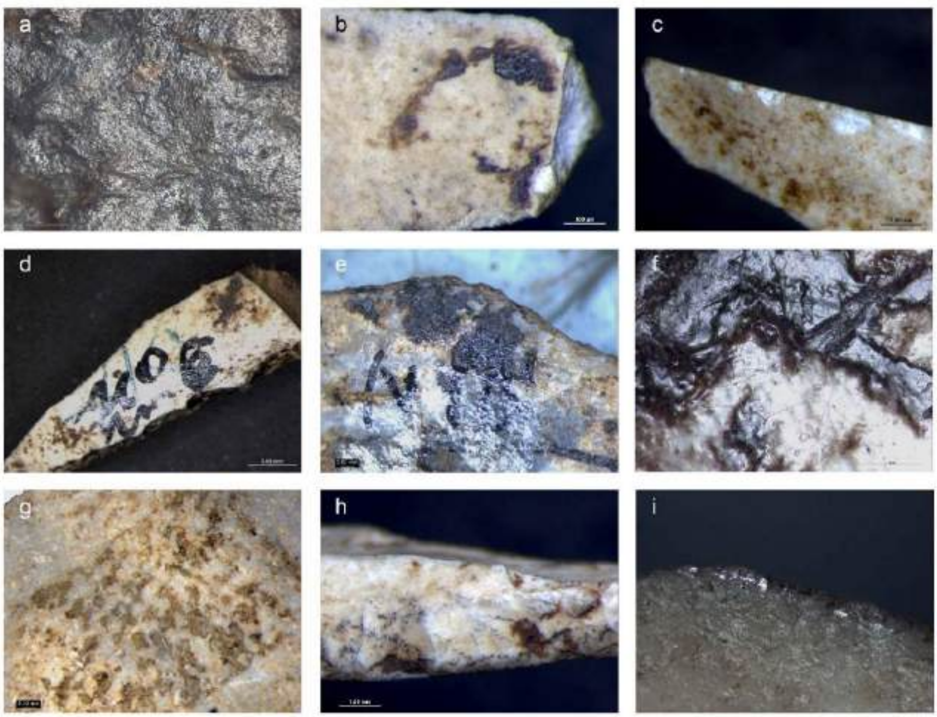

Figure 4.2: Selection of post-depositional and modern contamination on Morín tools. a) Surface alteration (100x); b) Modern tip fracture (25x); c) Modern edge-damage (12.5x); d) Ink, pencil, and clear varnish from labelling (7.5x); e) Clear varnish partially covering a residue (7.5x); f) Close-up of the varnish partially covering a residue (100x); g) Modern glue stain (7.5x); h-i) Pencil marks (12.5x, 100x).

Use-wear and residue analyses (optical microscopy, SEM-EDX, micro-Raman, ATR, GC-MS)

Twenty-three of the 35 analysed tools display evidence of use (Table SI1). Eleven tools are proximal-mesial fragments (N=7) or mesial fragments (N=4) of blades and bladelets. These include: i) one or more bending fractures with hinged terminations (N=4); ii) one or more bending fractures with hinged terminations associated with secondary damage (N=5), and/or a spin-off fracture (N=2). These fracture types can occur during use, during use as projectiles, or during knapping or shaping, but they can also form through accidental dropping and trampling (Rots & Plisson, 2014). Three pointed tools display impact fractures at their tip and/or base including two Châtelperronian points (MOR9 and MOR11). MOR9 displays on the distal extremity a bending fracture with a hinged termination initiated from the ventral face (Fig. 4.3a). The base exhibits a bending fracture initiated from the dorsal face with a complex termination (see Coppe & Rots, 2017) and secondary damage (Fig. 4.3c). Additionally, we documented isolated edge-removals on the mesial-distal right edge of the tool (Fig. 4.3b). MOR11 displays a distal snap fracture with an oblique profile (Fig. 4.3e), edge-damage on the right edge (Fig. 4.3f), and a proximal bending fracture with hinged termination from which a spin-off fracture (impact burination) originated (Fig. 4.3g, h). These traces suggest the tools were hafted as straight points and used as projectile tips (Coppe & Rots, 2017; Metz et al., 2023; Rots, 2016). Four endscrapers (MOR2, MOR17, MOR32, and MOR47) were used for working hide and display edge-rounding (Fig. 4.3i, j), varying from slightly to well-developed, and in a continuous band of greasy polish (Fig. 4.3j), in one case with a transverse directionality (cf. Aleo et al., 2021). Two tools (MOR59 and MOR60) were likely used to work soft animal material and display a greasy band of polish along the edge (Fig. 4.3l), isolated or close edge-removals (Fig. 4.3m), and a slightly developed edge-rounding (cf. Van Gijn, 1990). One tool (MOR12) was likely used to work soft/medium hard plants and displays a band of smooth and matt polish, isolated edge-removals, and a slightly developed edge-rounding (cf. Van Gijn, 1990). One tool (MOR49) was likely used to work siliceous plants and displays a band of smooth and matt polish with longitudinal directionality (Fig. 4.3n), close edge-removals, and a slightly developed edge-rounding (cf. Van Gijn, 1990). One tool (MOR25) displays on one edge a greasy band of polish, light edge-rounding, and isolated edge-removals indicative of contact with soft animal material (Fig. 4.3r). The opposite edge displays a thin line of smooth and matt polish, edge-rounding, and isolated edge-removals (Fig. 4.3p, q). Some more developed spots are domed and exhibit a pitted topography. We interpreted these traces as resulting from contact with fresh bone (cf. Keeley, 1980; Van Gijn, 1990). Eight tools display no wear traces or only minimal traces not indicative of use. The tools with mineral/manganese deposits (N=4) were not analysed for use-wear.

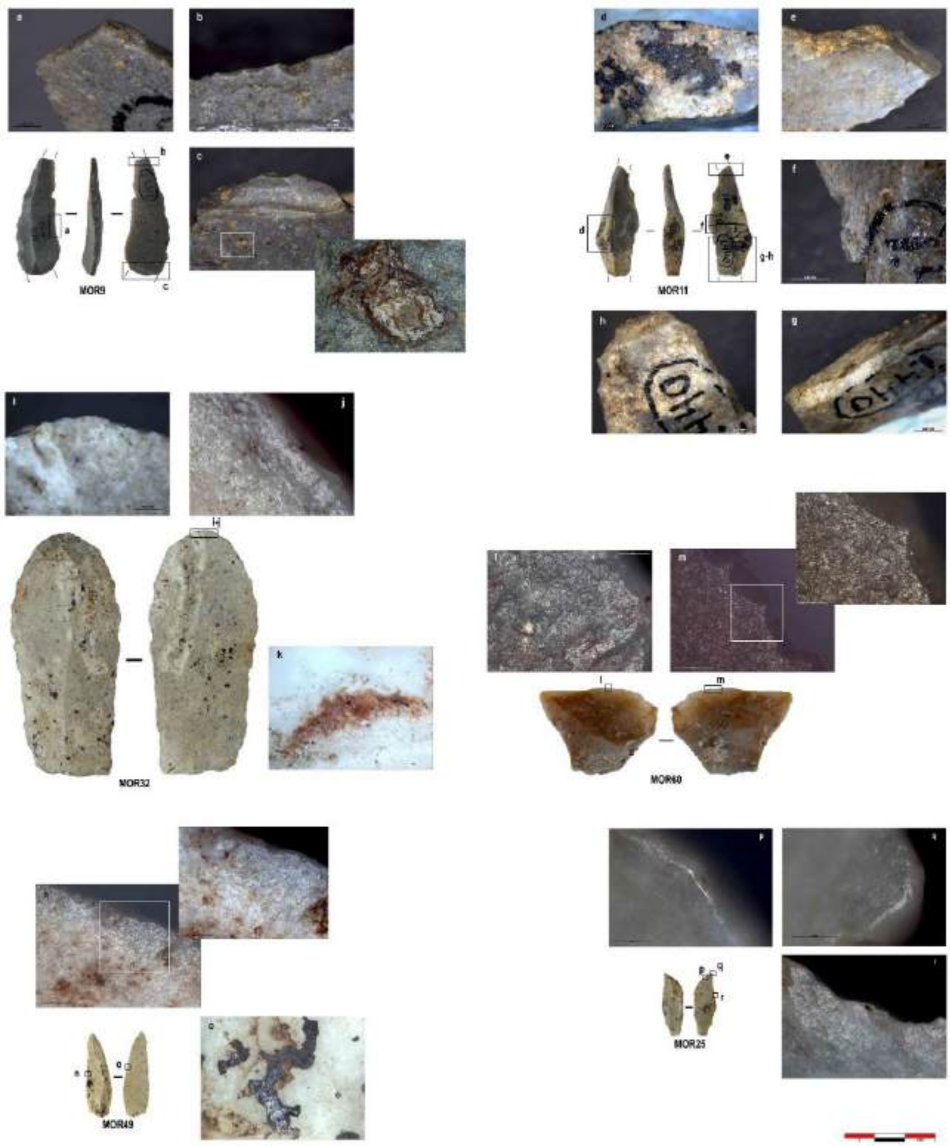

Figure 4.3: Selection of wear traces on Morín tools. MOR9 projectile point a) Distal bending fracture (7.5x); b) Lateral edge-damage (16x), c) Proximal bending fracture with complex termination and secondary damages

(7.5x) and close-up of a residue spot (200x); MOR11 projectile point d) Black compact residue on the lateral edge (7.5x), e) Distal snap fracture (10x); f) Lateral edge-damage (10x), g-h) Impact burination fracture (7.5x) originated from a proximal bending fracture (7.5x); MOR32 hide scraper i) Edge-rounding of the distal edge (7.5x), j) Edge-rounding and band of polish (200x), k) Close-up of a residue spot (200x); MOR60 hide-working flake l) Band of greasy polish along the edge (200x), m) Close edge-damage (50x) and close-up of the polish (100x); MOR49 plant-working flake n) Smooth and matt polish with longitudinal directionality and edge-rounding (100x, 200x), o) Close-up of a residue spot (200x); MOR25 multipurpose tool p) Line of smooth and matt polish (200x), q) Edge-rounding and line of polish (200x), r) Edge-rounding and band of greasy polish (200x).

Twenty-one tools display a black/brownish residue, which is usually smooth and matt, but occasionally granular (Fig. 4.4a, b). When thin, the residue is flat with angular terminations. Sometimes thin, flat, brownish, semi-translucent/translucent residues are associated with this group (Fig. 4.4c). Seven tools display a brown to orange residue, smooth and matt, and with a rounded shape (Fig. 4.4d). When the residue is very thin, it is flat and angular. Cracks are sometimes present on the surface. Sporadically, dark angular inclusions and ochre grains are visible in this residue (Fig. 4.4e). Two tools display a combination of the previously mentioned residue types. One tool displays a reddish granular residue with clear limits and angular terminations.

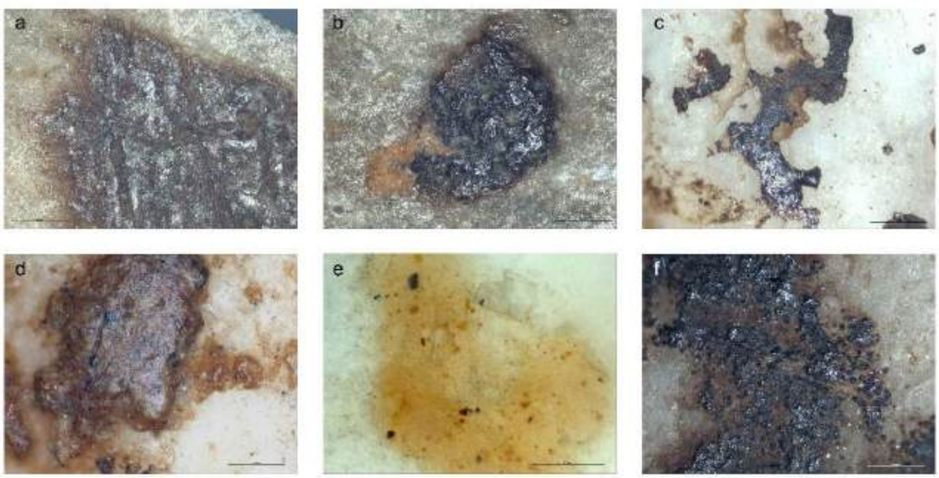

Figure 4.4: Selection of residues on Morín tools. a-b) Black/brownish residues (100x); c) Flat, brownish, angular residue with some cracks on the surface (200x); d) Orange, smooth, and rounded residue with dark inclusions (200x); e) Close-up on a residue spot with granular reddish inclusions (500x); f) Manganese staining (100x).

The results of SEM-EDX analysis (Tables SI2-11) indicate that the residues on four of 10 tools are of organic origin (MOR2, MOR4, MOR9, and MOR13). For all the organic residue samples (N=4), carbon (C) is the most abundant element (always >40%); furthermore, sulphur (S) was consistently registered. Sulphur, together with calcium (Ca) and potassium (K), is linked in the literature with birch tar (Dinnis et al., 2009; Pawlik & Thissen, 2011); however, more recently this interpretation was challenged, and calcium, potassium and/or sulphur were related to the weathering of organic materials (Despotopoulou, 2022). Additionally, iron (Fe) was registered on these four tools, only in the residue spots, suggesting that ochre may have been added to the mixture (Table 4.3). Six residue samples are inorganic, and for all of them the C concentration is below 25%. These inorganic residues present different elemental compositions. Three samples (MOR22, MOR35, and MOR50) are likely manganese (Mn) deposits due to the relatively high proportion of Mn (between 4.5%-16.8%) compared to other elements such as aluminium (Al) (between 1.4%-7.5%), phosphorus (P) (<2%), K (<2%), Ca (<3.5%), magnesium (Mg) (<1%), and Fe (<5.6%). MOR50 differs from MOR22 and MOR35 with a higher concentration of Al over Mn. The sample from MOR12 is likely adhering compacted sediment since it presents a more or less equal distribution of sodium (Na), Mg, P, S, K (<1.5% in each case) and slightly higher proportions of Al (~5%), Ca (<3%), and Fe (<3.5%). The sample from MOR17 is likely an iron oxide deposit since it presents a high proportion of Fe (~26%) and significantly lower concentrations of Al, P, zinc (Zn), and Ca (<2% in each case). Lastly, the sample from MOR42 is likely a Ca P deposit since it presents high proportions of Ca (~20%) and P (~12%) and a significantly lower concentration of Al (1.10%). The concentrations of P and Ca outside the residue spots are 1.7% and 3.1% respectively. In addition to these elements, all the residue spots analysed display variable concentrations of silicon (Si) and oxygen (O) due to the contribution of the flint substrate.

Table 4.3: Summary of SEM-EDX results for tools with organic residues. The ranges represent the minimum and maximum value measured in the different residue spots. The dash symbol (-) means that the element was not identified.

Element	MOR2	MOR4	MOR9	MOR13
Carbon (C)	35.11-48.46%	36.55-55.71%	50.69-62.75%	40.29-55.15%
Oxygen (O)	38.95-42.53%	31.58-41.65%	31.12-36.12%	30.64-32.34%
Magnesium (Mg)	0.32%	0.33%	-	-
Sodium (Na)	-	-	0.34-0.82%	-
Aluminium (Al)	0.51-4.54%	0.69-3.78%	0.62-1%	0.84-3.69%
Silicon (Si)	8.65-13.39%	9.22-12.53%	1.06-8.25%	8.22-10%
Phosphorus (P)	0.77-1.96%	0.39%	0.32-0.76%	0.57-2.44%
Sulphur (S)	0.46-0.67%	0.46%	0.32-0.44%	0.53-1.09%
Chlorine (Cl)	-	0.40-0.42%	0.36-0.58%	0.83%

Potassium (K)	1.13-1.34%	0.22-1.12%	0.31-0.54%	1.39%
Calcium (Ca)	2.22-4.23%	1.03-1.58%	1.30-1.78%	2.67-6.29%
Iron (Fe)	2.40-3.08%	2.16%	0.51%	3.12%

The results of micro-Raman analysis (Tables SI12-16) confirmed the organic nature and plant origin of three of the four residue samples that displayed strong C concentrations (MOR2, MOR9, and MOR13). However, it is not possible to attribute the residue to a particular type of adhesive as many peaks, especially in the region 1655-1000 cm^{-1}, match with vibrations present in both pine resin/tar and birch tar (pristine and/or weathered) FTIR references and/or Raman spectra (Despotopoulou, 2022) (Table 4.4). The possibility that the peaks match with vibrations of other wood products is not excluded. For MOR2, a set of four peaks in the region of 1830-2300 cm^{-1} for both location 1 and 2 likely result from background noise. The Raman spectra of MOR4 show a very limited number of peaks assigned to organic molecular vibrations (Table 4.4). Conversely, multiple positions in the low wavenumbers, under 600 cm^{-1}, match with minerals like quartz, muscovite, iron oxides and (oxy)hydroxides (goethite, hematite). The interpretation of a mineral deposit on MOR22 based on SEM-EDX was further corroborated by micro-Raman.

Table 4.4: Summary of micro-Raman results for tools with potential adhesive remains showing peak positions and their assignments to molecular vibrations (Chen et al., 2022; Cîntă-Pînzaru et al., 2012; Despotopoulou, 2022; Hanesch, 2009; Monnier et al., 2017; Vahur et al., 2011; Yuen et al., 2009). The dash symbol (-) means that the peak was not detected.

Raman shift (cm^{-1})				**Assignment**		**Reference**	
MOR2	**MOR4**	**MOR9**	**MOR13**	**Molecular vibrations**	**Compound/marker**	**Pine resin/tar**	**Birch tar**
2995/96	-	2994	2991	ν(CH_2,CH_3) symmetric		√	
2969/70	-	2970	-	ν(CH_3) asymmetric	Betulin		√
2862/66	-	2869/70	2865	ν(CH_2,CH_3) symmetric		√	
				ν(CH_3) + ν(CH_2) + ν(CH) asymmetric	Betulin, lupeol		√
2676	-	-	2691	ν(O-H) (-COOH)		√	
2626	-	2628	-	ν(O-H) overtones		√	√
1770/78	-	1775/76	1774	ν(C=O)			√
1655/58	-	1654/55	1658	ν(C=C) trans conjugated, amide I	Abietic acid	√	
				δ(CH_2)+ν(C=C)+δ(CCH)	Betulin, betulinic acid, lupeol		√
1536/55	1555	1557/58	1556	ν(C=C) aromatic	Aromatic compounds	√	√
1426/44	-	1425/26	1429	δ(CH_2), δ(CH_3) scissors		√	√

1316/17	-	1316/17	1317	δ(CH_2), δ(CH_3)	Carboxylic acids	√	√
				ω(CH_2)	Amorphous/crystalline cellulose		
-	1235	-	-	δ(OH)+τ(CH2)+δ(CH)	Betulinic acid		√
				/	α-quartz		
1209	-	1211/13	1209	δ(COH), δ(CCH)		√	√
1121/23	-	1127	1121	ν(COC) symmetric		√	√
1099/1104	1100	-	-	ν(CC) ring breathing		√	√
1030	-	1033	-	/	Esters, acids (oleanolic acid?)	√	
				ν(C-O)+δ(CH)+ρ(CH_2,CH_3)	Betulin		√
-	1007	-	-	/	Phenylanine	√	
				δ(SO_4)	Gypsum		

ATR spectra of manganese-stained tools (MOR38, MOR44, and MOR46) match well with the spectrum of the cave sediment. The spectra of the remaining tools (MOR11, MOR40, MOR43, and MOR47) present a limited number of low intensity peaks which are absent in the spectrum of the sediment sample (Fig. 4.5; Table SI17). The tools (except MOR40) display a doublet of peaks in the region 3000-2800 cm^{-1} (C-H stretch bands), of which those in MOR11 have the highest intensity, and a bending band at 1456 cm^{-1}. These bands are characteristics of alkyl fragments (Vahur et al., 2011). Carbonyl (C=O) bands are detected at ~1735 cm^{-1}, 1715 cm^{-1}, and 1700 cm^{-1} and can be due to ester, ketones and carboxylic acids (Vahur et al., 2011). Aromatic compounds were recorded between 1650-1550 cm^{-1}. Other bands appear at ~1415 cm^{-1} and ~1320 cm^{-1} due to C-H deformation of phenolic groups and carboxylic acids. However, carbonates also present a peak at ~1415 cm^{-1} and the peak at ~1315 cm^{-1} can also indicate cellulose. Some of these bands match with plant extractives such as birch tar and pine resin/tar (Chen et al., 2022; Monnier et al., 2017; Vahur et al., 2011); however, the peaks are not unique to these materials, and they are also present in other plant products, like lignin and cellulose (Monnier et al., 2017; Traoré et al., 2018; Yuen et al., 2009), and soil deposits (Pärnpuu et al., 2022; Tinti et al., 2015).

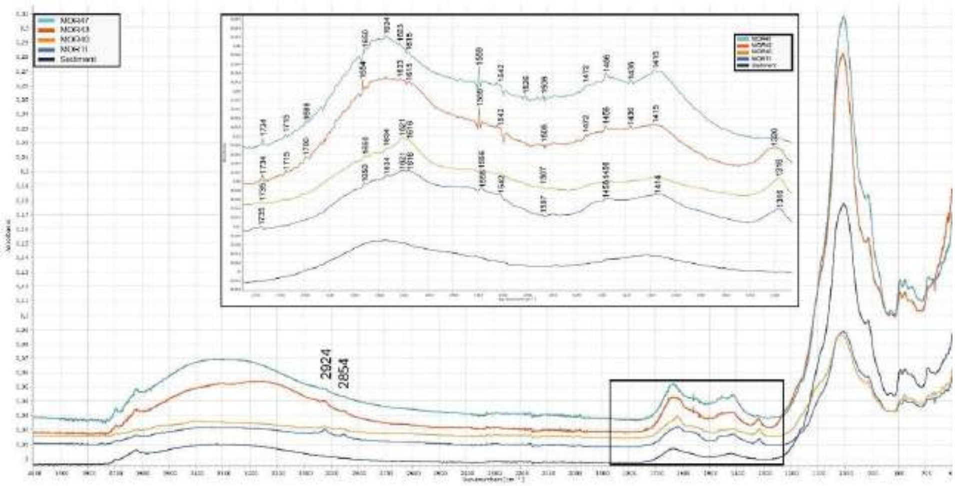

Figure 4.5: Comparison of ATR spectra of MOR11, MOR40, MOR43, MOR47, and cave sediment.

Of the 19 potential adhesive residues analysed by GC-MS, only two (MOR5 and MOR11) have significant preservation of lipids, while the others occasionally show uninterpretable residue signatures, including traces of fatty acids and even-numbered long-chain alcohols (C_{18}, C_{28}, and C_{30}) (Table SI18). The residue sample from MOR5 contains trace amounts of palmitic and stearic acid as well as 1-octacosanol and 1-triacontanol. The bulk of the residue consisted of long-chain *n*-alkanes, ranging from C_{25}-C_{29}, with no clear odd over even preference (Fig. 4.6a). While this *n*-alkane pattern can be suggestive of sedimentary contamination, this was ruled out because the *n*-alkanes were not identified in a sediment sample collected from the same layer. Instead, the signature may be related to the presence of a wax formed from a plant or insect or a bitumen deposit (Blomquist & Jackson, 1979; Bray & Evans, 1961; Bush & McInerney, 2013; Namdar et al., 2007; Scalan & Smith, 1970). The residue sample from MOR11 contains a high amount of saturated fatty acids, ranging from $C_{8:0}$-$C_{18:0}$ (maximizing at $C_{16:0}$). Two unsaturated fatty acids were also identified: $C_{16:1}$ and $C_{18:1}$. The most unique molecules are lactic acid, totarol, and a totarane ketone (Fig. 4.6b). The phenolic diterpenoids are characteristic of *Cupressaceae* that are native Spain, including *Juniperus thurifera* (Gauquelin et al., 1999). Those molecules were not detected in a sediment sample collected from the same layer; therefore, we can rule out sedimentary contamination. Except for MOR11, the three tools showing indication of organic compounds in the ATR spectra did not contain any lipids. Therefore, the identified bands are attributed to soil contamination and soil organic matter. The four residue samples interpreted as manganese stains during optical microscopy investigation were confirmed to not contain relevant organic residues. Two samples (MOR44 and MOR46) do not preserve lipids, while MOR26 and MOR38 display only trace amounts of fatty acids. MOR26 presents $C_{16:0}$ and $C_{18:0}$; the residue sample extracted mechanically from MOR38 presents short- and long-chain saturated fatty acids ($C_{6:0}$, $C_{8:0}$, $C_{9:0}$, and $C_{16:0}$), while the one extracted with the cotton swab imbued with methanol presents only long-chain fatty acids ($C_{16:0}$, $C_{18:0}$). $C_{16:0}$ and $C_{18:0}$ are common compounds found both in plant and animal tissues, but their presence without any other biomarkers suggests contamination, either from the burial environment or post-excavation.

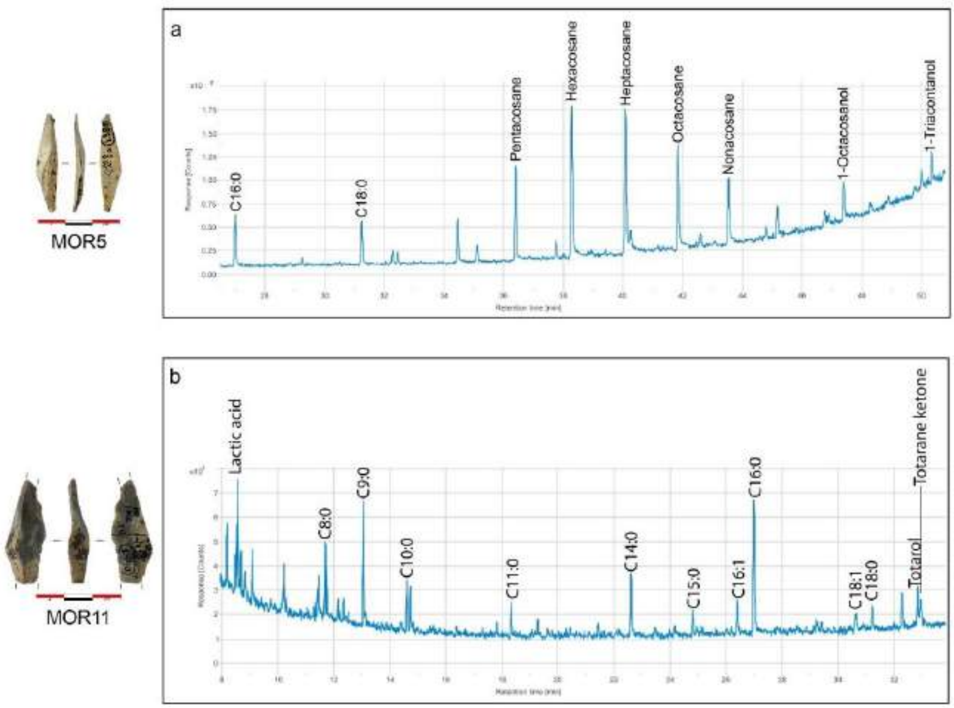

Figure 4.6: Partial chromatograms of MOR5 and MOR11 showing significant lipids preservation.

Discussion

Adhesive technology at Morín Cave: raw materials and mixtures

The detailed examination of stone tools and their residues from Morín Cave allowed the recognition of organic residues that we interpreted as adhesives based on their location, distribution, and molecular signatures. We identified adhesives in the Châtelperronian (N=2), Protoaurignacian (N=1), Early Aurignacian (N=1), and Gravettian (N=1) layers (Table 4.5). No adhesive residues were found on Mousterian tools although one Levallois point (MOR59) displayed a smooth, rounded, orange/brownish residue similar to those observed on MOR2 and MOR9. However, due to time and sampling constraints, the residue on MOR59 was only analysed with optical microscopy. The absence of adhesive residues may be explained by looking at the morpho-technological features of the Mousterian lithic assemblage. At Morín, the Mousterian toolkit displays heavy tools and naturally backed or modified backed tools obtained from different reduction schemes (Bernaldo de Quirós et al., 2010; Maíllo-Fernández et al., 2014). The backed edge increases the gripping of the tools enhancing direct prehension (Delpiano et al., 2019). Additionally, these tools may have been used with hafting configurations that do not require adhesives.

Table 4.5: Summary of results of artefacts with adhesive residues analysed in this study. The slash symbol (/) indicates no data provided. *SEM-EDX and Raman analyses were performed by Bradtmöller et al., 2016.

ID	Cultural affiliation	Tool type	Use	Adhesive	Additives	Analytical methods
MOR9	Châtelperronian	Châtelperronian point	Likely projectile	Tree extractive (resin/tar)	Ochre	Microscopy, SEM-EDX, Raman
MOR11	Châtelperronian	Châtelperronian point	Likely projectile	Cupressaceous resin/tar	/	Microscopy, FTIR, GC-MS
MOR2	Protoaurignacian	Endscraper	Scraping hide	Tree extractive (resin/tar)	Ochre	Microscopy, SEM-EDX, Raman
MOR13	Early Aurignacian	Dufour bladelet	Likely projectile	Tree extractive (resin/tar)	Ochre	Microscopy, SEM-EDX, Raman
MOR5	Gravettian	Partially backed blade	Likely projectile	Plant wax/insect wax/bitumen	Possibly ochre	Microscopy, SEM-EDX*, Raman*, GC-MS

Due to the overall poor preservation of residues and the inconsistent application of analytical techniques because of sampling constraints, only cautious interpretations of raw materials can be made. Raman results likely indicate a tree resin or tar including potentially pine resin and birch tar but could not be narrowed down to species or family level. The GC-MS indicates a resin or tar from a tree of the *Cupressaceae* family, such as *Juniperus thurifera*, was used as a hafting adhesive of one of the Châtelperronian points (MOR11). Palynological data from Morín (Leroi-Gourhan, 1971) should be taken with caution considering the inconsistency of the pollen record. Yet, other studies from this area indicate that the transition between the end of the Mousterian to the Aurignacian was characterised by a shift to more arid and cooler climatic conditions accompanied by a reduction of forest masses. In this cold and dry environment, conifers were the most represented taxa, with *Pinus* being the most abundant species, followed by *Juniperus*, *Fabaceae*, and *Salix* (Allué et al., 2018; Fernández-García et al., 2023; Ochando et al., 2022). Therefore, *Pine* and *Juniper* species were locally available in the environment surrounding the cave. *Betula* occurs in northern Iberia (e.g., in the Mousterian layers of Covalejos and in the Mousterian and Transitional Aurignacian layers of El Castillo (Uzquiano, 2006)), but it is notable only during later periods, <13 ka BP (Uzquiano, 2014). Conversely, *Pinus* and *Juniperus* charcoals are commonly recovered in Late Mousterian and Aurignacian sites of the Iberian Peninsula (e.g., Badal et al., 2011), attesting the wider exploitation of this taxa.

The molecular signature of the sample from MOR5, consisting of long-chain *n*-alkanes (from C_{25}-C_{29}) with no clear odd over even preference, could be attributed to a variety of sources. A plant or animal wax should be considered. Specifically, the signature is similar to degraded dark beeswax (Namdar et al., 2007). However, no other biomarkers of beeswax were identified in the sample. Beeswax is used as an additive in hafting adhesives as a plasticizer (Kozowyk et al., 2016) or used in its pure form as an adhesive on its own. Beeswax has been mixed with conifer resin in the Neanderthal site of Fosellone Cave (Degano et al., 2019), while pure beeswax was used as hafting material for a Late Upper Palaeolithic barbed bone point at Bergkame (Baales et al., 2017). Evidence of exploitation of bee products and beeswax in Iberia dates to later periods; beeswax and propolis were identified inside ceramic vessels as funerary offers or sealants in Bronze Age and Iron Age contexts (Frade et al., 2014; Parras et al., 2015).

The GC-MS results of MOR5 could also be interpreted as related to the use of bitumen, which contains a similar *n*-alkane profile (Scalan & Smith, 1970). Bitumen is a well-known hafting material, and evidence for bitumen adhesives is present in Europe (Doronicheva et al., 2022) and the Levant (Boëda et al., 2008b). In Iberia, molecular analysis supports the use of bitumen, which was identified in the dental calculus of a Neanderthal individual in the northern

Iberian site of El Sidrón (Hardy et al., 2012). This was connected to non-dietary activities such as using the mouth as a third hand, likely during the process of hafting stone tools with bitumen adhesives. The same individual also presented activity-related dental wear, particularly chipped enamel, supporting this hypothesis (Hardy et al., 2012; Radini et al., 2016; Radini et al., 2017). Several bitumen sources are available in Spain, including one at Lames de Parres in Asturias, located 15 km from El Sidrón and at Punta del Cuerno in the modern coastline of Cantabria, located ~30 km from Morín Cave (Kruge & Suárez-Ruiz, 1991). The latter would have been the likely source of bitumen for hunter-gatherer groups inhabiting Morín.

In addition to the organic components, all tools except one, showed evidence of iron oxide (ochre) combined with the adhesive (Table 4.5). Ochre grains were first identified with optical microscopy, and SEM-EXD and/or Raman confirmed their identification. The absence of iron oxides outside the residue spots strongly suggests an anthropogenic origin. We regularly documented ochre, in the form of small iron oxide nodules, in the Upper Palaeolithic layers of Morín, as well as in the Mousterian and Châtelperronian ones (see also Bradtmöller et al., 2016). These nodules did not bear any macroscopic traces of use, and no detailed microscopic examination was performed since it was beyond the scope of this study. Nonetheless, evidence of worked iron oxide nodules from Mousterian, Châtelperronian, and Upper Palaeolithic contexts testify that both Neanderthals and AMH were producing ochre powder for different uses, including that of a loading agent in adhesives. Ochre was likely added to the adhesive to modify its material properties (Kozowyk et al., 2016; Wadley, 2005; Zipkin et al., 2014). Mechanical tests demonstrated that the addition of ochre to plant resin/gum increases the strength of resin and gum adhesives and improves workability (Kozowyk et al., 2016).

The distinction of residues based on morphological characteristics we observed during microscopic investigation does not reflect differences on adhesive composition. Based on the results of elemental and molecular analyses, there is no correlation between the colour and morphology of residues and different adhesive materials. That corroborates previous studies that stated the unreliability of optical microscopy alone to differentiate between different natural sources of adhesives (Aleo et al., 2024; Soriano et al., 2015).

Except for one endscraper, the other tools with adhesive remains are interpreted based on the wear traces as lithic armatures. This is not surprising since projectiles require hafting to be used. However, at Morín, our evidence also supports the hafting practices of tools used in domestic activities, as already reported for other Upper Palaeolithic assemblages (Aleo et al., 2021; Taipale & Rots, 2020).

Comparing adhesives traditions at Morín

Morín Cave is to our knowledge one of the first sites to yield adhesive remains for hafting purposes associated with early Upper Palaeolithic techno-complexes, namely the Châtelperronian and Protoaurignacian. The Châtelperronian is a regional techno-complex restricted to France and northern Spain and dated between ~45-41 ka cal BP (Djakovic et al., 2022; Soressi & Roussel, 2014). It is traditionally associated with Neanderthals (Hublin, 2015; Welker et al., 2016), although several authors have raised doubts about the link between Neanderthal fossil remains and this industry (Teyssandier, 2024 for a synthesis). In the Iberia Peninsula, the Châtelperronian is documented in several sites, but it is only reliably dated at Labeko Koba and Aranbaltza II at 42,6-41,4 ka cal BP and 43,5±2,9 ka cal BP respectively (Marín-Arroyo et al., 2018; Rios-Garaizar et al., 2022). This chronology suggests that the Châtelperronian overlapped with the Aurignacian *senso latu*, which appeared in this area between 43-40,5 ka cal BP (Djakovic et al., 2022; Marín-Arroyo et al., 2018) and is associated with the dispersal of AMH in Europe.

Every attempt to date the Châtelperronian layer (layer 10) of Morín thus far failed (Maroto et al., 2012). Therefore, the chronology of this layer has been established based on its stratigraphic position and typological and technological indicators (Maíllo-Fernández, 2005). The lithic production is oriented toward the production of blades, and more marginally bladelets, from unipolar and bipolar cores. These are then transformed into typical Châtelperronian points (Maíllo-Fernández, 2005). In the overlaying Protoaurignacian layers (layers 9 and 8), tentatively dated at ~36,6 ka BP (Maíllo-Fernández et al., 2001), bladelets and blades are produced in a continuum from prismatic unipolar cores and they are the goal of the production. The former are almost exclusively transformed into Dufour bladelets (Maíllo-Fernández, 2006). Flake production using knapping methods with a discoidal concept also plays a significant role in these layers (Maíllo-Fernández, 2012). The Early Aurignacian (layer 6 and 7) is characterised by the high frequency of carinated pieces and two different laminar reduction methods: blades are produced from prismatic unipolar cores while bladelets from carinated cores (Cabrera et al., 2004).

Despite the presence of different lithic technological traditions within these layers, each with clear reduction schemes and production goals, adhesive technology appears stable in time. The similarity between the Raman spectra of adhesive residues from the Châtelperronian layer 10 (MOR11), Protoaurignacian layer 9 (MOR2), and Early Aurignacian layer 6 (MOR13) is striking (Fig. 4.7), suggesting that a similar organic material or mixture was employed. While, during the Gravettian, different sources were likely exploited for adhesive production. Additionally, a shared aspect we documented from the Châtelperronian to the Gravettian is the continuity in the use of crushed iron oxides as an additive (see also Bradtmöller et al., 2016). The findings from Morín are among the oldest evidence of ochre being used as a loading agent in adhesive mixtures in Europe, together with the one from Grotta del Cavallo and Le Moustier (Sano et al., 2019; Schmidt et al., 2024b). Moreover, if we accept the Neanderthal authorship for the Châtelperronian, this is among the earliest known instances of Neanderthals combining ochre with an adhesive. These finds indicate their sophisticated knowledge and understanding of different materials and their properties. Moreover compound adhesives require advanced executive functions of the brain, such as abstraction, forward planning, and multitasking (Wadley, 2010; Wadley, 2013). This may imply that Neanderthals likely had similar cognitive capabilities of contemporaneous AMHs making compound adhesives in South Africa.

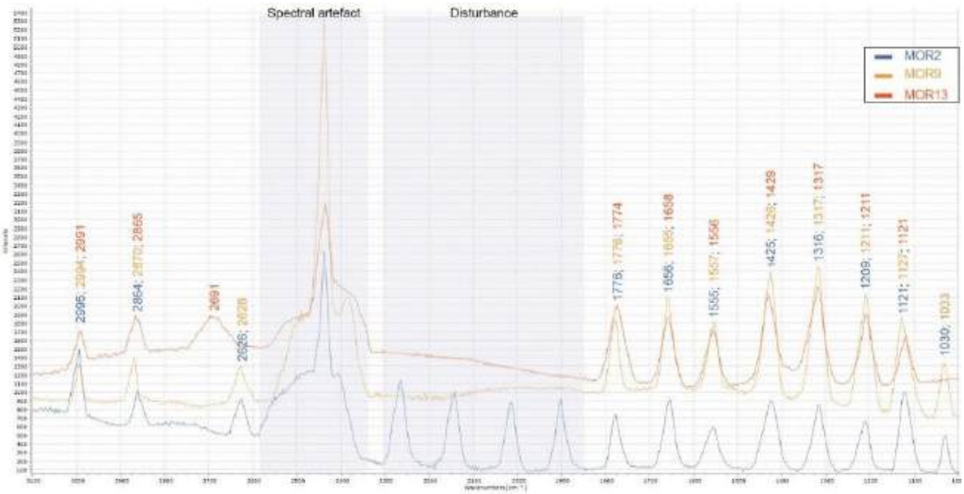

Figure 4.7: Comparison of Raman spectra of MOR2, MOR9, and MOR13. The similarities in the spectra suggest the use of a similar hafting material/mixture between the Châtelperronian, Protoaurignacian, and Early Aurignacian.

The homogeneity of adhesive technology across early Upper Palaeolithic techno-cultural units at Morín can be explained by the climatic deterioration that occurred during the Middle to Upper Palaeolithic transition (Fernández-García et al., 2023). The decrease in forest coverage and homogenisation of species, with an over representation of *Pine,* may have restricted the availability of plants suitable for adhesive production. However, it may also indicate interaction and cultural exchange between groups. Recently, it has been shown that Châtelperronian and Protoaurignacian groups possibly coexisted for ca. 1000 years in northern Iberia (Marín-Arroyo et al., 2018). While the nature of this coexistence is unknown, it is possible that different human groups, independent of their biological classification, interacted, influencing their technological systems. Sharing knowledge and diffusion of ideas may have contributed to the development of adhesive technology adopted by Châtelperronian and Protoaurignacian groups inhabiting this area. Other evidence that was proposed so far to potentially indicate technological/cultural transfer between Neanderthals and AMH are retouched bladelets types (Roussel et al., 2016) and bone lissoirs (Soressi et al., 2013). Although intriguing, at present there is not enough evidence to fully support this hypothesis from the Morín adhesives. The limited sample size from Morín and the paucity of adhesive remains identified dated to this period do not allow any large-scale discussion or intra-site comparison. Further research on this region, focusing on different technologies, may help shed light on the relationships of different human groups during this crucial moment of our history.

Conclusion

Morín Cave stands out as one of the few sites where adhesives dating to the early stages of the Upper Palaeolithic have been discovered. By utilizing multiple analytical methods to study residues, we were able to confirm the presence of adhesives on Châtelperronian tools, a techno-complex that has traditionally been attributed to Neanderthals, as well as Protoaurignacian, Early Aurignacian, and Gravettian tools, linked to AMH. Adhesives at Morín were used mainly to haft tools used as mechanically delivered projectiles, although traces of adhesive were also detected on one endscraper used for hide processing.

The Raman analysis revealed that tree extractives, possibly pine resin or birch tar, were exploited, but the results do not allow any further specifications. GC-MS results identified the use of resin or tar from a plant of the *Cupressaceae* family on a Châtelperronian point and insect or plant wax or bitumen on a Gravettian lithic armature. Additionally, crushed ochre was consistently added to the adhesive mixture in all these techno-cultural units. This discovery highlights the Neanderthals' use of mineral additives for adhesive production and is overall among the oldest evidence of ochre use as a loading agent in Europe. The direct comparison of adhesive traditions attributed to different techno-cultural groups inhabiting the cave highlight a continuity of adhesive technology over time, especially during the Châtelperronian, Protoaurignacian, and Early Aurignacian. Based on the Raman results, a similar material or mixture was used from the Châtelperronian to the Early Aurignacian to haft stone tools. Furthermore, the use of powdered ochre as an additive appears to be a common practice shared among Neanderthals and AMH in and out of Africa.

It is possible that the homogeneity in adhesive traditions during the early stages of the Upper Palaeolithic was due to environmental constraints. We also hypothesise that these similarities may have resulted from interactions between different human groups that coexisted in northern Iberia, exchanging ideas and influencing each other’s technological systems. Further research is necessary to corroborate this hypothesis; northern Iberia, thanks to its richness in archaeological sites with long stratigraphic sequences, has the potential to shed light on further technological and cultural/behavioural innovations among the last Neanderthals and AMH.

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Chapter 5

European Mesolithic adhesives: the bone points from the Dutch North Sea

"The dynamic lives of osseous points from Late Palaeolithic/Early Mesolithic Doggerland: A detailed functional study of barbed and unbarbed points from the Dutch North Sea"

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Supplementary Information:

Abstract

Osseous barbed and unbarbed points are commonly recovered from the Dutch North Sea and other Mesolithic sites of northern Europe. Interpreted as elements of projectile weaponry, barbed points are considered by archaeologists to be a technological innovation in the hunting equipment of hunter-gatherers. However, debate about their exact use and identification of the targeted prey species is still ongoing. To shed light on the function of these tools, we analysed a sample of 17 artefacts from the Netherlands with a multi-disciplinary approach encompassing morphometric, functional, and chemical analysis. ^{14}C-AMS dating yielded the oldest date for a barbed point from the Dutch coast (~13,000 cal BP). The observation of microwear traces preserved on the tools provides solid evidence to interpret the function of barbed and unbarbed points. We show that there were two distinct tool categories. 1) Barbed points hafted with birch tar and animal or vegetal binding were likely projectile tips for terrestrial and aquatic hunting. We provide strong clues to support the link between small barbed points and fishing using wear traces. 2) Points without barbs served as perforators for animal hides. Our results highlight the importance of use-wear and residue analysis to reconstruct prehistoric hunting activities. The functional interpretation of projectile points must also rely on microwear traces and not merely on the association with faunal remains, historical sources, and ethnographic comparisons.

Introduction

The appearance of barbed points in the archaeological record can be seen as a major innovation in the hunting equipment of hunter-gatherers linked to new predatory strategies (Pétillon, 2009). In Europe, the oldest self-barbed point dates to the end of the Gravettian (Pétillon, 2000). The use of barbed osseous points spread during the Magdalenian (Langley et al., 2016) and continued, in different forms, in the following technocomplexes. Barbed points are also widespread and typical artefacts from the Mesolithic. They were found in large amounts in Mesolithic contexts of northeaster Germany (Gramsch, 2019; Hartz et al., 2019), southern Scandinavia (Jensen et al., 2020), western Russia (Zhilin, 2019), southern Baltic (Galiński, 2013; Orłowska & Osipowicz, 2021), the Netherlands (Amkreutz & Spithoven, 2019), and Great Britain (Elliott & Little, 2018). Differences in technology and morphology that characterise barbed weaponry are linked to cultural traditions. Although techno-functional differences exist, their frequency in Mesolithic sites of northern Europe proves that these artefacts were part of a common technological practice shared by Mesolithic people.

For nearly a century there has been debate concerning the function of osseous barbed points. Their function has been inferred based on direct and indirect association with faunal remains, ethnographic comparisons, and sporadically with use-wear studies that are mostly limited to fracture patterns and macrowear traces. Clark (1936, 1948) first interpreted osseous points as fishing gear, while others (e.g., Hartz et al., 2019; Verhart, 2000 and references therein) have argued that these tools were used on both terrestrial and aquatic prey. No direct and exclusive association with fish has been found in the archaeological record so far (see Hartz et al., 2019). However, the indirect association of barbed points and fish bones, mostly from pike, is documented at numerous sites including Abri of Liesbergmühle VI, Switzerland (Nielsen, 2009), Odmut, Montenegro (Cristiani & Borić, 2016), Kunda, Estonia (Verhart, 2000) and the Trans-Urals (Zhilin & Savchenko, 2020). At the Late Mesolithic site of Abri of Liesbergmühle, for instance, many osseous points were found in association with fish remains, which constitute around 20% of the faunal assemblage (Nielsen, 2009). Large barbed points were recovered in direct association with elk remains at High Furlong, England (Hallam et al., 1973) and Tåderup, Denmark (Clark, 1936). These finds document the use of barbed tips for hunting large herbivores.

Historical and ethnographic accounts have shown a varied rather than specialised use of barbed points. Based on ethnographic accounts, detachable harpoons are specialised hunting tools intended for marine mammals and hunting in aquatic environments (Christensen et al., 2016; Weniger, 1992). Undetachable ones (or barbed points) are for fishing, for hunting birds, otters, land mammals, and even for war (Christensen et al., 2016; Pétillon, 2009; Weniger, 1992). Ethnography can provide possible explanations or be useful in forming hypotheses about ancient use but cannot be relied upon on its own. Thus far morphometric studies, fauna associations, and analogies have not resolved the function of Mesolithic osseous barbed points.

To shed light on the function of these objects we studied a collection of osseous points from the Dutch North Sea. Since these artefacts were recovered in secondary deposition, we based our interpretation only on wear traces documented on the points. In addition, to create a reference collection of relevant hafting traces, we carried out an experiment to test whether we can identify different hafting designs based on wear trace characteristics and residue distribution patterns. Hafting methods can inform us about the technological and cultural choices of the Mesolithic people of Doggerland. Different hafting designs may have been selected for different hunting activities or because of their efficiency over other methods (Rots, 2010).

Combining these results creates complete biographies of barbed and unbarbed osseous points. This adds new information relevant to the debate about the function of Mesolithic barbed tips and informs us of the technology and hunting strategies of the Doggerland inhabitants during the beginning of the Holocene.

Materials and Methods

Archaeological points

The assemblage of Dutch North Sea osseous points consists of more than 1000 barbed and unbarbed points that have been recovered from several locations in the province of South Holland. These finds come from waterlogged sediments, which predate the final inundation of the North Sea basin around 6000 years ago (Peeters & Momber, 2014). The sand is dredged from known locations situated several miles off the coast and used for beach replenishment and construction works (Amkreutz et al., 2018). Therefore, the points are subsequently found along the beaches. The finds document the early Holocene occupation of the drowned North Sea prehistoric landscape, named Doggerland (Coles, 1998), which stretched from the Netherlands to Great Britain, Denmark, and Norway (Peeters & Amkreutz, 2020). The Doggerland materials is precious for the Netherlands, where most Palaeolithic and Mesolithic sites are buried many meters below the surface. It has been suggested that Doggerland was the heart of the northwestern European Mesolithic and likely holds one of the most comprehensive records of the Holocene (Clark, 1936). However, since relevant archaeological layers in the sea can be challenging to access, surface finds are the only means of investigating the archaeological heritage in the North Sea. Thanks to the exceptional preservation of organic materials (bone, antler, and adhesive residues) the investigation of barbed points from the Dutch North Sea, despite being surface finds, has the potential to broaden our knowledge of technology and behaviour in Mesolithic Doggerland and provide us a unique window into the inhabitants of wetlands.

Direct ^{14}C dates on 15 barbed points from Dutch Doggerland confirmed their attribution to the Mesolithic period, roughly between 9950-7300 years ago (Amkreutz & Spithoven, 2019; Dekker et al., 2021). The first large-scale study of these objects was conducted by Verhart (Verhart, 1988). More recently, Spithoven analysed a larger assemblage of points with a morphometric and functional approach (Spithoven, 2016; Spithoven, 2018). Verhart distinguished two categories of points based on morphometric attributes: 1) Small points, less than 85 mm in length, were likely used as arrow tips for small prey, fishing, and fowling. 2) Large points, over 94 mm, were likely used as spear tips or harpoons for large marine and terrestrial animals (Verhart, 1988, 2000). The distinction between these groups has now been set at a length of 88.5 mm (Amkreutz & Spithoven, 2019). Both authors agreed to classify these tools as projectile tips but did not find enough evidence to identify with confidence the prey hunted.

This study consists of 17 osseous points (Fig. 5.1) that we selected because all show macroscopic indications of hafting, such as residues/staining, binding impressions, and a difference in surface morphology and wear between tip and base. We decided to analyse a small sample of tools in great detail, using a wide range of techniques that cannot be applied to large assemblages because then the analysis becomes too time-consuming and too expensive. To reconstruct their use-life, we analysed the objects with a multi-analytical approach integrating morphological, metric, chemical, and spectrographic methods. Destructive analyses were performed only on NSM1, 10, and 30 and on loose residues of NSM18 (Table 5.1). Regarding the other artifacts, the owners did not provide consensus for destructive sampling. No permits were required for the described study, which complied with all relevant regulations.

Figure 5.1: Overview of the archaeological points analysed in the study.

Table 5.1: Overview of the archaeological sample of osseous points and destructive and non-destructive techniques applied in this study. For NSM18, the destructive analyses (GC-MS, ^{14}C-AMS), indicated with *, were conducted on loose residues.

Tool ID	Find location	Raw material	Tool type	Preservation	Weathering	Max length mm	Max width mm	Max thickness mm	Hafting indicator	Analysis
NSM1	Maasvlakte	Bone/ antler	Barbed point	Poor	Cracking, exfoliation	78	11	5	Black residue	3D scan, use-wear, GC-MS, ZooMS, ^{14}C
NSM2	Rockanje	Bone	Barbed point	Excellent	Light corrosion	53	9	5	Brownish residue	Use-wear
NSM3	Rockanje	Possibly bone	Barbed point	Good	Modern damage	49	8	4	Black residue	Use-wear
NSM6	Rockanje	Possibly bone	Barbed point	Excellent	No	45	9	5	Binding impressions	3D scan, use-wear
NSM7	Rockanje	Bone	Barbed point	Excellent	Light corrosion	52	9	5	Incisions?	Use-wear
NSM8	Rockanje	Bone	Barbed point	Good	No	38	9	3	Black residue	Use-wear
NSM9	Rockanje	Bone	Barbed point	Good	Corrosion, cracking	34	6	3	Difference in surface preservation	3D scan, use-wear
NSM10	Rockanje	Bone	Barbed point	Moderate	Exfoliation, modern fracture	108	15	6	Black residue	3D scan, use-wear, GC-MS, ZooMS
NSM15	Rockanje	Possibly bone	Unbarbed point	Moderate	Cracking, exfoliation	68	7	4	Black residue	Use-wear
NSM16	Pijnacker	Possibly bone	Barbed point	Good	Cracking, exfoliation	127	13	6	Black residue	3D scan, use-wear
NSM17	Rockanje	Bone	Unbarbed point	Excellent	No	80	9	4	Binding impressions	Use-wear
NSM18	Maasvlakte 2	Bone/ antler	Barbed point	Good/ Excellent	Cracking	51	9	4	Black residue	3D scan, use-wear, GC-MS*, ^{14}C*
NSM22	Maasvlakte 2	Bone/ antler	Barbed point	Good	Cracking, exfoliation, modern residue	58	11	5	Black residue	Use-wear
NSM26	Maasvlakte 2	Bone/ antler	Barbed point	Good/ Excellent	Corrosion	113	13	6	Binding impressions	Use-wear
NSM28	Maasvlakte	Bone/ antler	Barbed point	Moderate/ Good	Cracking, Modern fractures	118	13	3	Black residue	Use-wear
NSM29	Maasvlakte	Bone/ antler	Barbed point	Excellent	No	58	9	5	Discolouration	Use-wear
NSM30	Zandmotor	Bone	Barbed point	Moderate	Cracking, exfoliation	138	15	7	Discolouration	3D scan, use-wear, ZooMS

The points come from several find locations, generally present-day beaches (Fig. 5.2). The points were found along the coast at Rockanje beach (N=9), the Maasvlakte (N=6), and the Zandmotor (N=1). One point was found at Pijnacker, which is located roughly 20 km from the coast, during the construction of a residential area (Spithoven, 2016). Most of the points are owned by private collectors except for four (NSM22, 26, 28, 29) that belong to the Rijksmuseum van Oudheden (Leiden, NL).

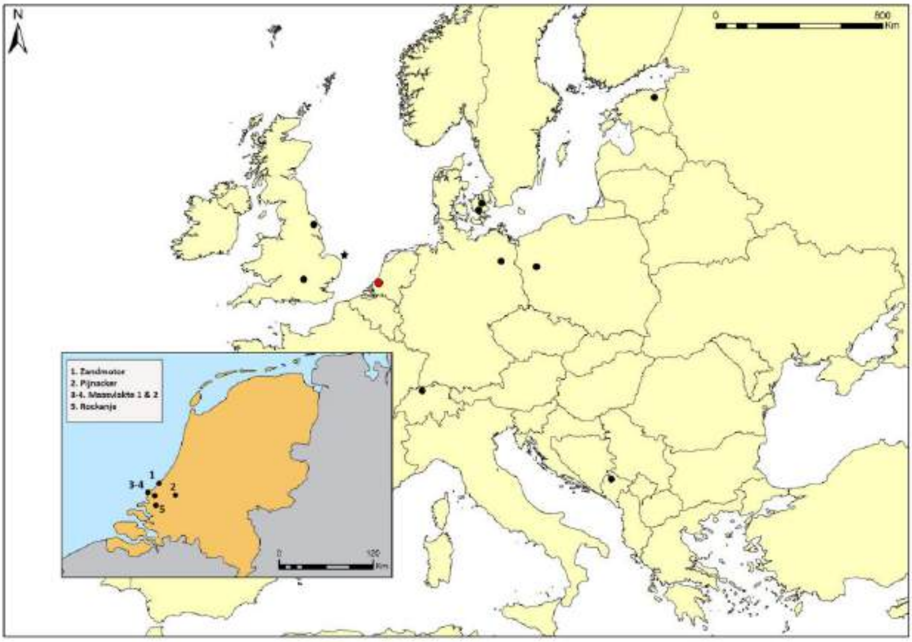

Figure 5.2: Finding locations of the analysed Dutch North Sea points (in the close-up) and locations of archaeological sites cited throughout the text. From the north: Kunda (Estonia), Ulkestrup and Tåderup (Denmark), Star Carr and High Furlong (England), Friesack (Germany), Krzyż Wielkopolski 7 (Poland), Abri of Liesbergmühle (Switzerland), Odmut (Montenegro). The star represents the finding location of the Colinda point at Leman and Ower Banks.

The state of preservation of the points varies from poor to excellent. This was assessed based on the macroscopic presence of weathering and modern damage. Cracking, corrosion, and exfoliation are the most common natural surface modifications observed on the points, with most showing multiple alterations.

The experimental program

Replicas of Mesolithic bone barbed points were made from deer metapodials (S3 Table SI1). Cutting the blanks and the rough shaping was done with modern tools while the barbs were produced using flint tools. The final shaping was done using flint flakes and a grinding stone (sandstone) to match the production traces observed on the archaeological tools. The experimental bone points were then hafted to fletched pine arrow shafts (length: 830 mm, diameter: 9 mm). Two points were hafted with birch bark tar only. On 16 points tar was used in combination with deer sinew (N=8) and lime bast (N=8) bindings. Dried deer sinews, collected from metapodia, were first pounded with a rounded cobble to separate the fibres, moistened with spit, and then wrapped around the point. Raw lime bast fibres, obtained from lime bark stripes, were moistened in water before being used as bindings. Fibres were used plain and not twisted. On 10 points the tar served as bonding material, on eight points it was used for coating lime bast and sinew bindings. The shafts were either bevelled (N=9) or split (N=9) (S4 Fig. SI1). In total, we tested six different hafting arrangements, and all tests were duplicated (Table 5.2) (inspired by Pétillon et al., 2011; Verhart, 2000).

Table 5.2: Hafting arrangements tested during the experiment.

Hafting type	Shaft	Adhesive	Bindings	Hafting method	Nr of experiments
1a	Split	Birch tar	Sinew	The base of the point (covered in tar) is inserted into the split extremity of the shaft. Bindings are used to secure the point	2
1b	Split	Birch tar	Lime bast	The base of the point (covered in tar) is inserted into the split extremity of the shaft. Bindings are used to secure the point	2
2a	Split	Birch tar	Sinew	The point (without tar) is inserted into the split extremity of the shaft and secured with bindings. The bindings are then coated with tar	2
2b	Split	Birch tar	Lime bast	The point (without tar) is inserted into the split extremity of the shaft and secured with bindings. The bindings are then coated with tar	2
3a	Bevelled	Birch tar	Sinew	The point is hafted with tar on a bevelled shaft and secured with bindings	2
3b	Bevelled	Birch tar	Lime bast	The point is hafted with tar on a bevelled shaft and secured with bindings	2
4a	Bevelled	Birch tar	Sinew	The point is secured on a bevelled shaft with bindings. The bindings are then coated with tar	2
4b	Bevelled	Birch tar	Lime bast	The point is secured on a bevelled shaft with bindings. The bindings are then coated with tar	2
5	Split	Birch tar	-	The base of the point (covered in tar) is inserted into the split extremity of the shaft	2
6	Bevelled	Birch tar	-	The point is hafted with tar on a bevelled shaft	2

The arrows were shot into a ballistic jelly cube covered with leather (2 mm thick) (Coppe & Rots, 2017; Jin et al., 2018) using a wooden self-bow mounted on a shooting mechanism (S4 Fig. SI2). The use of a mechanical shooting device has the advantage of reducing variations related to a human archer (Gaillard et al., 2016; Lepers & Rots, 2020). The distance between the front face of the target and the bow was 2 metres to improve accuracy and reduce variability. A piece of foam was placed behind the target to prevent arrows from getting lost after a missed shot. The arrows were shot with an average speed of 39 m/s, simulating the average speed of a traditional bow (Lepers & Rots, 2020). Speed was recorded with a Caldwell ballistic precision chronograph. The arrows were shot a maximum of 25 times to allow hafting traces to develop. When the haft-bond failed, we did not re-haft the projectiles unless the failure was caused by hitting the foam behind or below the target or passing through the target and hitting the ground. The ballistic jelly target was replaced approximately every 18 shots to ensure similar impact conditions and penetration resistance.

Morphometric analysis and 3D

For each archaeological object we recorded the raw material (bone or antler), tool type, maximum length, maximum width, maximum thickness, number of barbs, presence of broken, damaged, and reworked barbs, barb incision shape, base morphology, and base cross-section accordingly to previous classifications (Spithoven, 2018; Verhart, 1988). The identification of the raw material was based on optical examination of the inner material surface and comparison with bone and antler natural and modified fragments from the reference collection of the Laboratory for Material Cultural Studies (Leiden University, NL).

We created 3D models using close-range photogrammetry to create a permanent record of the points selected for destructive analysis and points with relevant hafting traces (SI, S2). Pictures were taken with a Sony A6300 camera equipped with a 50 mm lens. The points were placed on a hand-operated turntable and manually photographed. The smaller objects (NSM1, 6, 9, 19) were photographed at two different height stages. One image every 5° was captured for each face, totally 72 per whole rotation. The larger objects (NSM10, 16, 30) were photographed at three height stages for each face. For large points, a whole rotation comprised 45 photographs, one every 8°. The images were processed, and high-resolution models were created and properly scaled in Agisoft Metashape 1.6.5.

Use-wear analysis

Macro and microscopic wear traces and residues on the archaeological and experimental samples were analysed using established methodologies (e.g., Langejans & Lombard, 2015; Rots, 2010; Van Gijn, 2010). For the low-power examination, we employed a Leica M80 stereomicroscope with an external light source and magnifications ranging from x7.5 to x60 and equipped with a Leica MC120 HD camera. The high-power examination was done with a Leica DM6000 M metallurgical microscope fitted with incident light, bright field illumination, polarising filters, and magnifications ranging from x50 to x500. Images were taken with a Leica DFC450 camera. We documented edge-removals, rounding, polish, striations, and residues. We also recorded asymmetry, rough finishing, axis changes, and striations superimposed to production traces as evidence of tool curation and maintenance following the methodology developed for Magdalenian bone points (Langley, 2015, 2016). Since the archaeological points have a complex post-depositional story and have been handled and curated, we used the location and distribution of wear traces and residue and their association as fundamental criteria to discern between post-depositional and use evidence (Langejans, 2012). Wear traces were evaluated based on the experiment included in this paper, the experimental reference collection available at the Laboratory for Material Cultural Studies (Leiden University, NL), which comprises more than 4000 experiments including more than 200 bone tools used in varied activities and contact materials, on a reference collection of three unmodified bone fragments recovered from the Zandmotor beach (NL), and previously published literature.

The points were subjected to macrofracture analysis to assess whether they were used as projectiles (Bradfield & Brand, 2015; Bradfield & Lombard, 2011; Rots, 2016; Ruta et al., 2022). We analysed fracture types, their position, and distribution patterns, to infer the activity that caused the breakages (Bradfield, 2015). Experiments have shown that the only diagnostic impact fractures (DIFs) resulting from the longitudinal impact on bone tools are spin-off fractures larger than 6 mm and bifacial spin-off fractures (Bradfield, 2015; Bradfield & Brand, 2015). Burin-like fractures (impact burination), which develop on stone projectiles, are generally very rare in bone points (Bradfield & Lombard, 2011). Other fracture types (bending fracture with step, hinged or feathered termination, snap fracture, and crushing) can be caused either by impact, accidental breakage, or post-depositional processes such as trampling. Since fracture variability is extremely high and fractures could also occur accidentally or after deposition, the combination of the macrofracture method, use-wear and residue analysis is fundamental to identify prehistoric hunting tools.

Dating, GC-MS, and ZooMS

We directly dated two adhesive residues to establish the ages of points 1 and 18. ^{14}C-AMS dating was performed at the Centre for Isotope Research at Groningen University (NL). The sample from point 1 was pre-treated with acid only (A) because the material was too vulnerable and too small for the additional base and second acid pre-treatment steps. The sample from point 18 was pre-treated consecutively with acid, a base and a second acid step (ABA-protocol) (see Dee et al., 2020). The base-step was performed at room temperature instead of 80°C since tar/resin can be vulnerable when treated with alkaline solution at higher temperature (SI, S1). The results were calibrated with OxCal v.4.4 (Bronk Ramsey, 2009) using IntCal20 calibration curve (Reimer et al., 2020).

The black residues on points 1, 10, and 18 were sampled for gas chromatography coupled with mass spectrometry (GC-MS). The residue samples were analysed at Inorganic Systems Engineering (ISE) Laboratory (TU Delft, NL). This method allows the identification of materials-specific organic components, or groups of components, that are used as biomarkers to characterise unknown mixtures (Evershed, 2008). A sample of ~10 mg was removed from each object with a sterile scalpel blade. The samples were prepared and analysed by GC-MS following the same methodology employed by Regert et al. (2006) and Urem-Kotsou et al. (2018) (SI, S1). The GC-MS analyses were performed on an Agilent 7890B gas chromatograph system with a split/splitless inlet, coupled with an Agilent 5977B EI MSD interface, an FID and a splitter with corresponding EPC pressure control to achieve this. The GC was fitted with a nonpolar Agilent J&W DB5 MS column. GC-MS chromatograms are interpreted using National Institute Standard and Technology (NIST). The mass spectra were matched against those of authentic standards (betulin and lupeol), by using previously published data and the NIST library.

Bone samples were collected from points 1, 10, and 30 for zooarchaeology by mass spectrometry (ZooMS) analysis, conducted at the York University BioArCh Laboratory (UK). With this technique unique collagen biomarkers are used to fingerprint and identify species of origin from small amounts of bone. One sample of ~10-20 mg was taken from each point using a sterile metal scalpel blade. The samples were analysed with the ammonium-bicarbonate (AmBic) protocol (Van Doorn et al., 2011). The non-destructive buffer extraction was opted because of the small sample sizes (SI, S1). The samples were run on a

Bruker ultraflex MALDI-ToF instrument. The resulting mass spectra were interpreted by comparing the peaks to a list of published peptide marker series for all European, Pleistocene medium to large size mammals (Welker et al., 2016).

Results

Shape and morphometric of archaeological points

The sample features 15 unilateral barbed points and two unbarbed points. Seven points have three barbs; points with more than four barbs are rare. Four points have at least one broken barb. Ten points have at least one damaged barb. Five display traces of a reworked barb. The barbs were mainly cut with oblique incisions (type 2; see (Verhart, 1988)). Different base morphologies and base cross-sections are visible across the sample without one being predominant (Table 5.3). Twelve points belong to the group of small points (length <88.5 mm), while five to the group of larger points (length >88.5 mm) (Amkreutz & Spithoven, 2019; Verhart, 1988).

Table 5.3: Results of the typological and morphometric analysis of the archaeological points. Barb incisions shapes as define by L. Verhart (Verhart, 1988).

		N	**%**
Raw material	Bone	7	41
	Possibly bone	4	24
	Bone/antler	6	35
Tool type	Barbed point	15	88
	Unbarbed point	2	12
N of barbs	1	2	11
	2	2	12
	3	7	41
	4	2	12
	5	1	6
	8	1	6
	N.A.	2	12
N broken barbs	0	11	65
	1	3	17
	2	0	0
	>2	1	6
	N.A.	2	12
N damaged barbs	0	5	29
	1	9	53
	2	1	6
	>2	0	0
	N.A.	2	12
Reworked barb	0	10	59

	1	5	29
	N.A.	2	12
Barb incision shape	Type1. One horizontal incision	2	11
	Type2. One oblique incision	8	47
	Type3. Horizontal parallel incisions	0	0
	Type4. Series of oblique incisions	1	6
	Type5. Two crisscross oblique incisions	1	6
	Type6. Series of crisscross oblique incisions	2	12
	Type7. Incisions like type 5 and 6 where the bottom of the inside angle was widened by cutting away some bone	1	6
	Type8. Incisions like type 5 and 6 where the bottom of the inside angle is widened by parallel horizontal incisions	0	0
	N.A.	2	12
Base morphology	Oval	3	18
	Squared	3	18
	V-shape	4	23
	Asymmetrical V-shape	5	29
	N.A.	2	12
Base cross-section	Flat	7	41
	Oval	4	24
	D-shape	5	29
	Flat/D-shape	1	6
	Tot	**17**	**100**

Ballistic experiment and experimental hafting traces

Six arrows lasted 25 shots, two of them hafted with the split shaft and four with the bevelled shaft, all of which were secured with sinew bindings (S2 Table SI2). However, overall, the arrows hafted with the split shaft lasted longer than the ones with the bevelled shaft (mean 19.28 vs 14.5). Arrows secured with sinew bindings lasted longer than those fixed with lime bast (mean 23.5 vs 9) (Fig. 5.3). Sinew bindings were more resistant than lime bast ones, broke less frequently and allowed a better fixation of the point. We assessed these results with a non-parametric Mann-Whitney U test in Statistica by StatSoft. There are no significant differences in the performance of split and bevelled shafts ($U=30.50$, $p=0.91$). However, sinew bindings are significantly more effective than lime bast bindings ($U=5.50$, $p<0.01$). Five arrows were re-hafted because the tips dislodged by accident during firing. The point hafted on the bevelled shaft without bindings dislodged at the first shot. This experiment was repeated with the same result. The point hafted on the split shaft without bindings lasted for 11 shots before the shaft split. This experiment was repeated, and the shaft split after nine shots.

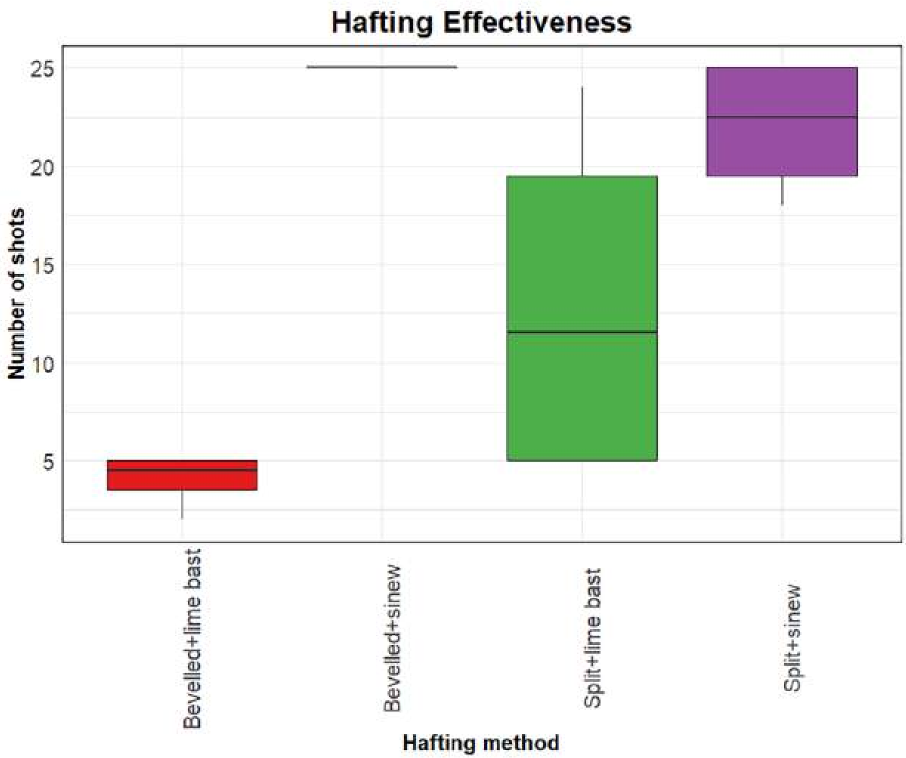

Figure 5.3: Box and whiskers plot showing the relationship between hafting methods and hafting effectiveness.

Hafting traces developed on 10 of 18 experimental points (Fig. 5.4, S2 Table SI2). Discolouration of the hafted part is visible on six points. On four points, discolouration was caused by the tar, while on two, the discolouration is due to bindings (Fig. 5.4, a-b). The discolouration caused by bindings is distributed in bands parallel to each other; this pattern was not observed for the tar-stained pieces (Fig. 5.4, c). Additionally, binding discolouration affects areas of the tool where tar was absent. Discolouration due to tar is visible on both faces on the points hafted in the split shaft while on one face only for those hafted with the bevelled shaft. Macroscopic binding impressions did not develop on the points even though some of them were left hafted for several weeks. None of the experiments displays macrofractures.

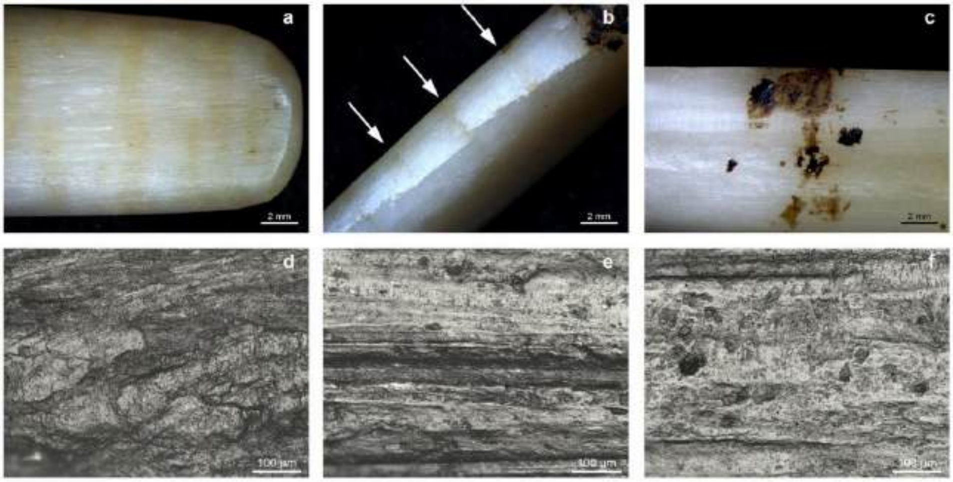

Figure 5.4: Selection of use-wear traces visible on the experimental bone points. a-b) Discolouration in parallel bands due to bindings (7.5x); c) Tar residue at the haft limit and tar discolouration. Note the difference in colour between the hafted part and the non-hafted one (7.5x); d) Greasy dull polish from sinew bindings (200x); e) Smooth and matt polish from lime bast bindings (200x); f) Smooth, domed polish on the mesial area probably from contact with the wooden shaft (200x).

Nine points show hafting polish. Eight of the nine points with polish display a rough, greasy, and predominately dull polish resulting from the contact with sinew (Fig. 5.4, d). One displays a smooth, matt, and bright polish from contact with lime bast (Fig. 5.4, e). Polish developed on seven of the eight points bound with sinew, and on one of eight points bound with lime bast. Four points display a transverse directionality in the polish. The polish is limited to the proximal end of the tools. On the points hafted on the bevelled shaft, the polish is visible on one of the flat surfaces of the tool and on the lateral sides. On the points hafted with the split shaft, the polish developed only on the lateral sides (Fig. 5.5). Two points display a smooth, domed, bright polish on the mesial area (Fig. 5.4, f) probably resulting from the contact with the wooden shaft. Areas in contact with the adhesive did not develop microwear traces.

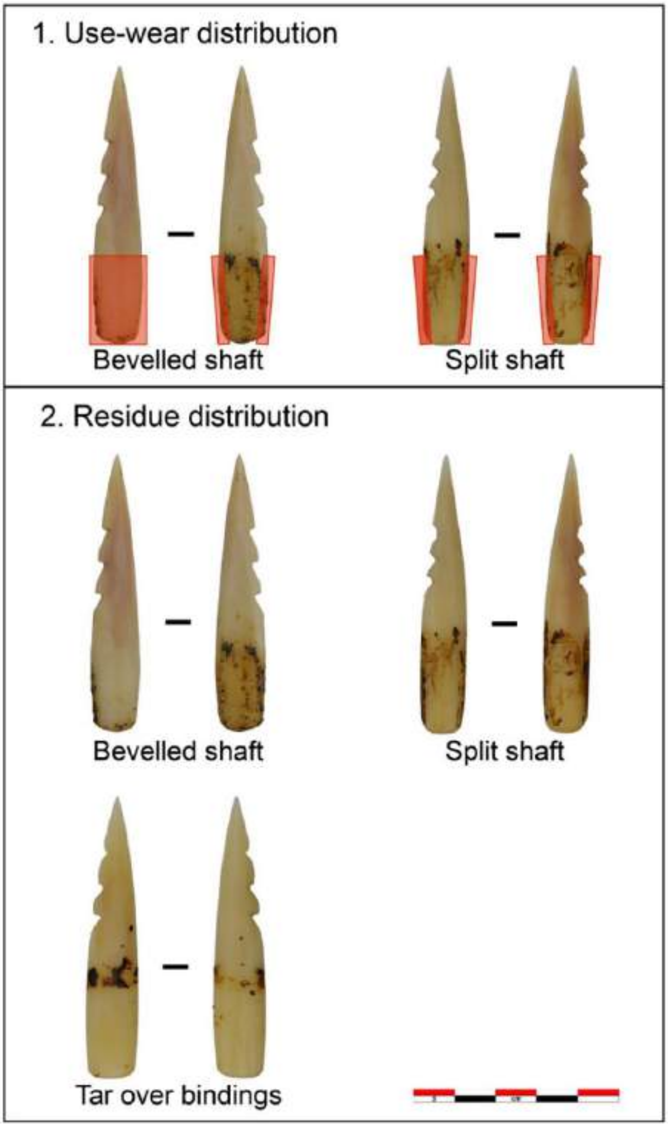

Figure 5.5: Location and distribution of use-wear traces and residue according to the different hafting methods tested in the experiment.

Residue distribution varies according to the hafting arrangement employed (Fig. 5.5). The residues are distributed on both faces of the points in the split haft and one face only in the bevelled points. In both cases, residue may also be present on the lateral sides. On points on which the adhesive was used for coating the bindings, the residue and discolouration preserve only at the haft limit.

Use-wear and residue analysis of archaeological points

The examination of the unmodified bone fragments collected at the Zandmotor beach provided a comparison for interpreting post-depositional traces. Post-depositional polish on these bone fragments has a random distribution with no directionality. Some locations of the surfaces display a flat, smooth, and reflective polish with deep long striations.

Modern contaminations visible on the archaeological osseous points consist of ink and clear varnish for labelling, glue for repairing, plasticine, and wood glue. Wood glue, commonly applied to consolidate organic tools after desalting, hindered the microwear analysis resulting in two of 17 examined points being excluded from the use-wear analysis.

Macrowear traces

We documented a total of 24 macrofractures on 15 points, with most showing multiple fractures (Table 5.4). All of these 15 points present at least one fracture on the tip. Nine of these points also display damage at the base. Crushing is the most visible fracture type (N=7), followed by bending fracture with step termination (N=6) and hinge termination (N=3). Unifacial spin-off fractures are visible on three points, while only one bifacial spin-off fracture is visible. Three points display a snap fracture with a diagonal profile. A single impact burination fracture is observed in the studied assemblage. Only NSM29 displays a diagnostic impact fracture (DIF) (cf. Bradfield & Brand, 2015). This point has on the tip a bending fracture longer than 6 mm with step termination (Fig. 5.6, a) from which a bifacial spin-off fracture was initiated. This fracture type can hardly occur in another way than through use as a hafted projectile (Bradfield, 2015, p. 7). However, considering that osseous barbed points are known to be used as projectiles, it is likely that more of the fractures documented resulted from impact damage; either direct (tip) or recursive (base). Additionally, the fractures visible at the proximal end of the barbed points are consistent with wear from fixed hafting (Langley et al., 2023).

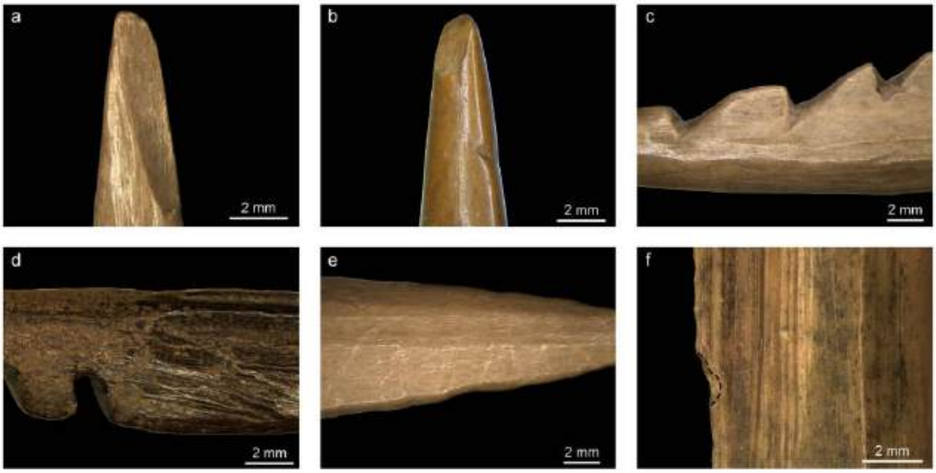

Figure 5.6: Selection of macrowear traces documented on the archaeological points. a) Impact fracture on NSM29 (12x); b) Reworked barb on NSM02 and tip fracture (10x); c) Difference in barbs shape on NSM29 (7.5x); d) Difference in surface preservation between the tip and the base on NSM09 (12.5x); e) Binding impression on NSM26 (7.5x); f) Edge-rounding and edge-removal caused by bindings on NSM06 (16x).

Table 5.4: Results of use-wear and residue analysis on the archaeological points. N.A.= not applicable.

Tool ID	Macrofracture	Barbs macrowear	Use microwear	Hafting microwear	Residue	Observations
NSM1	Tip: fracture but bad preservation	-1 damaged	-Bright and smooth polish domed topography longitudinal directionality	-Isolated spots of bright smooth polish with transverse directionality and striations	-Black residue, mesial proximal part	
NSM2	Tip: bending hinge termination, minor damage	-1 reworked -1 damaged	Not visible	-Left proximal edge: compression due to bindings	-Brownish layer full of cracks on platform	
NSM3	Tip: snap and unifacial spin-off step termination	-1 damaged	Not visible	-Binding impressions on lateral edges -Edge-damage right mesial edges	-Staining base and distal part	-Modern damage on one side
NSM6	Tip: snap	-1 reworked -1 damaged -Difference in shape and size. 2nd barb smaller than 1st one	-'Corrugate' polish. Mainly dull rough and greasy, but with smooth matt and bright spots	-Binding impressions on lateral edges -Edge-damage left mesial-proximal edge -Edge-rounding left mesial-	-Staining all over the surface -Post-depositional residues all over the surface	

				proximal edge -Smooth, greasy, bright polish with flat topography and fine transverse striations		
NSM7	Tip: minor damage Base: bending step termination	-1 reworked -1 damaged -Difference in shape and size. 3rd barb is larger and squared compared to the others -Axis change	-'Corrugate' polish. Mainly dull rough and greasy, but with smooth matt and bright spots	-Edge-damage right mesial edge -Smooth, greasy, bright polish with flat topography and fine transverse striations	-Staining on barbs -Post-depositional residues	
NSM8	Tip: minor damage Base: crushing with hinge and step terminations	-2 damaged/reworked -Difference in shape and size. 2nd barb bigger and rounded than 1st one -Axis change	-'Corrugate' polish. Mainly dull rough and greasy, but with smooth matt and bright spots	-Smooth, greasy, bright polish with flat topography and fine transverse striations	-Staining distal part -Black residues, reddish at extremities, proximal part	
NSM9	Tip: bending step termination Base: bending step termination, minor crushing	-1 damaged	-Not visible, very corroded tip	-Difference in surface preservation. Tip very corroded compared to base -Binding impressions lateral edges -Edge-damage right mesial edge	-No residue	
NSM10	Tip: modern fracture Base: impact burination hinge termination	-3 broken (modern)	N.A.	-Binding impressions lateral edges	-Staining base, barbs, and tip -Clump of residue proximal part. Black.	-Modern wood glue on the surface except on the residue -Modern fracture. Point repaired with glue
NSM15	Tip: minor damage	N.A.	-Fine transverse striations	-Surface very worn and corroded	-Staining all over the surface	
NSM16	Tip: crushing with hinge step terminations, unifacial	-2 minor damage	N.A.	Not visible	-Staining proximal and distal -Clump of residue	-Wood glue all over the surface except on the residue

	spin-off step termination Base: bending step termination, minor crushing with hinge step terminations				proximal part. Black.	
NSM17	Tip: bending hinge termination	N.A.	-Smooth and bright polish -Fine transverse striations	- Smooth, greasy, bright polish with flat topography and diagonal and longitudinal striations	-Post-depositional residues all over the surface	
NSM18	Tip: crushing Base: step terminations	-1 chipped	-'Retrieval' cut marks	Not visible	-Black residue proximal-mesial part with fibres impressions.	
NSM22	Tip: minor damage Base: hinge termination		-Bright and smooth polish domed topography longitudinal directionality (2nd barb)	-Binding impressions lateral edges -Edge-damage right mesial edge -Post-depositional polish	-Staining proximal-mesial part. Black, compact, homogeneous, very polished with lot of striations -Modern residue (plasticine)	
NSM26	Tip: snap Base: crushing with step termination and unifacial spin-off step termination	-1 reworked	Not visible	-Binding impressions encircling the base -Edge-rounding proximal part	-Grey staining (modern?) -Microscopic residues mostly located on the distal part (rougher). Residues covered with glue/varnish. Post-depositional - A few reddish/orange residues, very granular	-Curation hinder the observation of use-wear and residues. Striations everywhere probably connected to brushing
NSM28	Tip: minor damage	-1 broken -1 chipped -7th 8th barbs very rounded and not pointed compared to others	Not visible		-Staining barbs and distal part -Black residues, reddish at extremities, proximal part -Reddish/brownish residue proximal part with diagonal orientation	-Modern fractures. Point repaired with glue

NSM29	Tip: bending step termination (>6mm) and bifacial spin-off step termination. DIF Base: crushing hinge terminations	-1 reworked -3rd barb sharper compared to the others	Not visible	-Binding impressions lateral edges -Edge-damage right mesial edge -Edge-rounding proximal part -Smooth, matt, metallic polish with flat topography and fine transverse striations -Discolouration proximal part	-Isolate black residues on the platform, inside barbs incisions and tip. Very reflective	-Modern glue/varnish in several locations
NSM30	Indet. Bad preservation	-1 chipped	Not clear	-Binding impressions lateral edges -Discolouration proximal part	-Staining proximal part	

Five barbed points display a reworked barb (Table 5.4). Reworked barbs were mostly partially removed (N=4), leaving only a slightly raised scar (Fig. 5.6, b), or completely removed (N=1). These scars are always located on the distal part of the points close to the tip and never observed on the mesial or basal area. Those traces may be associated with removed fractured/damaged or blunted barbs (cf. Langley, 2015). Besides, resharpening and repairing the tip by grinding and scraping would have resulted in a shortening of the point at the tip affecting the barbs at that end which may have been removed to maintain a sharp functional extremity. In addition, we identified traces of rejuvenation of the distal area. Rejuvenation resulted in a visible modification of barb shape and sometimes asymmetry and axis change (Langley, 2015). Five points show a clear difference in shape or size between the top barb(s) and the lowest ones (Fig. 5.6, c). In two cases (NSM7 and 8), a change in the point axis is also visible. This evidence is associated on two points with a rough finishing of the distal area, with coarse striations macroscopically visible and overlaying production traces. These traces strongly suggest that these objects were often repaired and reworked and their use-life extended as much as possible. We also observed the so-called 'retrieval' cut marks on the distal-mesial section of NSM18. These marks are described as short, oblique, and isolated incisions that form when the point is retrieved from inside a carcass or cut away from the haft for repair or retooling (Langley, 2015, p. 347).

Two points display surface corrosion at the tip, while their proximal surfaces are better preserved (Fig. 5.6, d). Two points show discolouration of the proximal part (Table 5.4). The differences in surface modification between the proximal and distal parts demonstrate that these parts of the points were exposed to different environments. Ten points have macroscopic binding impressions (Table 5.4). Binding impressions appear as regularly spaced depressions on the bone surface (Fig. 5.6, e). Nine points have binding impressions on the lateral sides only, whereas one point (NSM26) has binding impressions encircling the base. These impressions are associated with edge-damage on six objects and with edge-rounding on three (Fig. 5.6, f).

Microwear traces

We documented microwear traces related to use on five small barbed points (Fig. 5.7, Table 5.4). None of the large barbed points display distinctive microwear traces. On three points (NSM6, 7, 8), a 'corrugated' polish is visible on the active part. The polish is mainly dull, rough and greasy, but with smooth, matt, and bright spots (Fig. 5.7.1, a-b). Based on the characteristics of the polish (location, distribution, texture, and topography), extensive visual comparisons with the experiment reference collection in Leiden -which includes two bone points shot into a salmon and 48 flint tools used on fish- and existing literature, we interpret these traces of wear to result from contact with fish (Clemente-Conte et al., 2020; Högberg et al., 2009; Van Gijn, 1986). This polish closely resembles the polish on an experimental point shot into a salmon (Fig. 5.8, a-b) and on experimental flint tools used to process fish (Fig. 5.8, c). Polish from contact with fish displays on both flint and bone features of contact with soft and hard materials and it is characterised by a corrugated texture and a dull greasy polish with smooth and bright spots. Two points have a bright smooth polish with domed topography and clear longitudinal directionality. Based on its characteristics, this polish is interpreted as associated with contact with bone and it probably resulted from contact with animal bones during impact. It is located close to the tip of NSM1 and on the second barb of NSM22 (Fig. 5.7.2, a). A similar bone polish is also visible on an experimental point used to shoot a carcass (Fig. 5.8, d-e). Besides, polish directionality, longitudinal on both experimental and archaeological tools, corroborates the interpretation of the use motion (shooting). Therefore, the use-wear traces on barbed tips (NSM1, 6, 7, 8, 22) most closely corresponds to the use-wear traces on experimental tools used to hunt fish and land animals.

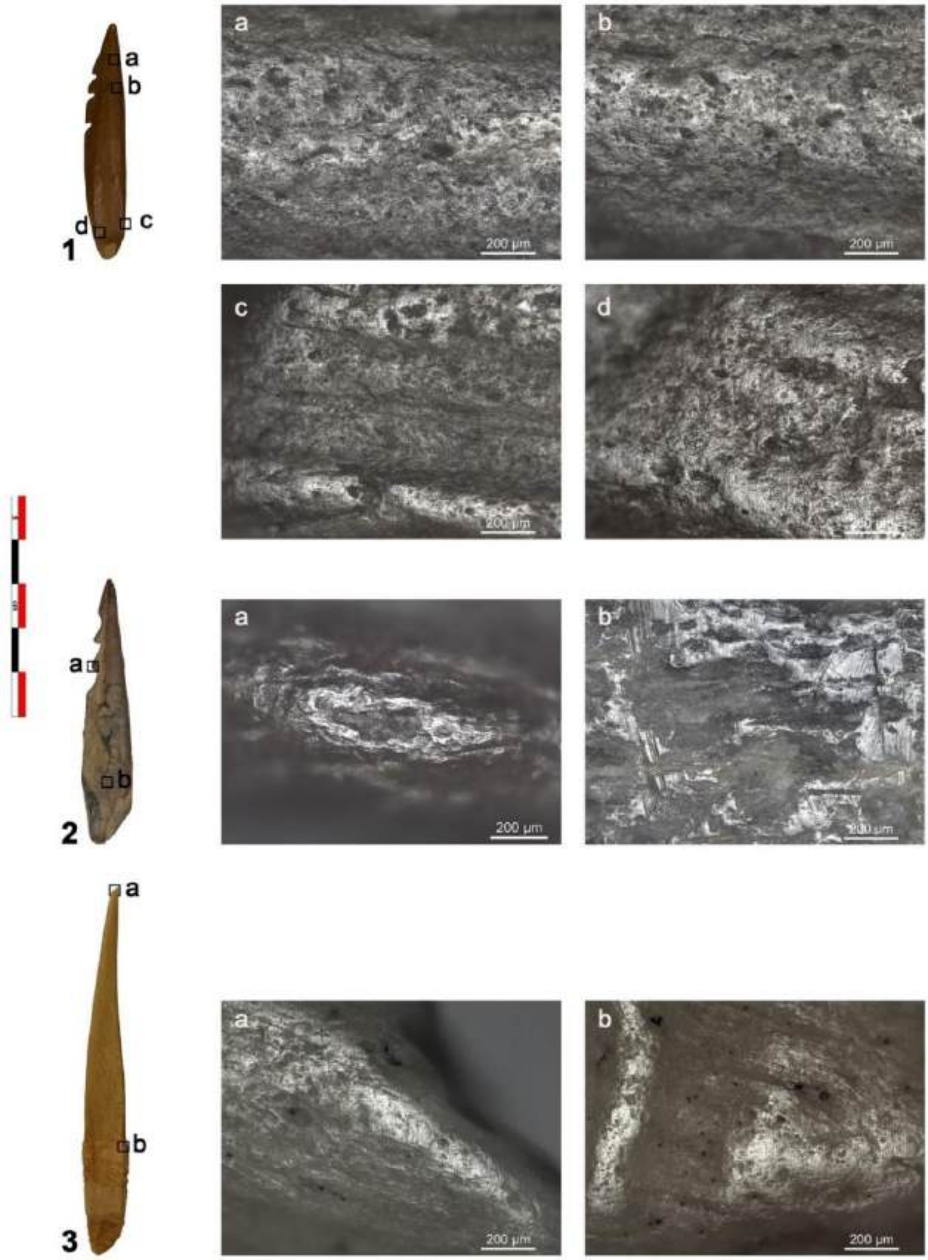

Figure 5.7: Selection of microwear traces documented on the archaeological points. 1. NSM07 a-b) Polish with corrugated texture likely resulting from contact with fish; c-d) Polish and fine transverse striations from sinew bindings. 2. NSM22 a) Bright smooth polish with longitudinal directionality likely resulting from contact with bone; b) Post-depositional polish with long deep striations. 3. NSM17 a) Polish and transverse striations from boring animal hide; b) Smooth and bright polish from hafting. Magnifications 100x.

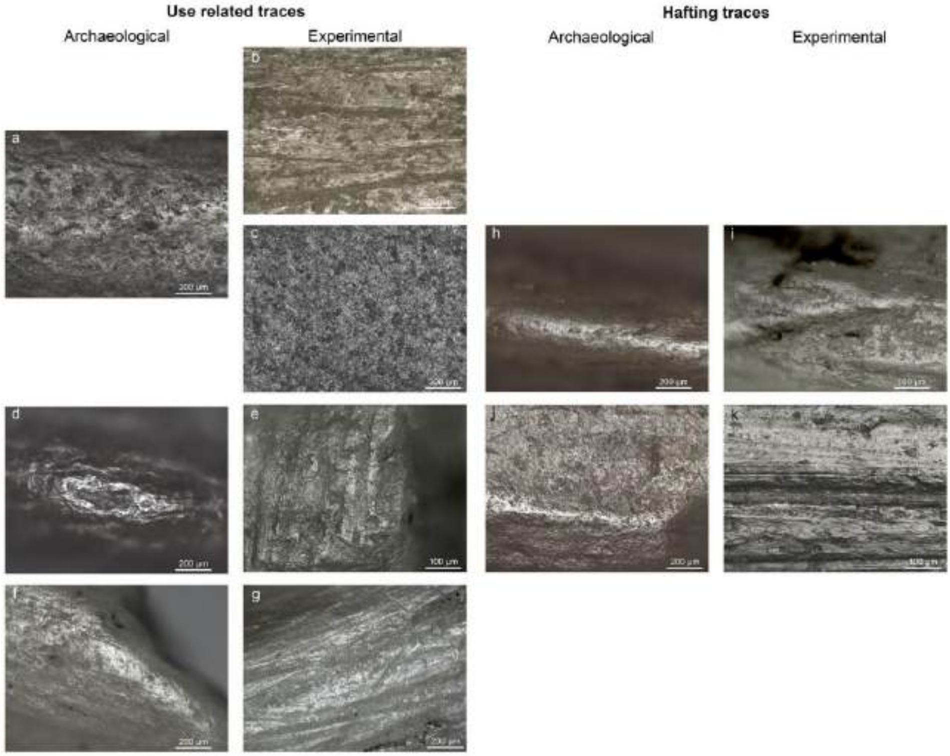

Figure 5.8: Comparison between archaeological and experimental wear traces. a) Fish polish on NSM07 (100x); b) Polish on an experimental bone point from shooting salmon (100x); c) Polish on an experimental flint tool used to process fish (red snapper) d) Bone polish on the second barb of NSM22 (100x); e) Bone polish on an experimental point used to shot a carcass (100x); f) Polish and short transverse striations on NSM17 from boring animal skin (100x); g) Polish and short transverse striations on an experimental borer used to perforate deer skin (100x); h) Polish and short transverse striations on the base of NSM08 from sinew bindings (100x); i) Polish and short transverse striations from sinew bindings (200x); j) Smooth bright polish on the base of NSM29 from plant bindings (100x); k) Flat polish from lime bast bindings (200x).

Two unbarbed points (NSM15, 17) display striations with a transverse orientation, indicative of a boring motion, on the tip. On NSM17, striations are associated with a smooth, greasy, flat, bright polish (Fig. 5.7.3, a). Based on the wear traces and comparison between archaeological and experimental microwear, the unbarbed points were likely used as perforators to work animal materials, likely hide (Fig. 5.8, f-g). At least one of the two unbarbed points bears evidence of hafting. The polish visible at the base resembles the one at the tip (Fig. 5.7.3, b). Thus, it is likely that a strip of hide or leather was wrapped around the tool to provide a better grip (Rots, 2010).

Hafting traces were documented on five barbed points (Table 5.4). Four points (NSM1, 6, 7, 8) display a smooth, greasy, flat, and bright polish on the mesial-proximal area. This polish is mostly visible on the lateral sides of the tools and is better developed on the high reliefs of the surface compared to low areas (Fig. 5.7.1, c-d). On the base of NSM29, a smooth, matt, flat, almost metallic polish is present. Fine, short, transverse striations are always associated with these micro-polishes. These traces are interpreted as originating from sinew and vegetal bindings of the hafting arrangement. Sinew is identified due to the similarities between the archaeological and experimental traces (Fig. 5.8, h-i). The experimental polish from lime bast does not provide an accurate match for the archaeological material (Fig. 5.8, j-k) but we can still interpret some of the binding polish as being related to contact with plant material. NSM22 displays a very flat, smooth, and reflective polish with long deep striations on the mesial-proximal area that we interpret as post-depositional (Fig. 5.7.2, b).

Residue analysis

Residues are present on 12 out of 17 points. Four points (NSM3, 15, 22, 30) display only black staining/discolouration, meaning no physical three-dimensional residues are preserved. NSM9 has no residue. We excluded NSM26 because the residues are located under a layer of wood glue.

Three points (NMS6, 7, 17) have microscopic residues randomly distributed on micro-cracks and grooves of the bone. They are elongated like the cracks, black in colour, and highly reflective when examined with the metallographic microscope in normal light. They are interpreted as post-depositional, most likely related to rooting.

NSM2 displays a brownish residue which is limited to the platform. The residue appears as a homogeneous, smooth, and reflective layer full of cracks. Cracks are not visible in other locations of the point. SEM-EDS analysis confirmed the inorganic nature of the residue. All the EDS spectra show a strong contribution of calcium (Ca) and phosphorus (P), probably originating from the underlying bone's hydroxyapatite ($Ca5(PO4)3(OH)$) (Despotopoulou, 2022).

Four points (NSM1, 10, 16, 18) display large, visible, black residue on the proximal/mesial portion. These residues are preserved on one side of NSM1, NSM10, and NSM16, and on both sides of NSM18. The residues have a three-dimensional rounded shape, a granular texture, and are sometimes cracked. When examined with the metallographic microscope, the residues are black, brownish at the limits, reflective in normal light, and dull in cross-polarised light. On NSM10 and NSM16, the residues are located at the base of the points. On

NSM1, the residue extends 47 mm from the base toward the tip of the object. On NSM18, the residue covers almost half of the point (maximum length 25 mm), covering the third and fourth barbs (Fig. 5.9.1). The surface of this residue displays elongated white/grey striations with a diagonal orientation that may be the remains of fibres or their impressions (Fig. 5.9.1, a-b). However, when analysed with high magnifications (200x-300x), they lack any visible structure, e.g., elongated cells organised in fibrous bundles. Thus, they are likely fibre impressions. The residue on this object is black, terraced, and granular, with some orange inclusions that are semi-translucent in cross-polarised light (Fig. 5.9.1, c). On top of the residue, a granular rusty orange layer is visible, likely the result of the degradation of organic material (Fig. 5.9.1, d). Based on their distribution, morphology, surface characteristics, and the reference collection, these residues are interpreted as organic adhesive remains.

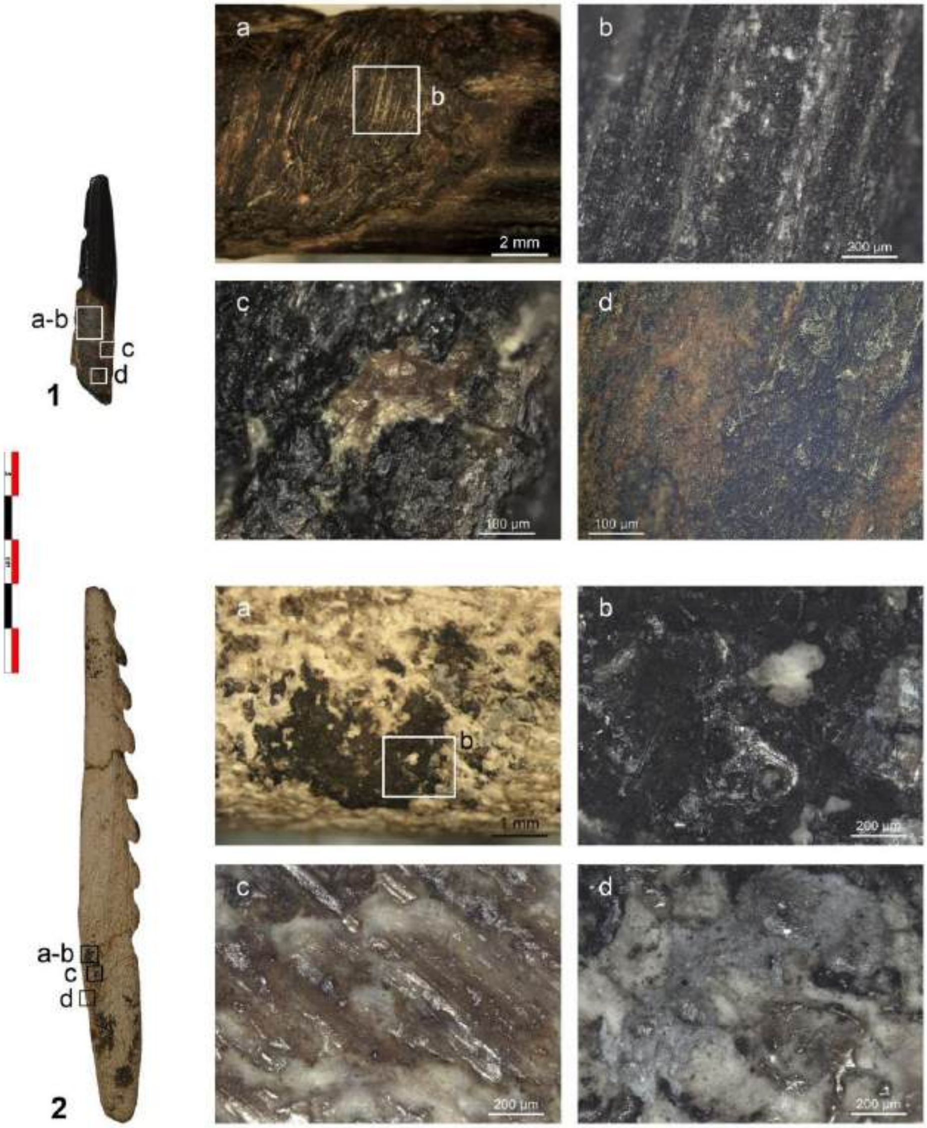

Figure 5.9: Adhesive residues documented on the archaeological points. 1. NSM18 a) Black residue (10x); b) Close-up of the possible fibre impressions (100x); c) Detail of the orange semi-translucent inclusion (200x); d) Granular rusty orange layer on top of the residue (200x). 2. NSM28 a) Granular black residue (20x); b) Close-up of a (100x); c) Granular brownish residue with oblique orientation (200x); d) Modern grey residue (200x).

Residues on NSM8 and NSM28 are also interpreted as potential adhesive remains. Micro-residues are visible on both sides of NSM8 towards the base. Bigger residues are black, some are brownish at the limits, with a three-dimensional rounded shape and a granular texture. NMS28 displays a combination of residues (Fig. 5.9.2). Spots of granular black residue are visible at the base, partially covered by a modern grey residue (Fig. 5.9.2, a-b-d). Micro-Raman and micro-FTIR indicated the organic nature of these residues. The detected peaks can be assigned to pine tar, pine resin, or birch tar (Despotopoulou, 2022). In addition, on the lateral side of the point, a granular brownish residue with an oblique orientation is visible (Fig. 5.9.2, c). The latter is located very close to a modern fracture that was repaired with glue. Therefore, its modern origin cannot be ruled out completely.

Destructive analyses: dating, GC-MS, and ZooMS

Two 14C-AMS dates were obtained from the residue samples belonging to points 1 and 18. NSM1 (GrM-27499) has an estimated age of 9275 years BP and NSM18 (GrM-27889) of 11,065 years BP. Calibrated dates range between 10,573-10,298 cal BP for NSM1, and 13,095-12,843 cal BP for NSM18 (S2). These ages confirm the attribution of NSM1 to the Mesolithic, while NSM18 is attributed to the Upper Palaeolithic (Table 5.5).

Table 5.5: Results of the destructive analyses on the archaeological barbed points. X= no result; N.A.= not applicable. ^{14}C ages (in yrBP) are calibrated to calendar years with software program: OxCal, version 4.4 (Bronk Ramsey, 2009) using calibration curve IntCal20 (Reimer et al., 2020). *age range for 95,4% probability.

Tool ID	^{14}C age (yrBP)	Calibrated age range (yrBP)	ZooMS identification	GC-MS
NSM1	9275±35	10,573-10,298	*Cervus elaphus*	Birch tar
NSM10	N.A.	N.A.	*Bison - Bos primigenius*	Birch tar
NSM18	11,065±50	13,095-12,843	N.A.	Possibly birch tar
NSM30	X	X	*Cervus elaphus*	N.A.

The GC-MS analysis of the residues indicates the presence of pentacyclic triterpenoids with a lupane skeleton and their degraded derivates, and saturated and unsaturated fatty acids and diacids (Table 5.6). These markers are typical of birch bark tar (Orsini et al., 2015; Regert, 2004). Lupeol and/or betulin are present in two adhesive samples (NSM1 and NSM10), while in NSM18 only degraded products of these compounds are identified. Although degraded, residue samples from NSM1 and NSM10 can be confidently interpreted as birch bark tar. The peaks in the chromatogram of sample NSM18 are low in intensity which makes it hard to identify the chemical components. Some peaks provide a very low match with compounds with betulin structure. Probably, there are remnants of birch bark tar, but the sample is too degraded to allow a confident interpretation based only on GC-MS results. Degradation can be induced by the natural ageing of the material during the burial period or as a consequence of intentional transformation involving heating processes. Since the bone points come from different primary sites, it is possible that NSM18 was subjected to different taphonomic processes compared to NSM1 and NSM10 which influenced the preservation of biomarkers. Besides this, differences in the Palaeolithic and Mesolithic tar production methods can also be responsible for the dissimilarity in the results. It is possible that the adhesive for NSM18 was produced with a different technique or by using higher temperatures, which affected the preservation of molecules. No chemical compounds for other typical adhesive materials, such as pine resin, waxes, or gum are present. It is therefore likely that birch bark tar was used as a single-component adhesive for hafting the points.

Table 5.6: Chemical compounds identified with GC-MS on each sample. Y stands for yes; N stands for no.

Chemical component	**Retention Time**	**Tool ID**		
		NSM1	**NSM10**	**NSM18**
Glycerol, 3TMS derivative	11.7	Y	Y	Y
Nonanoic acid, TMS derivative	13.0	Y	Y	Y
Palmitic Acid, TMS derivative	27.0	Y	Y	Y
Bisphenol A, 2TMS	27.3	Y	Y	N
Cyclic octaatomic sulfur	27.2	N	N	Y
Stearic acid, TMS derivative	31.3	N	Y	N
13-Docosenoic acid (E)/ Euricic Acid	34.1	N	N	Y
13-Docosenoic acid, (Z)-, TMS derivative	38.9	Y	Y	N
α-Lupane	43.7	Y	Y	Y
α-Lupane	44.8	N	N	Y
Lupa-2,20(29)-diene	46.0	Y	Y	N
α-Betulin I, TMS	46.4	Y	Y	N
α-Allobetulin	47.2	Y	Y	N
Allobetul-2-ene	49.5	Y	N	N
Lupeol, trimethylsilyl ether	51.0	Y	Y	N
Betulone, TMS derivative	52.8	Y	Y	N
Betulin, bis-TMS	53.2	Y	Y	N
Betulinic acid, O,O-bis-TMS	53.5	Y	Y	N
Allobetulin, TMS derivative	54.2	Y	N	N

Since osseous points are heavily modified by manufacturing, use, reuse, and post-depositional alterations, it is not always possible to identify the raw material. Based on macroscopic observation, seven points are bone, four are probably bone, and six are either bone or antler. Based on the collagen peptide markers NSM1 and NSM30 are identified as Bovidae/Cervidae and NSM10 as cattle (Table 5.5). For the Mesolithic North Sea area, the label Bovidae/Cervidae refers to a group of species that all share the same markers and consists of either red deer or elk (see Dekker et al., 2021 and references therein). In addition, the presence of a peak at m/z 2216 in both samples also suggests we can further specify this to red deer (Jensen et al., 2020). NSM10 is identified as cattle, which includes bison (*Bison)*, aurochs (*Bos primigenius*), and yak (*Bos grunniens*). The geographical location and age narrowed this down to either bison or aurochs (Mol et al., 2006). Even though exceptions are possible, such as three Danish brown bear points (Jensen et al., 2020) and two Doggerland human bone points (Dekker et al., 2021), blanks for bone tools were generally derived from herbivores hunted and brought to the sites (Knecht, 1997).

Discussion

Comparisons

The two barbed points dated with ^{14}C-AMS yielded Early Mesolithic and Upper Palaeolithic dates. The Mesolithic age of NSM1 falls in the range of other dates available for Doggerland points (~10,000-7000 BP) (Dekker et al., 2021), while NSM18 is the oldest barbed point from the Dutch coast with an age of approximately 13000 years. Only one other barbed point from Doggerland, off the coast of Great Britain, the Colinda point, dates from the Late Palaeolithic. Direct AMS dating of this specimen yielded an age of 13,500 cal BP (Bailey et al., 2020).

The Upper Palaeolithic point in our sample matches the Magdalenian barbed points of the Iberian Mediterranean. Compared to contemporaneous French and Cantabrian barbed tips, Mediterranean ones usually have a single row of small barbs that do not protrude much from the shaft, although a certain degree of variability within the assemblages is visible (Román & Villaverde, 2012). The same features characterise both our Upper Palaeolithic and Mesolithic points.

The Mesolithic points of Doggerland compare well in terms of overall morphology, size, and shape of the barbs, with other western European assemblages (Maglemosian tradition) of similar age (e.g., Star Carr, Clark, 1936; Friesack, Gramsch, 2000), except for their reduced length. Despite the high internal variability, which characterises all bone point assemblages, these points show a preference for a unilateral row of small barbs, the absence of distinct bases, and very simple or absent decorations. Base incisions, with an aesthetic and/or functional meaning, are documented on barbed points from Star Carr (Clark, 1954; Elliott & Little, 2018) and the Colinda point (Allain & Rigaud, 1986) but are absent in the Doggerland assemblage. Bilateral barbed points, which are represented by one fragmented specimen in the Doggerland assemblage (Verhart, 1988), have a more north-eastern European distribution and they likely have roots in the French and Cantabrian Magdalenian tradition (Bergsvik & David, 2015; Orłowska & Osipowicz, 2021). The Doggerland points fit well in a unilateral tradition of bone points that has its roots in some of the technocomplexes of the final Upper Palaeolithic.

The function of Dutch Mesolithic osseous points

Our results suggest that barbed and unbarbed points were different tool types with different functions. Use-wear traces indicate that barbed points (N=15) likely served as hunting weapons. Unbarbed points (N=2) were used to perforate animal hide. However, a study of a bigger sample is needed to check if this conclusion fits all unbarbed points, or if their function was more diverse.

Some of the studied small barbed points bear traces of contact with mammal bone and others with soft fish tissue, but the size of the prey is unknown. The use of bone points on small mammals like beavers and otters is reported in numerous historical and ethnographic sources (Christensen et al., 2016; Hartz et al., 2019; Russell, 1992). Direct archaeological evidence for the use of barbed points to hunt beavers comes from the Middle Neolithic layer of Sakhtysh 1, Central Russia, where a fragment of an osseous point was found stuck in a beaver skull (Zhilin & Savchenko, 2020). The direct association of barbed points with elk remains confirms their use for hunting large size ungulates as well (Clark, 1936; Hallam et al., 1973). Ethnographic evidence certifies the use of barbed points for fishing (Christensen et al., 2016) while, for archaeology, this link was often suggested based on indirect associations of osseous points and fish bones (e.g., Cristiani & Borić, 2016; Nielsen, 2009). The microwear traces presented here provide clues supporting the theory of small barbed points being fishing gear as previously proposed (Verhart, 2000).

These results reinforce the data on Doggerland environment and the presumed diet of Mesolithic human groups who inhabited this area. At the beginning of the Holocene, the southern North Sea was a rich and diverse landscape characterised by a forested environment with interspersed lakes and marshes (Amkreutz et al., 2018; Peeters & Momber, 2014). This environment provided a wide range of food resources including medium to large ungulates (e.g., red deer, roe deer, aurochs, elk, wild boars), beaver, otter, fish, shellfish, and birds (Kitagawa et al., 2018; Mol et al., 2006; Van der Plicht & Kuitems, 2022). Stable isotope analysis of Mesolithic skeletal remains from Doggerland showed a significant freshwater component of their diet, highlighting the importance of aquatic resources (Van der Plicht et al., 2016). Barbed points were an important part of the hunting equipment and probably complemented the fishing toolkit alongside hooks. Other methods, such as nets and fish traps, which would have yielded a greater number of fish, were likely employed as well. Fish traps from the Netherlands date to the Neolithic (Out, 2008), but Mesolithic examples of fish traps, nets, and sinkers are well documented in northern and eastern Europe (McQuade & O'Donnell, 2007; Zhilin & Savchenko, 2020). From around 7,000 years BP, due to the rapid sea level increase, the Doggerland area transformed from an inland to a semi-marine and then a fully marine environment (Cohen et al., 2014). Isotope analysis showed that marine resources were also exploited by later Mesolithic hunter-gatherers, although less intensively (Van der Plicht et al., 2016). A different environment with different prey may have required different hunting tools.

Points size and prey targeted

Both large and small barbed points studied here show macroscopic evidence of their use as projectiles. However, the investigation of micro-polishes on large points in our sample was inconclusive. Therefore, it is still unclear if larger points were designed for different hunting activities or specific prey. More use-wear analysis and dating of points may help to identify different specific functions of barbed points or document a change in their function through time connected to changes in the environment. Such studies are underway in the project Resurfacing Doggerland by Dr. Hans Peeters (NWO AIB.19.009).

Previously it has been suggested that small points were arrow tips and the larger ones were spear tips based on size, weight, and shape of the barbs (Gramsch, 2000; Verhart, 2000). In addition, the small points would have been used on small prey and the larger tips on larger sized prey. Both ideas can be contested. Before making a connection between the size of the point and the prey, it would be necessary to conduct a functional study to separate proper projectile points from pointed bone tools used in activities other than hunting. As our analysis has demonstrated not all the pointed tools are projectiles (cf. Osipowicz et al., 2020b), although they are often grouped under this label (e.g., Amkreutz & Spithoven, 2019; Gramsch, 2000). Also, since barbed points are often reworked, the morphology of the barbs we see may result from the practical constraints of resharpening and repairing the object, such as the size or shape of the blank.

If we accept that small barbed points from the Dutch North Sea are unique in terms of their size among the European scenario (Amkreutz & Spithoven, 2019), then we cannot assess their function based on comparisons with other assemblages which are predominantly featuring large points. Many European bone point assemblages are found with ichthyofauna, often pike, but their association is not proven. Because these assemblages do not contain small points, we cannot deduce if there are prey differences based on the point type. Besides, direct evidence of large barbed points associated with large mammal bones, like the example of elk from High Furlong, is too scarce to suggest a strong connection between different sized points and the size of the prey targeted. Furthermore, ethnographic examples highlight the variability of shapes and sizes of points used as arrowheads and spearheads (e.g., Mason, 2007; Osgood, 1970). Therefore, a typological and functional interpretation based only on the size of the points is not reliable (cf. Hartz et al., 2019; Pétillon, 2009; Weniger, 1992).

Reconstruction of hafting methods

The location and characteristics of wear traces and residues, together with the experimental results and chemical analysis, provide clues as to how the barbed points studied here were hafted. The tools were attached to their shafts with birch tar and animal and vegetal bindings. The location of the residue on some points on one side of the tool indicates the use of a bevelled shaft, while residues on both sides indicate a split shaft. The location of binding impressions on both lateral edges may point toward either a split or bevelled shaft. There is no clear indication of the preference for the bevelled shaft over the split one. An exception is NSM26, which displays binding traces encircling the base. According to Verhart (1988, p. 183), this point may have been entwined to create a better fit into the shaft. We hypothesise that this point was reused and re-hafted several times on a bevelled shaft allowing traces to form on both sides. Moreover, based on the location of binding impressions on the meso-proximal area of the tools, we conclude that these points were not detachable. Impressions left from a harpoon line would have been limited to the mesial part of the implements (cf. Cristiani & Borić, 2016). We identified no 'classical' harpoon points with a detachable head and a line (category A in Pétillon, 2009). None of the analysed implements display features for fastening a line, such as the presence of a basal perforation (linehole), lateral spurs at the base, or the binding barb. In the total North Sea assemblage (N=~1,000) only two points classify as possible harpoons (Amkreutz & Spithoven, 2019). Both are large points and display a single or double notch at the proximal base where the line may have been attached (Verhart, 1988, Fig. 8 MS133 and Fig. 9 KF41). However, these points may have been firmly attached to a foreshaft and therefore still be part of a composite detachable weapon system. North American Shuawps' beaver harpoons, for instance, are composed of an osseous barbed point attached to a wooden foreshaft (Ignace & Ignace, 2017). The morphology of these harpoon points, however, does not differ from other fixed points (Pétillon, 2009). Therefore, it is almost impossible to identify their detachable nature if recovered without the shafts.

Micro-polishes on the meso-proximal area of some of the studied tools provide evidence of the binding materials used to secure the points. In some cases, sinew is identified while some of the other binding polishes are plant related. Although our experiments indicate that sinew is a stronger binding material and led to a lower failure rate during the shooting experiments, vegetal fibres are well documented in archaeology (e.g., Elliott & Little, 2018; Gramsch, 2000; Junkmanns et al., 2019). We also cannot completely rule out that the failure of the hafting bond was intentional. Failure would have allowed the point to detach upon impact and rankle in the wound causing more internal damage (cf. Mason, 2007).

Birch tar was used as a single-component adhesive. The use of birch tar as an adhesive for tools used for fishing is not surprising considering its material properties. Birch tar is not water-soluble and can be reheated and reused many times with almost no detrimental effects on its performance (Kozowyk & Poulis, 2019). Evidence of pure birch bark tar adhesive or a mixture of birch tar and pine resins is well-known in the European Palaeolithic and Mesolithic respectively (see Little et al., 2022 and references therein). However, the majority of points studied here did not have adhesive residues. Although this may be due to a preservation bias, it is conceivable that hafting methods of barbed points did not always necessitate adhesives. Direct evidence from the Mesolithic sites of Friesack (Germany) (Gramsch, 2000) and Ulkestrup (Denmark) (Andersen et al., 1982) shows that barbed points were not always mounted with adhesives. At Friesack, some points were bound to the shaft with strips of bast without glue. Others were hafted with tar and a combination of tar and bindings (Gramsch, 2000). In addition to this, in many examples, indigenous peoples of North America do not use adhesives to bond the points to the shafts but only sinew (Mason, 2007). This evidence may explain the low number of adhesive residues compared to the relatively high occurrence of binding traces on the North Sea points. Another possible explanation is that North Sea points were mounted with bindings and tar used only as a coating agent as seen, for instance, at Friesack (Gramsch, 2000) and Krzyż Wielkopolski 7, Poland (Kabaciński et al., 2023). Our experiments show that minimal residues preserve on the points when this arrangement is used. Moreover, when used as fishing gear, the bindings likely required some adhesive or sealant to waterproof them.

The long life of barbed points: reuse, rejuvenation, re-hafting

Reuse and rejuvenation of barbed points seem common technological behaviours. Our sample of osseous points shows traces of maintenance (rejuvenation, reuse, and reworking). Other studies also documented a large number of rejuvenated and reworked Dutch points (Spithoven, 2018; Verhart, 1988). Besides rejuvenated tips and barbs, the Doggerland assemblage features fragments of large points that broke and were roughly refurbished into an equivalent tool. The old barbs were ground away, leaving visible scars on the side, while new barbs were cut near the tip (Fig. 5.1, NSM8; Verhart, 1988, Fig. 14 KF69). Maintenance traces are common on other contemporaneous Upper Palaeolithic and Mesolithic barbed osseous points as well (Langley, 2016). Experimental work (Knecht, 1997; Pétillon et al., 2011) showed that bone points are more durable and are easier to repair when dull or broken compared to their lithic counterparts.

This evidence highlights that during their use-life, Doggerland barbed points were intensively curated, reused, and re-hafted many times before being discarded or lost. The bone material accommodated this intense reuse, but material selection, e.g., human and brown bear bones (Dekker et al., 2021), may also suggest that these points were imbued with specific cultural and symbolic connotations. The points may also have had special meaning and were therefore used for a very long time by their Mesolithic owners.

Conclusion

We presented the results of a detailed functional analysis on a sample of 17 barbed and unbarbed points recovered from the beaches of the Dutch North Sea. We reconstructed their use-lives and assessed the animals hunted.

Our sample features the oldest barbed point recovered from the Dutch North Sea, roughly 13,000 years old. The other point dates to the Early Mesolithic, roughly 10,500 years ago. Morphological similarities, dominance of unilateral barbs, small teeth, and general lack of decoration, between Doggerland and European bone points of the Upper Palaeolithic and Mesolithic periods indicate that these tools were part of a shared European tradition and a systematic component of the hunting kit since at least the end of the Palaeolithic. Macrofractures visible on the tips and bases of barbed points indicate their use as projectiles. We also show that on some small points, the polish closely resembles experimental fish polish, and on others, the polish resembles (mammal) bone polish. We suggest that barbed points were used for hunting both aquatic and terrestrial animals. Prey targeted may have included freshwater fish, ungulates, and animals hunted for fur, such as beavers or otters. Evidence of rejuvenation of small and large points, reuse of large barbed point fragments, and re-hafting underline that these were highly curated tools, and their use-life was extended as long as possible. The bone facilitates extensive reuse, and perhaps the use-lives of the points were extended because they bore a special meaning to the owners. Conversely, unbarbed points do not display impact fractures. The presence at the tip of striations oriented transversely to the axis of the tool indicates their use in boring/piercing activities, likely to perforate hides.

The characteristics of macro and micro hafting traces and chemical analysis help to reconstruct the hafting methods of bone points. Split and bevelled systems were used in combination with birch tar adhesive and sinew and vegetal bindings. Our experiment showed that sinew binding works better than vegetal ones and with tar can create an excellent joint. Considering the large number of points with binding traces and no adhesive residues, bindings may also have been used alone to secure hafts.

Our results highlight that barbed points were dynamic tools that transformed during their life through use, repair, and reuse. The situation may also be similar for unbarbed points. Such transformations may have led large points to become small ones, possibly with a consequent change in their function. Analogies show that both small and large points served multiple different purposes and were used on various prey. Therefore, a functional distinction of barbed weapons between arrow and spear tips based only on morphometrics is not sufficient to account for the complexity and variability of archaeological assemblages. Only by combining a functional approach encompassing use-wear analysis, ethnographic analogies, and problem-oriented experiments, can we concretely demonstrate the precise function of barbed osseous projectile points.

Acknowledgments

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Chapter 6

Discussion and conclusion

Discussion

This thesis has expanded the sample of securely identified prehistoric adhesives. By using a multi-analytical approach to the study of ancient adhesives from three different archaeological assemblages, I shed light on tool use, raw material selection, and adhesive recipes to compare the technological complexity and behaviour of Neanderthals and modern humans. In this chapter, I outline my results in response to my research questions, as outlined in Chapter 1, reflect on the methodology employed, and provide new insights to further discuss adhesive technology and the technological complexity of Neanderthals and modern humans.

Summary of Neanderthal and modern human adhesives

This dissertation contributes to the body of knowledge on Neanderthal and modern human adhesives (Table 6.1).

Table 6.1: Overview of the archaeological assemblages analysed in this dissertation and identified adhesives. Ka (kilo annum, thousand years) signifies "thousand calendar years ago".

Site	Screened artefacts	Identified adhesives and method	Tool type	Adhesive ingredients	Additives	Age
Morín Cave (Spain)	23,796	5: -3 SEM-EDX and Raman -2 GC-MS	-1endscraper -1 retouched bladelet -3 lithic points	-Cupressaceous resin or tar -Plant wax/ insect wax/ bitumen	Hematite Fe_2O_3	~43-23 ka BP
Dutch North Sea coast (The Netherlands)	17	3: -3 GC-MS	-3 barbed points	Birch bark tar	/	~13-10 ka BP
Steenbokfontein Cave (South Africa)	30	11: -5 FTIR -2 FTIR and GC-MS -4 GC-MS	11: -5 scrapers -6 cutting tools	-*Widdringtonia* or *Podocarpus* sp. resin/tar -Plant exudate containing pentacyclic terpenoids	Hematite Fe_2O_3	~5-2 ka BP

Evidence of the use of adhesive likely attributed to Neanderthals comes from Morín Cave, Spain (Chapter 4). At this site, I documented adhesive residues on at least one lithic point attributed to the Châtelperronian. The Châtelperronian is a regional techno-complex that, in Iberia, spans between 43,500-39,200 years BP (Rios-Garaizar et al., 2022). It is traditionally associated with Neanderthals

(Welker et al., 2016), despite some authors raising doubts about the Neanderthals' authorship of this industry (Teyssandier, 2024). At Morín, Neanderthals used plant extractives to haft the lithic point which displays evidence suggesting its use as a projectile. A second Châtelperronian point with orange-brownish residues of organic origin possibly related to adhesive, shows hematite mixed within the residue. Thanks to precise chemical methods for adhesive identification, it has already been demonstrated that Neanderthals exploited a range of natural resources for adhesive manufacture, including birch bark tar, bitumen, and conifer resin (Boëda et al., 2008b; Degano et al., 2019; Niekus et al., 2019; Schmidt et al., 2023). However, the use of hematite as an additive in adhesives is not well documented, and this is one of the first examples of its use for this purpose by Neanderthals. This finding is supported by the recent discovery of a goethite-loaded bitumen adhesive at Le Moustier (France), dating between 56,000-40,000 years BP (Schmidt et al., 2024b) However, no use-wear analysis was conducted on the Le Moustier tools, so their function is unknown.

All three assemblages analysed for this dissertation yielded evidence for the use of adhesives by modern humans (Table 6.1). Regarding the European sites, at Morín, I identified possible adhesive remains on one Protoaurignacian endscraper (>36,600 years BP), one Early Aurignacian Dufour bladelet (>36,600 years BP), and one Gravettian partially backed blade (~23,500 years BP). Raman results likely indicate a tree resin or tar, as the source of the Protoaurignacian and Early Aurignacian adhesives. The molecular signature of the Gravettian sample could be attributed to a variety of sources, such as plant or animal wax and bitumen. Hematite, identified with SEM-EDX analysis, was consistently mixed with the adhesive residues throughout the sequence. Based on the use-wear traces preserved on the tools, the endscraper was used to work hide, while the two backed implements may have been used as projectiles (Chapter 4). The three residue samples from the North Sea bone points (~13,000-10,200 years BP) were chemically identified as birch bark tar. In our samples, birch tar was used as a single-component adhesive in combination with animal and vegetal bindings. Bone points were likely used as projectile tips for terrestrial and aquatic hunting (Chapter 5). Regarding Steenbokfontein Cave in South Africa, I morphologically identified adhesive residues on 28 tools (between ~5250-2200 years BP), of which 11 were securely identified with molecular analyses (IR spectroscopy and/or GC-MS). *Widdringtonia* or *Podocarpus* resin/tar was mixed with hematite and occasionally with an additional tree extractive. Tools were used as hafted scrapers for hide-working and as elements of composite tools for cutting animal and plant matter, although other functions, such as use as barb-projections, cannot be excluded. I did not observe any differences in the adhesive recipes used to haft tools made of different raw materials or used in different tasks (Chapter 3).

Comparing Neanderthal and modern human adhesive traditions

By integrating use-wear with elemental and molecular data from the residues, as well as paleoenvironmental data from the literature, I can reflect and compare Neanderthal and modern human raw material exploitation, the use of additives, and the context of use of hafted tools in a new light. However, to fully evaluate these aspects and determine the extent to which adhesive technology was integrated into the toolkits of Neanderthals and modern humans, it is necessary to consider the data presented in this dissertation from a broader perspective and integrate it with data from the available literature.

Timing of adhesive technology: To date, the oldest adhesive residues from Middle Palaeolithic contexts are known from Campitello (Italy), dating to approximately 190,000 years BP (Mazza et al., 2006). Possible adhesive remains on a large sample of stone tools were identified by optical microscopy and SEM-EDX at Inden-Altdorf (Germany), dating approximately 120,000 years BP (Pawlik & Thissen, 2011), although they have never been molecularly identified. More securely identified Neanderthal adhesives are known for later phases of the Middle Palaeolithic; around 70,000 years BP in the Levant (Boëda et al., 2008a; Boëda et al., 2008b) and 55-45,000 years BP in Europe (Degano et al., 2019; Niekus et al., 2019; Schmidt et al., 2024b), including the evidence from Morín Cave reported in this dissertation (Chapter 4).The early evidence of Neanderthal adhesive technology is scattered in time and space, leading to the idea that, at an initial stage, Neanderthals may have possibly accidentally discovered, lost, and rediscovered adhesive technology (Kozowyk et al., 2017). Only later Neanderthals refined their adhesive technology, allowing adhesives to be maintained and transmitted (Kozowyk et al., 2017; Schmidt et al., 2023). That may account for the significant temporal gap between the very few older adhesive remains identified and the higher number of more recent occurrences

(Fig. 6.1). However, preservation and research biases can also account for this. Therefore, only by increasing the number of securely identified residues from well-dated Middle Palaeolithic contexts we can accurately gauge the level of embedment of adhesive technology in the toolkit of Neanderthals. Adhesive residues from initial Upper Palaeolithic contexts in Europe are also rare. The examples from Morín Cave (Chapter 4) and Grotta del Cavallo (Italy) (Sano et al., 2019) are among the oldest.

In South Africa, securely identified Middle Stone Age adhesives were found only at Diepkloof Rock Shelter (Charrié-Duhaut et al., 2013) and Sibudu Cave (Prinsloo et al., 2023; Soriano et al., 2015). More were only morphologically identified. Based on the characteristics, location, and distribution of these residues on the tools, they were convincingly linked to hafting adhesives (e.g., Gibson et al., 2004; Lombard, 2004; Lombard, 2007). These first finds occur almost simultaneously with the earliest evidence of mechanically delivered projectiles, dated around 54,000 years BP in Europe (Sano et al., 2019) and in Africa around 64,000 years BP (Brown et al., 2012). For bow hunting, hafting is essential; therefore, it is likely that projectile technology was accompanied by a well-developed hafting technology, which potentially included adhesives (cf. Lombard & Phillipson, 2010).

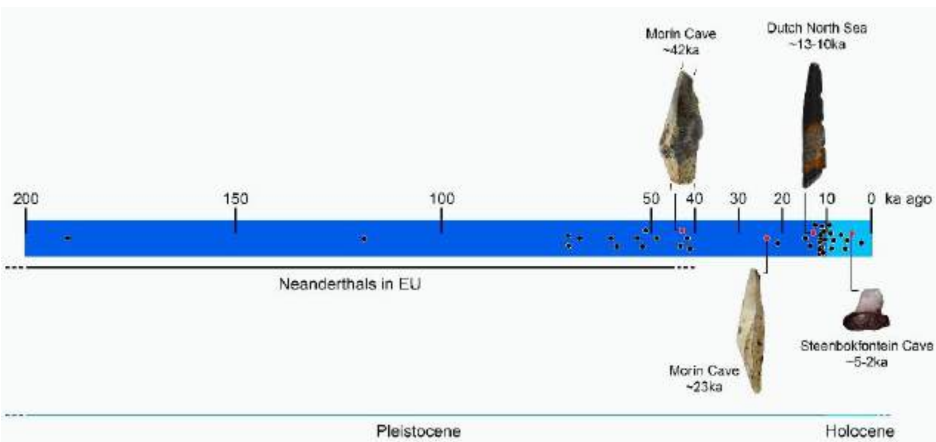

Figure 6.1: Timeline displaying the chronological distribution of molecularly identified adhesives cited in this dissertation. In red are the finds analysed by the author. Ka (kilo annum, thousand years) signifies "thousand calendar years ago".

The presence of adhesive remains intensified in the subsequent periods and reached its peak in the early Holocene, as demonstrated by adhesive finds both in Mesolithic sites (see Chapter 5) in Europe and Later Stone Age sites (see Chapter 3) in South Africa (Fig. 6.1). This trend corresponds with another cultural universal phenomenon: lithic miniaturisation (Pargeter & Shea, 2019). Assemblages containing a large number of microliths predominantly date to the final Pleistocene and early Holocene, but some early examples are found in Middle Stone Age industries of South Africa (Brown et al., 2012) and Protoaurignacian industries in Europe (Kuhn, 2002). Microliths (i.e., bladelets, backed bladelets, segments, small retouched tools) were predominantly used as cutting and piercing tools, often as components of composite tools in domestic activities or advanced projectile weapons (Chapter 3 (Charrié-Duhaut et al., 2016; Groman-Yaroslavski et al., 2020; Porraz et al., 2016)). As a result, adhesives were arguably a fundamental component of modern humans' hafting technology.

<u>Selection of ingredients of the adhesives:</u> At Morín Cave (Spain), only two out of five samples with possible adhesive residues provided information on the adhesive sources since the results of the others yielded only broad characterisations. Neanderthals likely used a plant extractive from the genus *Juniperus* as the main component of their adhesives, while modern humans used an unidentified plant or animal wax or bitumen (Chapter 4). Conifers were the most locally available plants during the Middle to Upper Palaeolithic transitions in Cantabria (Allué et al., 2018; Fernández-García et al., 2023; Ochando et al., 2022), while bitumen sources are located within a 30 km radius of Morín (Kruge & Suárez-Ruiz, 1991). Similarly, modern humans at Steenbokfontein Cave (South Africa) employed a tree extractive from *Podocarpus* or *Widdringtonia* as the primary source of adhesive (Chapter 3). Specimens of both genera today grow within a 40 km radius of the cave (Lombard, 2023); additionally, *Podocarpus* sp. charcoals were found in archaeological sites within ~50 km of Steenbokfontein (Cartwright et al., 2016; Cartwright, 2013), corroborating the availability of this plant in the environment surrounding the site. This data seems to suggest that the main driver of the selection of natural resources for adhesive production for both Neanderthals and modern humans was their availability and abundance in the local environment. The case of the bone points from the Dutch North Sea stands out. During the early Holocene, between the Pre-Boreal and Boreal, the environment of Doggerland was dominated by birch and pine (Gaffney et al., 2009). Although pine resin can make a suitable adhesive with minimal technological investment,

it seems that Doggerland hunter-gatherers preferentially selected birch to produce tar over readily available pine resin. All the samples that were analysed from the North Sea bone points are securely identified as birch tar (Chapter 5). Moreover, in the Mesolithic, the overall available molecular data point toward the almost dominant use of birch tar as an adhesive for hafting purposes (Aveling & Heron, 1998; Kabaciński et al., 2023; Little et al., 2022; Osipowicz et al., 2020a), even in locations where pine was more abundant than birch, such as at Friesack (Benecke et al., 2016). While this can be due to the low number of molecularly identified adhesive remains or the better preservation of terpenoids over other organic molecules, it is likely that birch tar may have been preferred for its superior material properties, i.e., cohesive strength, workability, and reusability (Kozowyk & Poulis, 2019).

Use of additives: Until recently, securely identified Neanderthal adhesives consisted of single-component glues as opposed to multi-ingredient adhesive mixtures (or compound adhesives) produced by modern humans in Africa (e.g., Veall, 2019) and Europe (e.g., Bradtmöller et al., 2016; Javier Muñoz et al., 2023). Evidence from Morín Cave (Chapter 4) and Le Moustier (France) (Schmidt et al., 2024b), however, reveals the likely use of iron oxides as loading agents in adhesives by Neanderthal populations. Additional evidence from Fosellone Cave (Italy) suggests that Neanderthals possibly mixed conifer resin with beeswax (Degano et al., 2019). Thus, Neanderthals produced compound adhesives just like contemporaneous modern humans in Africa. Several studies have shown that additives have different effects on adhesive material properties and can be used to modify the characteristics of the desired final product (Kozowyk et al., 2016; Wadley, 2005; Zipkin et al., 2014) although the right ratio of additives is required to enhance the performance (Kozowyk et al., 2016). The ‘right mix’ was achieved by sensory judgment, either visual or tactile, as demonstrated for other ancient crafts (cf. Kuijpers, 2013; Kuijpers, 2018; Sahle, 2019).

Iron oxides (generally referred to as ‘ochre’) are common additives of South African adhesives, including at Steenbokfontein Cave (Chapter 3), but other identified additives include latex, beeswax, fat, bone fragments, quartz, and sand (Charrié-Duhaut et al., 2016; Degano et al., 2019; Veall, 2019; Villa et al., 2012). The fabrication of compound adhesives demands the engagement of sophisticated technical and cognitive abilities, encompassing pyrotechnology, abstraction, forward planning, and multitasking (Lombard, 2007; Wadley, 2010;

Wadley, 2013). That suggests that Neanderthals and modern human adhesive technologies are more similar than previously thought and Neanderthals likely possessed technological and cognitive skills matching those of modern humans.

Context of use and hafted tool types: The two potential Neanderthal adhesive finds featured in this dissertation have been found on lithic points from Morín, which have been likely used as projectile tips (Chapter 4). Other published studies have reported the identification of Neanderthal adhesives on points or convergent tools, mostly Levallois points and convergent scrapers (Boëda et al., 2008a; Doronicheva et al., 2022; Hauck et al., 2013). However, the range of tool types with adhesive residues also includes 'domestic tools', such as side-scrapers, retouched flakes, and unmodified flakes (e.g., Degano et al., 2019; Mazza et al., 2006; Niekus et al., 2019). Unfortunately, in most cases, a detailed use-wear study of these tools is missing; therefore, the exact function of these objects is unknown (for an exception, Doronicheva et al., 2022). Furthermore, some of the oldest evidence of adhesive use was not intended to attach stone tools to handles but to serve as handles themselves. This is the case for the Zandmotor find (Niekus et al., 2019), one of the Königsaue tar lumps (Koller et al., 2001), Levallois flakes from Syria (Monnier et al., 2013), and possibly the tools from Le Moustier (Schmidt et al., 2024b). When the adhesive is used as a handle, it is possible that other properties, such as workability, mouldability, gripping, and handling, were preferred to pure adhesive strength, as the evidence of Le Moustier seems to suggest (Schmidt et al., 2024b). Once again, this highlights the level of knowledge of material properties and fillers of Neanderthals and the nuanced role of adhesives in prehistoric technologies.

Upper Palaeolithic and Mesolithic European adhesives from Morín Cave (Chapter 4) and on the bone points from Doggerland (Chapter 5) have been identified mainly on backed implements (N=2) or projectile points (N=3) and only on one endscraper. Other published studies have reported the identification of modern human adhesives on tools used as mechanically delivered projectiles or typologically classified as such (Aleo et al., 2023; Baales et al., 2017; Bradtmöller et al., 2016; Javier Muñoz et al., 2023; Langejans et al., 2023) and, to a lesser extent, on domestic tools (Bradtmöller et al., 2016; Cârciumaru et al., 2012). In South Africa, the older morphologically and chemically identified residues are also found on points and backed tools (Charrié-Duhaut et al., 2013; Lombard, 2005; Lombard et al., 2010; Soriano et al., 2015). However, more adhesive residues on other tool types, including microliths, scrapers, adzes, handles, and an ostrich eggshell flask, are known from later periods (Charrié-Duhaut et al., 2016; Jerardino, 2001; Veall, 2019). At Steenbokfontein Cave, most of the tools with adhesive residues are typologically classified as scrapers, and many of those that I analysed for use-wear functioned as hide-working and plant-working tools (Chapter 3). Considering the tools featured in this dissertation and other known tools with molecularly identified adhesives, Neanderthals and modern humans have hafted informal and formal tools related to domestic and hunting spheres. The over-representation of adhesive residues on projectiles, i.e., points and microliths, from the Upper Palaeolithic onwards is likely partially due to a research bias. While projectiles and microliths do necessitate hafting for their use - thus, it is more likely they bear adhesive residues - hunting tools have always attracted more scientific attention than domestic ones (cf. Taipale & Rots, 2021). When the latter are subjected to functional studies, they often display hafting traces, such as micro-polishes, edge damage, and even adhesive residues (e.g., Aleo et al., 2021; Taipale & Rots, 2020). Hence, future research devoted to domestic tools will likely lead to the identification of more adhesive residues and overcome historical research biases. Additionally, many objects typologically classified as projectiles may not have been used as such (e.g., Groman-Yaroslavski et al., 2020). A telling example is the unbarbed points from the Dutch North Sea presented in Chapter 4 that were used as perforators and not as projectiles. Therefore, without reliable data from use-wear analysis, it is impossible to confirm their functions and thus support a preference of early modern humans for hafting hunting tools over domestic ones.

Implications

Before discussing adhesives as indicators for technological complexity in the deep past, I want to reflect on some aspects of adhesive technology highlighted so far.

The information in this dissertation and the data available in the published literature indicate that the number of securely identified Neanderthal and early Homo sapiens adhesives is increasing. The available evidence suggests that adhesives were used in all spheres, domestic and hunting. The presence of adhesives on domestic tools used for daily tasks provides evidence of routine production of adhesives. Evidence from Campitello (Mazza et al., 2006), Zandmotor (Niekus et al., 2019), and Fosellone (Degano et al., 2019), demonstrates that Neanderthals often hafted unretouched simple flakes, obtained with limited technological investment, alongside scrapers, retouched blades, and points (e.g., Boëda et al., 2008b; Degano et al., 2019; Schmidt et al., 2024b). The same holds true for modern human tools hafted with adhesives (e.g., Bradtmöller et al., 2016; Charrié-Duhaut et al., 2016; Veall, 2022). Even though a detailed functional analysis of tools with adhesive residues was not always carried out, the majority were likely used for domestic activities. While some tasks do not necessarily require hafting, the technological investment required for hafting both informal and formal tools, whether domestic or hunting related, strongly suggests that Neanderthals and modern humans had mastered adhesive technology and integrated it into their toolkits.

The selection of ingredients for adhesive production was mainly driven by the availability of natural resources in the environment. However, the case of the bone points from the Dutch North Sea coast (Chapter 5) and other adhesive finds from the Mesolithic period highlight that material properties also play a crucial role in the selection. Therefore, these prehistoric adhesive makers were aware of the different properties of the natural resources in their environment and likely selected one over the other according to their needs. The same holds true for the selection of organic and mineral additives. Among mineral fillers, hematite-rich ones have the most impact on the workability, viscoelastic properties, and hydrophobicity of resin and gum (Kozowyk et al., in preparation). Prehistoric populations, although unable to quantify these properties, likely had a sense of it and may have purposely selected hematite-rich ochres (Dayet et al., 2013; Hodgskiss & Wadley, 2017; Kozowyk et al., in preparation).

Concerning technological and, by extension, cognitive implications, evidence points toward the technological complexity of prehistory adhesives. As stated in the introduction, complex technologies are often multi-stepped or structured hierarchically, involve the transformation of materials, and require enlarged functions of the brain and some forms of cultural transmission (Hoffecker & Hoffecker, 2017; Lombard, 2019; Schmidt, 2021; Wadley et al., 2009). Adhesive production, and overall, the production of multi-component tools, involves planning trips to collect the different raw materials that must be gathered and transported to the production site. It can be, therefore, more costly and time-consuming than other prehistoric technologies. This is also an indication of forward planning (Wadley, 2010). Although adhesives can be produced accidentally and with relatively simple processes (Schmidt et al., 2019), it is undeniable that the intentional transformation of bark into tar to make a relatively large quantity of adhesive or compound adhesive manufacture required invention, innovation, and other enhanced executive brain functions (Niekus et al., 2019; Schmidt et al., 2022; Wadley, 2010; Wadley et al., 2009). Adhesive production is organised in steps, and each ingredient must be prepared and added at the right time and quantity, as demonstrated by experiments and ethnographic accounts (Sahle, 2019; Wadley, 2010; Wadley et al., 2015). These steps necessitate forward planning, abstract thought, and mental fluidity (Wadley, 2010; Wadley et al., 2009). Additionally, it has been demonstrated that even simple tar-making methods can exhibit elaborate production sequences if the production is scaled up involving concurrent production assemblies, thus multi-tasking and attention-switching (Kozowyk et al., 2023b). Moreover, some production methods require some form of social learning and knowledge transmission (Schmidt et al., 2023; Schmidt et al., 2022), a criterion used to define complex technologies (Schmidt, 2021; van Schaik et al., 2019). Therefore, adhesive technology can be qualified as a complex technology able to shed light on technological and cognitive capabilities in the deep past.

Cultural biography of prehistoric adhesives

In the following biography, I attempt to outline some aspects of the cultural biography of prehistoric adhesives analysed in this thesis. The adhesives were produced from natural resources selected for their availability in the environment or material properties. Despite being difficult to demonstrate, specific materials may also have been chosen for other qualities, such as colour, smell, and translucency (cf. Hess & Riede, 2021; Little et al., 2022; Peresani et al., 2021). Evidence of prehistoric production features has not yet been identified; however, there is consensus around the complexity of certain adhesive production processes. This is the case for those involving concurrent production assemblies (Kozowyk et al., 2023b) or relying on underground processes potentially involving some forms of social learning or teaching (Schmidt et al., 2024b; Schmidt et al., 2022). The growing complexity of artefacts, involving hafted tools made of several (organic) parts joined together with a medium, may have favoured informal craft specialization (Barham, 2013) linked to teaching and learning across generations (Barham, 2013). Ethnographic studies of adhesive production systems in traditional societies in Zambia and Ethiopia confirmed that the makers learned how to make adhesives from their parents or mentors at an early age, and the knowledge is transmitted through generations (Fajardo et al., 2024; Sahle, 2019). Nonetheless, it is unlikely to only talk about the specialisation or specialism of adhesive technology in the Palaeolithic. On the contrary, it is reasonable that small-scale adhesive production for simple repairs or hafting and backing of daily-use tools that required limited skills was embedded in the domestic economy, similar to flint knapping.

Among the materials used for making adhesives, ochre deserves a special mention. Ochre was mixed into the adhesive mixture at Steenbokfontein (Chapter 3) and possibly Morín Cave (Chapter 4) and it is a common ingredient in South African adhesives (Lombard, 2007). Several experimental studies demonstrated the functional role of ochre in the adhesive formulation (Kozowyk et al., 2016; Wadley, 2010; Zipkin et al., 2014). Ochre increases adhesive strength, improves workability, and reduces the curing time of resin-based adhesives. However, its addition may not have been merely utilitarian (cf. Dapschauskas et al., 2022; Zipkin et al., 2014), as there are many archaeological and ethnographic accounts for the symbolic and ritual use of ochre (e.g., Dapschauskas et al., 2022; Henshilwood et al., 2009; Hodgskiss, 2020; Rifkin, 2015; Watts et al., 2016). It is possible that initially ochre was added to the adhesive to confer symbolic or magical properties to an object and later spread due to its simultaneous beneficial effects on the adhesive's material properties. Whether the use of ochre in adhesive technology was perpetuated over time due to its symbolic or utilitarian function is hard to tell, but neither option should be discarded beforehand.

Throughout their lifespan, the analysed adhesives were used to affix stone tools to shafts, and their tools displayed evidence of use which indicates they were functional objects. I did not observe any change in the function of the studied tools or residues. Yet, this biographical approach highlighted the long use-life of some of these hafted objects, particularly scrapers from Steenbokfontein Cave (Chapter 3) and the Dutch bone points (Chapter 5). These tools display evidence of resharpening, reuse, or re-shaping. While endscrapers can be resharpened when still hafted (cf. Rots, 2010), I observed on the bone points wear traces suggesting re-hafting. In this scenario, the adhesive was likely re-melted and reused. An indication of that may be seen in NSM18, a fragment of a large point used and reused many times. The chemical analysis of its residue showed a high degradation of biomarkers, which may result from multiple re-heating sessions.

In support of this argument, experimental work showed that birch tar can be reheated many times with almost no detrimental effects on its performance (Kozowyk & Poulis, 2019). Although birch tar in small quantities can be easily produced (Kozowyk et al., 2017; Schmidt et al., 2019), its reuse is far less costly (considering time, energy, and resources) than sourcing materials to manufacture a new adhesive. Adhesives may have facilitated the reuse of objects. Some adhesives are thermoplastics and others are water-soluble, meaning they can be reheated or wetted to allow the insets to be easily removed from the shaft for reshaping, repairing, or resharpening. Additionally, the reversibility of the adhesives can extend the durability of the handle and haft over time (Barham, 2013). Since manufacturing hafted tools requires significant investment in time and effort, their owners extended their use-life as much as possible until they became no longer usable or were accidentally lost.

Reflections on the methodology and future directions

My research was devoted to the material analysis of tools and their residues through optical microscopy, as well as the integration of data derived from elemental and molecular analyses to reconstruct the use-life of hafted tools. Despite the considerable advancement of research in the field of prehistoric adhesives over the last two decades, there are methodological aspects that can still be improved.

First of all, I would like to emphasise the importance of analysing large samples of tools rather than single finds. The analysis of single finds within an assemblage hampers the recognition of patterns in the use of adhesives and hafted tools, variability or homogeneity of adhesive mixtures, and possible continuity of adhesive traditions. This limits our understanding of past agents' technological complexity in terms of the selection of task-oriented adhesives, exploitation of natural resources, and knowledge of the material properties of different ingredients. On a larger scale, this also affects the recognition of chronological or diachronic trends in adhesive technology.

Moreover, it is fundamental to use a comprehensive analytical approach to study ancient adhesives, which systematically integrates traceology (use-wear and experiments) with elemental and molecular analyses. Traceology can provide unique insights into adhesive technology and the technological and behavioural choices of past agents in several ways. Firstly, the distribution and location of use-wear inform us on different hafting designs and configurations. When assessing technological skills and know-how, it is important to know the joint design. The use of adhesives and binders to create composite tools requires the integration of materials with different properties and an understanding of cause and effect (Barham, 2013; Rots, 2010). In hafted tools, the joint represents the weakest part of the tools, and it is subjected to stresses and strains during use. Hence, the makers had to understand and incorporate this knowledge into tool design and select suitable materials, including binders and adhesives, to improve its effectiveness and durability (Barham, 2013). In this scenario, experiments also play a crucial role. The results of the experiment conducted with replicas of Mesolithic bone points in this thesis showed that different hafting arrangements can be recognised based on the distribution and characteristics of wear traces and residues (Chapter 5). More systematic and controlled experiments will establish a comprehensive reference for exploring hafting technology and composite tools design on archaeological material (cf. Rots, 2010)

Secondly, a detailed examination of the objects using a combination of low- and high-power microscopes allows for the interpretation of the tool's context of use and the evaluation of morphological features of the residues. The context of use helps us determine whether adhesives were task-specific and how and to what extent past agents modified the adhesive's properties to accommodate their needs. The morphological description of residues allows for their qualitative evaluation as potential adhesives to be selected for molecular analyses. As seen in Chapters 3 and 4, inferences on the nature of adhesives based solely on optical microscopy are often unreliable. The colour, shape, and distribution of residues can be altered through use, reuse, and taphonomic processes. Despite this, some morphological features of adhesives such as cracks on the surface of the residue, smoothness, greasiness, sharp and straight limits with angular terminations, and the presence of orangish semi-translucent inclusions or stains, appear to be commonly shared between different adhesive types and can guide researchers towards a broader identification of unknown residues as possible adhesives.

Third, a comprehensive microscopic examination of adhesive residues can help identify organic and inorganic additives such as hematite, sand, burnt bones, charcoal and elements of the hafting system like micro-fibres related to bindings or shaft material, and select targeted protocols for their analysis. Despite GC-MS being the most common method to characterise archaeological adhesives (Langejans et al., 2022), its application without optical microscopy, XRD, Raman, or even μ-CT analyses provides only an incomplete picture of adhesives and the tools they adhere to. The former organic components can hardly be identified only through spectroscopy or GC-MS due to the broad characterization bands of the latter and weak/noisy spectra, while inorganic additives risk being ignored if specific methods, such as XRD, XRF, IR spectroscopy, are not employed for their identification.

Last, I discuss the role of experiments in archaeology specifically related to adhesive technology, since they have played a significant role in this field. The experiments I conducted during my PhD aimed to create a reference collection for use-wear traces. The experiment presented in Chapter 2 was a generalized experiment resulting in a reference collection for interpreting traces on non-flint tools from South Africa. The experiment with replicas of Mesolithic bone points allowed me to evaluate the performance of different hafting designs and materials (Chapter 5). In the latter, I controlled parameters such as the position of the bow, the distance between the bow and the target, and arrow speed using a shooting machine and a chronograph to ensure minimal variations and reproducibility of results. Similarly, other ballistic experiments have been conducted, informing about hafting design performance and impact performance of different adhesives (Iovita et al., 2014; Lombard & Pargeter, 2008; Pargeter et al., 2022; Wilson et al., 2021).

Researchers have been largely investigating methods for birch bark production and have come up with possible methods prehistoric populations may have used (Groom et al., 2015; Koch & Schmidt, 2022; Kozowyk et al., 2017; Schenck & Groom, 2016). In addition to that, there is a current trend in research to quantify the material properties of recreated prehistoric adhesives with laboratory tests (Gaillard et al., 2016; Kozowyk et al., 2016; Kozowyk et al., 2017; Tydgadt & Rots, 2022; Wilson et al., 2021; Zipkin et al., 2014). These experiments led to the interpretation that some materials, such as *Podocarpus* tar and birch bark tar, were preferred over other natural resources due to their superior mechanical properties (Kozowyk & Poulis, 2019; Schmidt et al., 2022). Furthermore, the tests demonstrated that adding mineral additives improves the performance of recreated resin and gum adhesives (Kozowyk et al., 2016; Zipkin et al., 2014). However, these mixtures have rarely been tested in actualistic experiments (for an exception see Wadley, 2005; Wadley et al., 2009).

Prehistoric makers were not aware of properties such as shear or impact strength, adhesive melting point, or glass transition temperature. They relied on their senses and practical use of adhesives for specific activities rather than formal measurements to judge the readiness and quality of their adhesives (Kuijpers, 2013; Sahle, 2019). Prehistoric populations were interested in more than just adhesion performance. An adhesive might be effective for particular activities or with a specific hafting configuration, even if it underperforms in laboratory tests. Brittle adhesives may have been preferred for specific hunting strategies, such as hunting with poisoned arrows, where the dislodgment of the weapon tip upon impact was sought (cf. Wadley, 2010). Thus, predictions regarding the preferential use or the selection of a specific adhesive over another based solely on laboratory performance tests should also be corroborated by actualistic experiments (Van Gijn, 2014).

Final remarks

The objective of this study was to evaluate the technological complexity between Neanderthals and modern humans through a comparative analysis of adhesives. By expanding the dataset of securely identified Pleistocene and Holocene adhesives and employing a comprehensive analytical approach, I have shown significant parallels between the adhesive technologies of the two groups. Nevertheless, it is acknowledged that additional data will help substantiate this conclusion.

The application of a multi-analytical approach, encompassing optical microscopy, experiments, spectroscopy, and chemical analysis, to the study of ancient adhesives allowed me to characterise the nature of the materials, recipes, tool design, and context of use. These components are crucial for understanding the technological complexity and variability in the Palaeolithic record. This research illustrates that by applying different methods to analyse larger samples of tools, we can overcome the limits of a single technique, strengthen our results, and achieve a more accurate reconstruction of adhesive technology.

Drawing from available evidence, adhesives were integrated into the toolkits of late Neanderthals and modern humans. Adhesives were sourced from diverse natural materials primarily selected for their availability in the surrounding environment or material properties. Growing evidence indicates that both human species modified the material properties of natural adhesives through the incorporation of additives to achieve products with desired attributes. These adhesives, whether single-component or multi-component, were utilized for hafting lithic and organic projectiles and for domestic tools. The identification of adhesives on a growing number of tools employed in domestic activities supports the proposition that the production and use of adhesives was likely a widespread practice among Neanderthals and modern humans. The similarities in the adhesive technologies of Neanderthals and modern humans highlighted in this thesis support the technological complexity of Neanderthals. Neanderthals' selection, transformation, and use of adhesive materials hint towards comparable procedures and reasoning with those of modern humans. On a higher level, these results likely imply that Neanderthals were capable of the same cognitive processes and had similar technological abilities to anatomically modern humans.

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Curriculum vitae

Alessandro Aleo was born in Casale Monferrato (AL), Italy on June 17th, 1989. He obtained a bachelor's degree in Cultural Heritage (archaeological curriculum) from the University of Turin, Italy in 2013. In 2016 Alessandro graduated *cum laude* with a Master of Art in Quaternary, prehistory, and archaeology from the University of Ferrara, Italy. His master's research, supervised by Prof. M. Peresani and Dr. R. Duches, focused on the study of early Upper Palaeolithic endscrapers from Fumane Cave. To continue his research, he was awarded a scholarship from the University of Ferrara to complete an internship in use-wear analysis at the Traceolab, University of Liège, under the supervision of Prof. V. Rots.

Between 2017 and 2019, Alessandro worked as a field archaeologist for different archaeological companies located in northern Italy. Starting at the end of 2019, he joined as a PhD candidate in the large ERC project "Ancient Adhesive: a window on prehistoric technological complexity" at Delft University of Technology and in collaboration with Leiden University. His PhD project, supervised by Dr. G. Langejans, Prof. A.L. van Gijn, and Prof. J. Dik, aims to study and compare technological complexity between Neanderthals and modern humans through the analysis of adhesive remains. The achievements within the PhD project are presented in this dissertation.

From May 2024, Alessandro has worked as a researcher in the "BiDebA-Biobased Debondable Adhesives" project funded by Interreg North-West Europe.

List of publications

1. **Aleo, A.**, Bradtmöller, M., Chasan, R., Despotopoulou, M., Gonzalez Gomez, J.A., Kozowyk, P.R.B., Rodríguez, F., Langejans, G.H.J. *submitted*. Persistent use of ochre and plant extracts in Palaeolithic adhesive technology at Morin Cave, Spain. *Journal of Palaeolithic Archaeology*
2. Chasan, R., Veall, M., Baron, L.I., **Aleo, A.**, Kozowyk, P.R.B., Langejans, G.H.J 2024. *Podocarpaceae* and *Cupressaceae*: A tale of two conifers and ancient adhesives production in South Africa. *PLoS* ONE 19(11): e0306402. https://doi.org/10.1371/journal.pone.0306402
3. **Aleo, A.**, Jerardino, A., Rivka Chasan, Myrto Despotopoulou, Dominique J.M. Ngan-Tillard, Ruud W.A. Hendrikx, Geeske H.J. Langejans 2024. A multi-analytical approach reveals flexible compound adhesive technology at Steenbokfontein Cave, Western Cape. *Journal of Archaeological Science* 167, 105997. https://doi.org/10.1016/j.jas.2024.105997
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7. **Aleo, A.** 2022. Comparing the Formation and Characteristics of Use-Wear Traces on Flint, Chert, Dolerite, and Quartz. *Lithic Technology*, 1-19. https://doi.org/10.1080/01977261.2022.2103297
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