Footprints of fire: understanding the formation and preservation of Pleistocene fire traces through laboratory-based experimental research

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‘Come let’s light a fire, drink a beer, and sing a song’

- Will Varley

Chapter 1

Introduction

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Abstract

The *origins of fire use* is a major research theme within Pleistocene archaeology due to the potential role of fire in many aspects of human evolution. This thesis argues that the impact of fire use cannot be properly understood without considering the chemical processes that shape the archaeological fire record (i.e., fire and diagenesis). The best way to do this is through a fundamental research approach, where those processes are broken down into individual measurable variables. Therefore, this thesis examines the effect of fire (heat) and diagenesis (pH conditions) on the physical and chemical properties of bone through a series of controlled laboratory experiments. This is done to 1) provide a more comprehensive understanding of the formation and preservation of heated bone as a fire proxy, and 2) build an analytical toolkit and reference datasets to identify and reconstruct the characteristics of ancient fire more adequately. This chapter provides the archaeological embedding of this chemistry-based work within the field of Pleistocene fire research. It presents the main themes and hypotheses on the role of fire in human evolution, gives an overview of the available archaeological evidence for Pleistocene fire use, and outlines the different approaches used to study ancient fire proxies. Then, some key issues are identified and presented along with a suggested approach to address them, leading into the aims and research questions of the thesis. The chapter closes with a brief outline of the rest of the thesis.

1.1 Present day people and their relationship to fire

Fire is an integral part of human culture, showing up in expected and unexpected aspects of our daily lives. Humans use fire in ways that no other species ever has. People rely on combustion-generated energy for light, warmth, cooking, transportation, and to power electronic devices, while in many places around the world people - about a third of the world's population - still depend on open fires for food preparation and to provide light and warmth (Boudewijns et al., 2022). However, fire is valued for more than as a source of energy; it has symbolic significance in many cultural and religious settings as well. In addition, the way in which fire is embedded in language shows it is associated with a variety of different activities and cultural meanings. For example, in certain Australian Indigenous languages the word for fire is also used to refer to 'family' or 'heart country' (Ward and Friesem, 2021; Ward et al., 2023). A Western example of how deeply intertwined fire is with our cultural fabric is its widespread use as a metaphor in song lyrics[1].

1 Check this Spotify playlist, curated by the author, for examples of songs from different genres that include references to fire: https://t.ly/amIJs

These examples show that people around the world have an intimate relationship with fire for practical purposes, in both direct and indirect ways, and that fire still shapes different aspects of people's languages and cultural practices. Understanding how this relationship came to be and when our ancestors started to use fire is one of the fundamental questions in the fields of Human Evolution and Pleistocene Archaeology.

1.2 The importance of fire in human evolution

The mastery and use of fire is widely considered to have played a key role in human evolution and the development of the human niche, and has provided our ancestors with many adaptive advantages. Similar to ethnographically-documented groups, our hunter-gatherer ancestors likely used fire for a wide range of purposes, including for warmth and light, to cook food, to manipulate and manufacture materials, and to ward off insects and predators (e.g., McCauley et al., 2020). The adoption of fire into the human technological repertoire has been linked to several biological, behavioural, and cultural developments within human evolution, informed by a combination of data from archaeology and other disciplines. Since fire use touches on so many different aspects of the development of the genus *Homo*, it is a topic with potentially far-reaching implications for our understanding of human origins. The most important of these themes, and their implications, are briefly outlined below.

Cooking

Cooking is the most reported use of fire among ethnographic hunter-gatherer groups (McCauley et al., 2020), and was likely also an important part of early hominin fire use. Cooking has several known nutritional and energetic benefits. It improves the digestibility of starch- and protein-rich foods, reduces the energetic costs of chewing and digestion, and provides a tool to get rid of certain toxins and pathogens (Boback et al., 2007; Carmody and Wrangham, 2009a, 2009b; Wrangham and Conklin-Brittain 2003; Wrangham, 2017). The consumption of cooked foods would have provided early hominins with improved energy yield and the ability to broaden their diets, which in turn may have resulted in other behavioural shifts (e.g., Carmody and Wrangham, 2009a, 2009b; Gowlett, 2006; Henry et al., 2011). Research by Richard Wrangham and colleagues suggests that it might have been the incorporation of cooked foods into the human diet, and the related energetic benefits, that facilitated the development of smaller teeth, a smaller gut, and larger brains in *Homo erectus*, around 1.5 Ma (e.g., Carmody and Wrangham, 2009a, 2009b; Wrangham, 2009, 2017). These morphological changes are used to suggest an early control of and reliance on fire, despite a mismatch with the available archaeological evidence of fire use

(e.g., Gowlett and Wrangham, 2013; Wrangham, 2017; see also **Section 1.3**). The oldest direct evidence for the consumption of cooked food, in the form of charred tubers, comes from South Africa and dates to 170 ka (Wadley et al., 2020). Direct evidence for cooking in the Levant and Europe is much younger and dates to 70 ka and 40-50 ka, respectively, and concerns charred fragments of a processed plant mixture, and cooked starches trapped in dental calculus (Henry et al., 2011; Kabukcu et al., 2023).

Warmth

The use of fire to provide warmth is the second most reported application among ethnographically documented hunter-gatherer groups (McCauley et al., 2020). Fire use is often cited as a major adaptation that allowed hominins to move into northern latitudes and survive in the temperate climate of Eurasia (e.g., Attwell et al., 2015; Gowlett, 2006; Hosfield, 2016; Rolland, 1998). In cold winters and harsh climate conditions fire provides several selective advantages, including reducing the energetic costs associated with maintaining core body temperature (Scholander et al., 1958; Wrangham and Carmody, 2010). However, hominin range expansion into Eurasia considerably predates the earliest evidence of fire use outside of Africa. Hominins first move out of Africa around 1.8 Ma and are present in northern Europe by at least 800 ka, while the earliest evidence for regular use of fire in Europe dates to 400 ka (e.g., MacDonald, 2017; Roebroeks and Villa, 2011). More sustained occupation of northern parts of Europe during cooler climatic conditions is visible in the archaeological record from around 500 ka onwards. While this development provides a somewhat better match with the record of available fire evidence, the scarcity of sites dating to that period makes it difficult to establish a causal link (MacDonald, 2017). In addition, the absence or scarcity of fire evidence at certain Middle Palaeolithic sites during cold phases has raised questions about the obligate nature of Neanderthal fire use and their reliance on fire to keep warm (Dibble et al., 2018; Sandgathe et al., 2011a, 2011b). This mismatch between the Lower and Middle Palaeolithic occupation of Europe and the fire record implies that hominins may have had alternative strategies to deal with the energetic pressures of surviving in cold environments (e.g., Hosfield, 2016; MacDonald, 2017, 2018; Speth, 2017).

Landscape management

Deliberate burning of vegetation has been suggested as a form of niche construction by early humans and a way for them to modify and manage the landscapes they inhabited (e.g., Nikulina et al., 2021; Roebroeks et al., 2021a; Thompson et al., 2021). This is a practice widely documented among ethnographic hunter-gatherer groups that improves the quality, predictability, and yield of animal and plant recourses and results in more open mosaic landscapes (e.g., Bliege Bird et al.,

2020; Scherjon et al., 2015). Several researchers have hinted or hypothesised that the initial stages of fire use by early *Homo* may have involved the exploitation of fire-prone environments and the active propagation of landscape fires, specifically for their energetic and nutritional benefits (e.g., Chazan, 2017; Gowlett, 2016; Parker et al., 2016). However, finding evidence for these landscape modification practices in the archaeological record is challenging due to the resolution and chronological constraints needed to relate hominin behaviour to records of environmental change (Nikulina et al., 2021; Roebroeks et al., 2021a; Thompson et al., 2021). Currently the oldest evidence comes from the Eemian site of Neumark Nord 2 (125 ka, Germany), where it has been suggested that Neanderthals used fire as a landscape modification tool (Roebroeks et al., 2021a).

Pyrotechnology

Our ancestors also used fire as a tool to manipulate different materials, e.g., for the production of tar for hafting (e.g., Kozowyk et al., 2017; Mazza et al., 2006; Niekus et al., 2019), for heat treatment of lithics to facilitate tool production (e.g., Delagnes et al., 2016; Schmidt et al., 2013), or for fire-hardening of wooden tools to make them more durable (e.g., Aranguren et al., 2018; Revedin et al., 2019). These activities are used as a proxy for developments in behavioural complexity and cognition because the steps involved imply a certain degree of control over the fire and understanding of both variation in fire and material properties (e.g., Brown et al., 2009; Kozowyk et al., 2023a; Roebroeks and Soressi, 2016; Wadley, 2010; 2013). In addition to identifying the products of pyrotechnologies in the archaeological record and relating these to specific hominins, the discussion often centres around the potential steps of the production process and their inferred complexity.

For example, tar production requires the heating of an organic material, usually birch bark, under low oxygen conditions and at low-medium temperatures (NB: exact temperatures still debated) (e.g., Koller et al., 2001; Kozowyk et al., 2017; Mazza et al., 2006). There are multiple ways of achieving these conditions involving different steps and varying degrees of (temperature) control, efficiency, and complexity (e.g., Blessing and Schmidt, 2021; Koch and Schmidt, 2022; Kozowyk et al., 2017; Schmidt et al., 2019a). In the absence of archaeological evidence on specific production processes, discussions rely on rare tar finds and their chemical signatures (e.g., Niekus et al., 2019; Schmidt et al., 2023). However, both are not without problems due to preservation issues and lack of extensive reference datasets (see Kozowyk et al., 2020; 2023). In relation to Neanderthals, there appears to be consensus that they had access to the required materials and possessed the necessary skill to have invented and developed tar production, likely increasing

efficiency and complexity of the technique by improving on early simple methods (Kozowyk et al., 2017; Niekus et al., 2019; Schmidt et al., 2023). The oldest archaeological evidence currently dates these developments back to around 200 ka (Mazza et al., 2006).

Similarly, the heat-treatment of lithics requires specific steps and skilled use of fire. The lithic material, often flint or silcrete, needs to be exposed to medium-high temperatures (i.e., ~300-500 °C) to evacuate water and induce a phase change in the quartz crystals, but heated and cooled carefully to avoid thermal stress and fracturing (e.g., Schmidt et al., 2013; Wadley and Prinsloo, 2014). As with tar production, there are different ways to achieve the desired result, all with varying degrees of required control and complexity (Schmidt et al., 2013; Wadley and Prinsloo, 2014). The heat-treatment of flint is thought to require more control over specific heating conditions than needed to improve the flaking properties of silcrete (Schmidt et al., 2013). Furthermore, variability in thermal properties between different sources of raw material will influence the degree of necessary temperature control (Mackay et al., 2019; Schmidt et al., 2019b). At present, the oldest evidence for heat-treatment of lithic material comes from the South African Middle Stone Age and dates to around 160 ka (e.g., Brown et al., 2009).

Fire production

Like tar production or heat-treatment of lithics, fire production (i.e., the ability to ignite fires at will) often features in discussions about hominin technological capabilities and their behavioural and cognitive implications. The ability to produce fire at will is widely agreed to be the final step in the development of fire use (see **Section 1.3** for the other steps). Evidence for controlled or habitual fire use has been taken by some researchers as an indication that the associated hominins were likely capable of fire production, suggesting quite a long chronology and placing its origins anywhere between 1.0 Ma and 300-400 ka (e.g., Alperson-Afil, 2008; Beaumont, 2011; Roebroeks and Villa, 2011). Yet others view the absence of fire evidence from (find layers of) certain sites associated with cold climates as a sign that Neanderthals were unable to produce fire at will, suggesting that the technology was invented (and brought to Europe) by Modern Humans (Dibble et al., 2018; Sandgathe et al., 2011a, 2011b). Lombard and Gardenfors (2023) evaluated archaeological evidence for regular fire use and ethnographic fire-making techniques using cognitive models, specifically focusing on fire-drill and strike-a-light technology (i.e., wood-on-wood friction and percussive fire-making, respectively). They concluded that Neanderthals and contemporaneous *Homo sapiens* in Africa likely had the cognitive capacity to have invented and used fire production techniques. They further hypothesise that the fire-drill technique was invented by Modern humans in Africa, while the strike-a-light technique originated in Europe

with the Neanderthals (Lombard and Gardenfors, 2023). However, without evidence for the fire-making tools themselves, testing these hypotheses is extremely difficult. Wood-on-wood friction techniques are unlikely to leave any direct traces due to the organic nature of the materials involved. Percussive fire-making, involving lithic material and pyrite nodules, has a higher preservation potential, but evidence for this technique is still extremely rare for the Pleistocene (Sorensen et al., 2014). This scarcity is related to several factors, including poor preservation of pyrite, the expedient character of Middle Palaeolithic technologies, and a lack of targeted use-wear studies (Sorensen et al., 2014; 2018). Nevertheless, use-wear traces found on MTA bifaces provide direct evidence that suggests that percussive fire-making was a recurring techno-cultural feature among late Neanderthals in France around 50 ka (Sorensen et al., 2018). In addition, indirect evidence, in the form of a distinct biomarker signal (i.e., PAHs and *n*-alkanes), has been used to suggest fire production at Lusakert cave (Armenia) around 40-60 ka (Brittingham et al., 2019).

Social behaviour and culture

In addition to the range of practical applications presented here, fire use has been suggested to have helped shape the intensity and content of social interactions, contributing to the evolution of language, culture, and our social brain (e.g., Gowlett et al., 2012; Lynn, 2014; MacDonald et al., 2021; Wiessner, 2014). The social brain hypothesis posits that the disproportionally large brains seen in primates, including humans, evolved to support their characteristic complex social systems (e.g., Dunbar, 1998; Gowlett et al., 2012). The use of fire is suggested to have facilitated this process in two ways. First, the consumption of cooked food would have provided the calories needed to sustain larger brains (Carmody and Wrangham, 2009a; Gowlett et al., 2012). Second, the use of hearths would have provided a focal point around which social behaviour was encouraged and could develop further. Fire light also extends the time available for social interactions (as well as other tasks) to take place. In addition, the use of a communal fire requires a certain degree of cooperation within a group to maintain (Gowlett et al., 2012; Twomey, 2014). A study on present day people showed that the audio-visual input from campfires lowers blood pressure and induces a relaxation response, which in turn promotes prosocial behaviour. It is not unlikely that sitting around a campfire would have promoted relaxation and social behaviour in our hominin ancestors as well (Lynn, 2014). For ethnographically documented hunter-gatherer groups the content of the social interactions taking place around campfires has been shown to differ from those taking place elsewhere. Fireside conversations tend to involve storytelling, while conversations during e.g., hunting expeditions are much more practical in nature. This implies that certain aspects of language and culture may have evolved around central hearths

(Wiessner, 2014). Finally, fire evidence has been used as a proxy to support hypotheses about the evolution of different cultural mechanisms. For example, the spatiotemporal pattern of regular fire use from around 400 ka onwards (see **Section 1.3**), and particularly its rapid emergence in large parts of the Old World, has been suggested to be the first clear evidence of cultural diffusion. This further implies the existence of a degree of social tolerance and extensive social networks to facilitate the transmission of complex skills like fire use (MacDonald et al., 2021).

The costs of fire

Next to the many benefits, the use of fire also came with costs that may have impacted the behaviour, health, and genetic make-up of early humans (e.g., Aarts et al., 2016; 2021; Henry et al., 2018a; Hubbard et al., 2016). Smoke generated during the combustion of biomass fuel (e.g., wood, dung, peat) contains a range of toxins (e.g., polycyclic aromatic hydrocarbons, dioxins) that are known to be carcinogenic and cause infertility issues in present day people (e.g., Aarts et al., 2016; Mishra et al., 2005; Smith et al., 2000). To combat the adverse health effects of environmental pollutants (incl. smoke) humans and other primates possess detoxification genes (Aarts et al., 2016). It has been suggested that changes in the detoxification capacity of our ancestors may have been linked to the introduction of regular fire use. However, when exactly these changes occurred, if they were in fact related to regular exposure to smoke, and which of the hominin species studied so far possessed the more efficient genetic detoxification make-up (i.e., Neanderthals, Denisovans, or early modern humans) is still debated (Aarts et al., 2016; 2021; Hubbard et al., 2016). Nevertheless, it seems reasonable to assume that frequent exposure to smoke toxins would have resulted in some adverse health effects for hominins regularly using fire.

In addition to the impact of toxins, there are energetic costs associated with the production and maintenance of fire. Based on behavioural ecology models it has been suggested that regular use of fire may have been a flexible adaptation, as opposed to obligate use, where fire was only used as a tool when the benefits it provided outweighed the required investment of time and energy (Henry 2017; Henry et al., 2018a). One of the main costs associated with the use of fire is the time and energy investment required for the procurement of fuel (Henry et al., 2018a; Pryor et al., 2016). Consequently, environmental constraints will influence behavioural choices such as fuel management strategies, mobility, and subsistence strategies (e.g., Henry et al., 2018b; Sorensen, 2017; Thery-Parisot and Meignen, 2000), and may result in alternative fire-free adaptive strategies (e.g., MacDonald, 2018; Speth, 2017). In turn, these choices and adaptations will influence the archaeological pattern of fire evidence.

1.3 Fire evidence in the Pleistocene archaeological record

Tracing the origins of hominin fire-related behaviour, and testing hypotheses on the themes outlined in **Section 1.2**, relies on the (additional) identification of evidence of fire use in the archaeological record. Comprehensive efforts to bring together fire evidence from different sites and critically evaluate the resulting patterns were first presented in the 1950s by Kenneth Oakley (1954; 1956a; 1956b). This section presents a brief overview of the archaeological evidence available in the Pleistocene record in different parts of the world, highlighting some of the key sites. A more extensive examination is beyond the scope of this work but can be found in various in-depth review papers (e.g., Bentsen, 2014; Gowlett and Wrangham, 2013; James, 1989; Murphree and Aldeias, 2022; Roebroeks and Villa, 2011).

Hominin use of fire is assumed to have evolved in several stages: 1) an initial phase of opportunistic (uncontrolled) use that relied on the interaction with natural sources, 2) an intermediate phase of more (habitual) controlled use of fire without the means of producing it, and 3) a final phase of habitual controlled use of fire that could be produced at will (e.g., Burton, 2009; Chazan, 2017; Parker et al., 2016; Preutz and Herzog, 2017; Roebroeks and Villa, 2011; Sandgathe, 2017). Despite this wide consensus, the timing, nature, and universality of these developments are still debated. Fire use is identified in the archaeological record through the presence of various heated materials, found in association with traces of human activity. These heated materials serve as proxy data for the presence of fire itself and include charred organic matter, ash, soot, heated bones, heated lithics and stones, heated sediment, tar, cooked food remains (e.g., cooked starches trapped in dental calculus), and heated biomolecules found in sediments. These proxies can appear as scattered finds, as layers or patches, and under favourable conditions as combustion features, i.e., *hearths*. The latter are often taken as the most reliable indication of anthropogenic fire evidence (e.g., James, 1989; Murphree and Aldeias, 2022; Roebroeks and Villa, 2011).

The Early Pleistocene

The earliest examples of preserved traces of fire found in association with evidence of hominin activity come from a handful of sites in Africa and date to around 1.5 Ma (e.g., Bellomo, 1994; Clark and Harris, 1985; Gowlett et al., 1981; Isaac, 1982; James, 1989). They are found in open air contexts and the evidence mainly consists of patches of rubified sediments and clay aggregates that are presumed to result from heating (e.g., Gowlett and Wrangham, 2013; James, 1989).

The most prominent of these early sites is Koobi Fora in Kenya, which has received extensive re-evaluation in recent years (e.g., Cutts et al., 2019; Hlubik et al., 2017; 2019). In addition to patches of heated sediment, the evidence for fire at this site consists of heated bone and heated lithics closely associated with those patches. Thermal alteration of the suspected fire traces was confirmed using experimental approaches and analytical techniques (Hlubik et al., 2019).

Dating to around 1 Ma, Wonderwerk and Swartkrans (both in South Africa) are often cited as examples of the oldest evidence for fire use found in a cave setting (e.g., Bentsen, 2014; Gowlett and Wrangham, 2013; Stahlschmidt et al., 2015). The evidence at Wonderwerk consists of macroscopically observed ash lenses and heated bone, as well as ashed plant remains and heated bone fragments, identified through micromorphology (Beaumont, 2011; Berna et al., 2012). However, a more recent micromorphological study suggests that some of the identified ash at the site is actually calcified plant remains (Goldberg et al., 2015). At Swartkrans the evidence consists of a collection of heated bones, some of which also display cutmarks (e.g., Bentsen, 2014; Brain and Sillen, 1988; Pickering et al., 2008). A combination of experimental work and analytical techniques was used to confirm that the bones were heated and not black due to manganese staining (Brain and Sillen, 1988).

Outside of Africa the evidence for possible fire use by early Pleistocene hominins is very limited and restricted to only two sites: Bogatyri (Russia) and Cueva Negra (Spain). Alongside these, the early Middle Pleistocene site of Gesher Benot Ya'aqov (Israel) is also often mentioned (e.g., Attwell et al., 2015; Bentsen, 2014; Gowlett and Wrangham, 2013; Hlubik et al., 2019; Scott and Hosfield, 2021). At Bogatyri, dated to around 900 ka, the reported evidence consists of fragments of heated bone, some of which are calcined (Bosinski et al., 2003; Bosinski, 2006; Gowlett and Wrangham, 2013). Cueva Negra is dated to around 800 ka and produced small pieces of heated chert and fragments of heated bone (incl. microfauna) found in association with an area of reddened sediment. Heat alteration of the chert and bones was confirmed using analytical techniques (Rhodes et al., 2016; Walker et al., 2016). At Gesher Benot Ya'aqov (780-700 ka) repeated use of fire is suggested based on so-called '*phantom hearths*', identified using spatial analysis of heated lithic material (e.g., Alperson-Afil, 2008; Alperson-Afil et al., 2007; Alperson-Afil and Goren-Inbar, 2006; Goren-Inbar et al., 2004). Recently, claims for the consumption of cooked fish have also been made for the site (Zohar et al., 2022).

Despite decades of research these very early claims for hominin fire use are still contentious. Traces are often ephemeral and easily removed from the archaeological record by taphonomic

processes (e.g., Roebroeks et al., 2021b). In addition, the anthropogenic nature of the fire traces remains difficult to verify, despite their association with evidence of hominin behaviour (e.g., James, 1989; Pickering, 2012; Roebroeks et al., 2021b). This is particularly challenging for evidence found in open air contexts, where a combination of patchy vegetation and wildfire could leave similar traces (Goldberg et al., 2017; McNiven et al., 2019). Because of these uncertainties, considerable effort is focussed on establishing the degree of integrity of the stratigraphic context and verifying the heat-altered nature of the observed traces (e.g., Brain and Sillen, 1988; Goldberg et al., 2015; Hlubik et al., 2019). Due to the low site density in general and limited number of sites with fire evidence it is difficult to establish a pattern that might lend (additional) support to claims from individual sites and hypotheses on the evolutionary impact of Early Pleistocene fire use (e.g., Hlubik et al., 2019; Roebroeks et al., 2021b).

The Middle Pleistocene

Evidence for fire use continues to be virtually absent from the archaeological record in many research areas throughout the first half of the Middle Pleistocene. This scarcity may be attributed to low site density, preservation issues, hominin behaviour, or a combination of all three and should therefore be treated with caution (e.g., Gowlett, 2006; Roebroeks and Villa, 2011; Roebroeks et al., 2021b; Scott and Hosfield, 2021). An extensive review of the European record, with its high site density and rich research history, showed that a pattern of widespread regular fire use does not emerge until the second half of the Middle Pleistocene (Roebroeks and Villa, 2011). From around 400 ka onwards sites with multiple proxies start to appear and become increasingly frequent. This trend is further illustrated by sites with long sequences and good stratigraphic control, like Caune de L'Arago (France), where fire evidence does not appear until after 350 ka (de Lumley, 2006; Roebroeks and Villa, 2011; but see Deldicque et al., 2021 for possible older traces). During this period evidence for fire is present in open-air contexts, rock shelters, and caves, with some sites showing evidence of repeated use over long periods of time (MacDonald et al., 2021; Roebroeks and Villa, 2011; Roebroeks et al., 2021b). Examples of the latter can be found at Bolomor Cave (Spain) and Biache-Saint-Vaast (France) (Herisson et al., 2013; Peris et al., 2012).

The change in the frequency and visibility of fire evidence in the European record, from 400-350 ka onwards, can also be seen at sites in other parts of the world. At Tabun Cave (Israel) the long and well-dated sequence shows a considerable increase in the frequency of heated lithics starting between 350-320 ka, a trend that can also be seen at other sites in the region (Shimelmitz et al., 2014). At Qesem Cave (Israel, 400-200 ka) there is a marked increase in fire

proxies such as ash, heated lithics, and heated bone from the lower to the upper part of the sequence (Karkanas et al., 2007; Stiner et al., 2011). In addition, the site contains a large central hearth dating to 300 ka that shows signs of repeated use (Shahack-Gross et al., 2014). A notable example from Asia is the site of Zhoukoudian (China). Evidence for anthropogenic fire at the site has been contested since the 1980s (e.g., James, 1989; Goldberg et al., 2001; Weiner et al., 1998). However, renewed excavations, coupled with the application of different analytical techniques, have uncovered unambiguous traces of anthropogenic fire (e.g., Gao et al., 2017). These include heated sediments, ash remains, and heated bones (e.g., Huang et al., 2022; Zhang et al., 2014; Zhong et al., 2013). Various dating techniques suggest an age in the range of 500-250 ka (Gao et al., 2017). In northern Africa the Middle Pleistocene change in the fire signal can be found at Jebel Irhoud (Morocco). Here evidence includes heated lithics, heated bone, and possible combustion features, dating to around 300 ka (Richter et al., 2017). A review of the archaeological record in southern Africa shows a comparable pattern of regular fire use during the Middle Stone Age (~ 270 ka onwards). Like in Europe and the Levant, sites often contain multiple proxies including combustion features (Bentsen, 2014; Roebroeks et al., 2021b).

The clear change in the scale of available fire traces in the second half of the Middle Pleistocene is accompanied with the first evidence for specific types of fire use. Simple combustion features start to appear and gradually become more common (e.g., Peris et al., 2012; Richter et al., 2017; Roebroeks and Villa, 2011; Scott and Hosfield, 2021; Shahack-Gross et al., 2014). In addition, there is direct evidence for hominins cooking their food (Wadley et al., 2020), as well as using pyrotechnology to produce tar and heat-treat lithic material (Brown et al., 2009; Mazza et al., 2006). Finally, there are early indications of landscape management by Neanderthals (Roebroeks et al., 2021a).

The Late Pleistocene

The Middle Pleistocene trend of increasing frequency of sites with fire evidence and preservation of multiple proxies continues into the Late Pleistocene, in Europe and the Levant. Evidence for the use of fire is present in caves, rock shelters, and open-air sites (e.g., Murphree and Aldeias, 2022; Roebroeks and Villa, 2011). Combustion features further increase in frequency, with stone-lined and structured hearths becoming more common in the Upper Palaeolithic (Murphree and Aldeias, 2022). Examples of sequences with repeated use of fire in the form of (well-studied) combustion features can be found at sites like Abric Romani, El Salt (both in Spain), Abri Pataud (France), and Kebara (Israel) (e.g., Albert et al., 2012; Braadbaart et al., 2020; Leierer et al., 2019; Vallverdu et al., 2012). A similar trend can be seen in the African record, with prominent South

African examples of combustion features at Sibudu and Klasies River (e.g., Goldberg et al., 2009; Larbey et al., 2019; Wadley, 2012). At the latter, remains of cooked starchy food were also found (Larbey et al., 2019). Further east, in Asia and Australia, examples of suggested Late Pleistocene evidence for the use of fire can be found at sites in the Belan valley (India), Tolbor-17 (Mongolia), Shuidonggou (China), and Riwi Cave (Australia) (e.g., Gallo et al., 2021; Gao and Dennell, 2014; Jha et al., 2021).

Despite regular fire use being a common and widespread phenomenon in the Late Pleistocene, combustion features and other traces of fire are not preserved everywhere or in all layers (e.g., Allue et al., 2022; Dibble et al., 2018; Murphree and Aldeias, 2022; Sandgathe et al., 2011a, 2011b). At Pech de l'Azé IV and Roc de Marsal, two well-studied sites in the Dordogne (France), various occupation levels lack evidence of fire. At both sites there is abundant fire evidence, including combustion features, in layers associated with temperate conditions, while fire evidence is virtually non-existent in layers deposited during cold phases (e.g., Dibble et al., 2018; Sandgathe et al., 2011a, 2011b). This striking absence of evidence and inconsistency in the record has led to discussions on the nature of hominin fire use during these periods (e.g., Dibble et al., 2018; Roebroeks and Villa, 2011; Sandgathe et al., 2011a, 2011b; Sorensen, 2017). In particular, the team working on Pech de l'Azé IV and Roc de Marsal has questioned if Neanderthal fire use can truly be viewed as habitual, rather than opportunistic, and if Neanderthals were able to produce fire themselves (e.g., Dibble et al., 2018; Sandgathe et al., 2011a, 2011b). However, there are other behavioural factors, as well as taphonomic ones, that could explain the observed pattern (e.g., Sorensen, 2017; Sorensen and Scherjon, 2018). Regardless, there is direct evidence that suggests that, at least at certain times and places, Neanderthals were able to produce fire (Sorensen et al., 2018). In addition, the Late Pleistocene sees the first direct evidence of cooking outside of Africa (Kabukcu et al., 2023; Henry et al., 2011).

1.4 Approaches to the study of ancient fire use

Research on the origins and nature of hominin fire use has largely relied on traditional archaeological approaches, including documenting the presence of fire proxies, their spatial relationship to other artefacts and site elements, and typological descriptions of combustion features (e.g., Alperson-Afil et al., 2007, 2009; Clark et al., 2022; Pop et al., 2016; Roebroeks and Villa, 2011; Vallverdu et al., 2012; Vaquero and Pasto, 2001; Whitau et al., 2018). Traditional disciplines like archaeozoology and archaeobotany are employed to answer questions related to subsistence and fuel use (e.g., Costamagno, 2013; Fladerer et al., 2014; Kabukcu et al., 2023;

Thery-Parisot, 2002; Vidal-Matutano et al., 2017, 2019). Originally introduced in the 1950s by Oakley, the past decade has seen a marked increase in the application and importance of analytical techniques and experimental data in fire research (e.g., Aldeias, 2017; James, 1989; Mentzer, 2014; Oakley, 1954, 1956a, 1956b; Roebroeks et al., 2021b). While still a developing field, this shift is starting to provide more high-resolution data and a broader understanding of different aspects of fire use. This section provides a brief overview of the contribution of analytical approaches (i.e., techniques that target the physical, chemical, structural, and micro-contextual properties of fire proxies) and experimental work to different aspects of the study of Pleistocene fire use, with a focus on the identification of proxies and the use of their properties to study variation in fire use. An extensive explanation of different techniques and breakdown of different proxies and variables is beyond the scope of this work, but can be found elsewhere (e.g., March et al., 2014; Mentzer, 2014; Weiner, 2010).

Analytical techniques

The application of analytical techniques to Pleistocene fire research is primarily geared towards (verifying) the identification of fire proxies and distinguishing heat-alteration from other forms of weathering (e.g., Brain and Sillen, 1988; Goldberg et al., 2015; Hlubik et al., 2019; James, 1989; Stahlschmidt et al., 2015). An example of the latter is the use of FTIR to determine whether black-coloured bones are the result of heating, manganese staining, or both (e.g., Brain and Sillen, 1988; Shahack-Gross et al., 1997). Effort has also been put towards devising analytical approaches to differentiate between natural and anthropogenic fire evidence, e.g., through palaeomagnetism (e.g., Bellomo, 1993). Various techniques are available to detect and verify heat-alteration in different macroscopic materials. In general, organic fire proxies are identified by characterising changes in their elemental, structural, and molecular properties. This can be done using techniques like CHN analysis, Reflectance analysis, FTIR spectroscopy, and different types of Mass Spectrometry (e.g., Braadbaart and Poole, 2008; Brain and Sillen, 1988; Gosling et al., 2019; Koller et al., 2001; Weiner, 2010). In the case of bone, which has both organic and inorganic components, FTIR is used to characterise heat-induced changes in the bone mineral as well (e.g., Ellingham et al., 2015a; Lebon et al., 2008, 2010; Snoeck et al., 2014; Thompson et al., 2009). Those same changes can also be detected by XRD analysis (e.g., Ellingham et al., 2015a; Gallo et al., 2021; Piga et al., 2016). For fully inorganic materials the focus is on structural and mineralogical changes (e.g., Weiner, 2010). For example, heated lithics, rocks, and sediments can be identified using archaeomagnetism and luminescence techniques (e.g., Bellomo, 1993; Carrancho et al., 2019; Pop et al., 2021; Rapp et al., 1999; Richter et al., 2011).

At the micro scale, fire proxies are commonly identified through micromorphology based on visual as well as chemical and structural characteristics (e.g., Mentzer, 2014; Weiner, 2010). Micromorphology allows fire evidence to be studied within its sedimentary context and for the integrity of that context to be assessed. This can help determine whether heating took place *in situ* and which (post-depositional) actions might have affected a combustion feature (e.g., Berna et al., 2012; Goldberg et al., 2017; Mentzer, 2014). The technique can also be used to verify suspected heat-alteration of individual proxies and combustion features (e.g., Goldberg et al., 2015; Stahlschmidt et al., 2015). In addition to verifying the identification of fire proxies for which heating is already suspected based on visual characteristics, analytical techniques can be used to identify traces that are not visible to the naked eye, either in conjunction with micromorphology or on their own. Magnetic susceptibility has been used to identify changes in ferromagnetic properties of sediments that could be the result of heating (e.g., Deldicque et al., 2021; Herries and Fisher, 2010; Kapper et al., 2014). Mass spectrometry techniques have been used to identify heated biomolecules in sediments (e.g., Brittingham et al., 2019; Leierer et al., 2019; Lejay et al., 2019). Polycyclic aromatic hydrocarbons (PAHs) in particular are a useful proxy to distinguish between natural and anthropogenic fire (e.g., Brittingham et al., 2019).

In addition to their use in the identification of fire proxies, in recent years work is emerging that applies analytical approaches to move beyond simple presence and absence and addresses questions related to variation in fire-related behaviour. This includes a focus on micro-structure, environmental setting, signatures of different plant and animal tissues, and variation in heating conditions (e.g., Aldeias et al., 2012; Braadbaart et al., 2020; Jambrina-Enriquez et al., 2019; Leierer et al., 2019, 2020). So far, the focus has been on combustion features as these present clear anthropogenic records of localised fire use in a primary context, often with multiple proxies available (e.g., Braadbaart et al., 2020; Goldberg et al., 2012; Leierer et al., 2019, 2020; Mallol et al., 2019). This makes them the ideal context to test new methods and approaches. Insight into different aspects of fire use can be obtained by studying the micro-structure of combustion features with micromorphology (e.g., Mentzer, 2014). The technique can provide information on different phases of use and reuse of a feature, as well as on maintenance activities like ash clearing, and intensity of (site) use (e.g., Allue et al., 2022; Goldberg et al., 2009; Leierer et al., 2019; Miller et al., 2008; Shahack-Gross et al., 2014). Micromorphology can be coupled with (geo)chemical techniques, like FTIR, XRF, and organic geochemistry (e.g., lipid biomarkers and their isotopic signatures), to identify and characterise microscopic fire traces within their sedimentary matrix (e.g., Aldeias et al., 2012; Allue et al., 2022; Berna et al., 2012; Goldberg et al., 2012; Leierer et al., 2019). Organic geochemistry can provide data on environmental conditions

and the types of organic materials heated in and around the fire (e.g., plant vs. animal tissues) (e.g., Braadbaart et al., 2020; Jambrina-Enriquez et al., 2019; Leierer et al., 2019). Variation in use can also be examined by determining the conditions under which fire proxies have been produced (e.g., temperature, oxygen availability), providing insight into the characteristics of the fire (e.g., Braadbaart and Poole, 2008; Braadbaart et al., 2020; Gosling et al., 2019; Rasmussen et al., 2012; Stamataki et al., 2021). Variation in fire characteristics may relate to how the fire was used and managed, e.g., maintaining a flaming or smouldering fire or creating reducing conditions all require different temperatures, degrees of oxygen flow, spatial configurations, and fuelling strategies (e.g., Rein, 2009, 2013; Richter et al., 2021; Santoso et al., 2019). In turn, these characteristics may eventually be linked to specific fire functions (e.g., Aldeias, 2017; Braadbaart and Poole, 2008).

Experimental research

Within the study of Pleistocene fire use experimental research is aimed at improving our understanding of the effect of heat on various materials, gaining insight into the types of fire proxies produced under different conditions, producing reference data, and investigating the link between fire characteristics, proxies, and fire function (e.g., Aldeias, 2017; March et al., 2014). Archaeological experimental work often takes an actualistic approach, where research questions are addressed by recreating realistic settings that involve the interplay of different variables (e.g., March et al., 2014). Studies that explore the effect of fire on different materials in this way include work focussed on lithics (e.g., Cutts et al., 2019; Halbrucker et al., 2021), stones (e.g., Backhouse and Johnson, 2007; Oestmo, 2013), sediments (e.g., Brodard et al., 2016; Canti and Linford, 2000), (molecular) organic residues (e.g., Buonasera et al., 2019; Lejay et al., 2016, 2019; Wadley et al., 2019), and various types of faunal remains (e.g., Aldeias et al., 2019; Perez et al., 2017; Shipman et al., 1984; Stiner et al., 1995). These studies largely focus on visual characteristics of heat-alteration, but some link these to chemical changes as well (e.g., Aldeias et al., 2019; March et al., 2014; Stiner et al., 1995). Experimental research has also been conducted to explore different aspects of fuel use, including heating properties of different fuels, efficiency for specific tasks, and variation in resulting proxy signatures (e.g., Albert and Cabanes, 2007; Braadbaart et al., 2012; Buonasera et al., 2019; Hoare, 2020; Lejay et al., 2016, 2019; Thery-Parisot, 2002). Another subset of experimental research relates to understanding the formation of combustion features and their sedimentary signatures. This includes work focussing on variation in use and re-use, fuel strategies, different fire functions, duration of use, and taphonomic actions (natural and anthropogenic), where feature shape, size, (micro) layering, and molecular signatures are documented (e.g., Bentsen, 2012; Buonasera et al., 2019; Mallol

et al., 2013a, 2013b; March et al., 2014). The most extensive of these studies is the work lead by March which presents a detailed report of 20 years' worth of combustion and preservation experiments, addressing a range of different variables (March et al., 2014). While not exactly experimental, worth briefly mentioning here as well are ethnoarchaeological studies on different aspects of the use and preservation of combustion features. These studies apply archaeological techniques, often micromorphology, to combustion features used by extant hunter-gatherer groups to gain insight into the relationship between fire use, abandonment, and the formation of archaeological signatures (e.g., Friesem et al., 2017; Friesem, 2018; Mallol and Henry, 2017; Mallol et al., 2007). Finally, actualistic experiments have been an important tool to explore the possible steps and choices involved in different pyrotechnologies, like the possible methods of aceramic tar production (e.g., Groom et al., 2015; Kozowyk et al., 2017; Schenck and Groom, 2016) or possible approaches to the heat-treatment of lithics (e.g., Wadley and Prinsloo, 2014).

In addition, or as an alternative to actualistic studies, some experimental fire research is conducted under laboratory conditions. This approach allows for a more detailed examination of individual variables and more control over experimental parameters. While common practice in the related field of forensics (e.g., Ellingham et al., 2015b, 2016; Krap et al., 2019, 2021; Marques et al., 2018; Snoeck et al., 2014; Thompson et al., 2009), the approach is not widespread within archaeology. However, a handful of laboratory-based archaeological experimental studies exist. These primarily focus on understanding the processes, and the role of individual variables, involved in heat-alteration of specific materials, providing detailed reference data, or method development (e.g., Aldeias et al., 2016; Braadbaart and Poole, 2008; Gallo et al., 2021, 2023; Milano et al., 2018; Pop et al., 2021). A laboratory-based experimental study on heat transfer in sediments, involving controlled heating by a heating element, has provided quantitative data on the relationship between temperature, exposure time, and penetration depth, among other variables (Aldeias et al., 2016). A recent study on the thermal alteration of lithics used controlled heating to explore the relationship between temperature, exposure time, and one specific visual characteristic of heat-alteration (i.e., potlid fractures) (Abdolahzadeh et al., 2023). Other experimental work has focussed on understanding the chemical and structural changes underlying the visual signs of heat damage in lithics, particularly in the context of heat-treatment of lithic raw materials (e.g., Mackay et al., 2019; Schmidt, 2014; Schmidt et al., 2013, 2012, 2016, 2019). Important work on bone was conducted by Shipman and colleagues (1984), who used controlled heating experiments to investigate the relationship between heating temperature and changes in colour, morphology, shrinkage, and crystalline structure. They identified five heating stages and proposed the use of XRD to identify bone heated to temperatures above

600 °C (Shipman et al., 1984). Some work has also been done on crystallinity changes in high temperature heated bone and the use of FTIR spectroscopy (e.g., Lebon et al., 2010; Snoeck et al., 2014). Finally, quite extensive work has been done on the effect of charring temperature on the physical and chemical properties of different plant materials, like wood and seeds (e.g., Ascough et al., 2010; Braadbaart, 2004, 2008; Braadbaart et al., 2004, 2007; Braadbaart and Poole, 2008). Notably, controlled experiments have also been performed to explore the effect of diagenesis on charcoal preservation and the reliability of the techniques used to reconstruct heating temperature (Braadbaart et al., 2009). A somewhat similar study on heated bone explored the effect of diagenesis on physical preservation (Kalsbeek and Richter, 2006).

1.5 Problem statement: the need for fundamental research

It is clear that fire likely played a key role in many aspects of human evolution. Therefore, gaining insight into the timing, nature, and development of hominin fire use has potentially far-reaching implications for our understanding of the genus *Homo*. However, there is a mismatch between hypotheses about the evolutionary role of fire use and the actual evidence available in the archaeological record. Pleistocene fire evidence is patchy and partially obscured by issues of preservation, especially early on. The resulting record is varied, with fire traces appearing together in different combinations and not all of them preserving everywhere. Furthermore, combustion features show up in the record relatively late. This variation is likely the reflection of a mix of preservation issues and diversity in hominin behaviour. As with many other topics in archaeology, the debates on the origins of fire use largely hinge on the availability of archaeological evidence and the correct identification and interpretation of fire proxies (cf. Roebroeks and Villa, 2011). The application of analytical techniques and experimental research has already proven to be valuable but is still very much a developing field. Moving research on Pleistocene fire use forward requires a more fundamental understanding of the formation and degradation of fire proxies and improved tools to study them, with a focus on three core themes: identification of fire evidence, studying variation in use, and the effect of preservation. These themes are briefly outlined below, followed by the suggested solutions and the focus of this thesis.

Identification

The reliability of hypotheses about Pleistocene fire use, and our ability to resolve the current debates and address the patchiness of the archaeological record, largely depends on the correct identification of the different fire traces (e.g., Roebroeks and Villa, 2011). Incorrect identifications

affect the interpretation of behaviour at the level of a single site and change the patterns of fire evidence that are built from those individual cases (e.g., Gao et al., 2017; Stahlschmidt et al., 2015). Identification of fire traces is traditionally (and often still) based on visual inspection, both macro- and microscopic. This approach results in problems with equifinality as certain types of weathering may closely resemble traces of heating (e.g., Pop et al., 2021; Shahack-Gross et al., 1997). Additionally, less clear and/or 'invisible' traces will go undetected. Other issues relate to the ability to distinguish between anthropogenic and natural fires (e.g., Hlubik et al., 2017). Consequently, there is a need for more objective and detailed methods for the identification of fire proxies. The application of such improved tools to verify existing fire evidence from old excavations and contentious sites will strengthen patterns and help generate new hypotheses (e.g., Roebroeks et al., 2021b). Analytical techniques have been successfully applied to verify heating, but there is a need for more extensive reference data and a formal approach, specifically one that works for dispersed traces as well as combustion features. The latter also requires a more comprehensive understanding of individual fire proxies.

Variation in use

In addition to identifying fire proxies, an important prerequisite to addressing different hypotheses about Pleistocene fire use is determining how the fire was used. Gaining insight into variability in use, based on how materials were heated (i.e., fire characteristics), can increase our understanding of how fire use developed over time (i.e., the different stages) and provide clues into the degree of control over the fire (e.g., Braadbaart et al., 2020; Braadbaart and Poole, 2008). The latter can also help answer questions related to behavioural and cognitive complexity, specifically in relation to the development of pyrotechnologies. Research on those technologies has highlighted the importance of control over specific heating conditions and fire characteristics like temperature and oxygen availability (e.g., Kozowyk et al., 2017; Schmidt et al., 2013). Since all fire use is the application of heat energy (or sometimes light and smoke) to a specific task, the importance of reconstructing fire characteristics extends beyond research on pyrotechnologies (e.g., Braadbaart et al., 2020; Braadbaart and Poole, 2008). Furthermore, studying the characteristics of a fire adds a perspective that cannot be achieved through more traditional means. Determining fire characteristics requires four things: an understanding of fire chemistry, an understanding of the material properties of the different proxies, appropriate analytical tools, and comprehensive reference data. There are various techniques available to target different proxies (see **Section 1.4**), but a formal approach is still lacking. There is also a wealth of experimental research focussed on the relationship between different types of use and resulting proxies (see **Section 1.4**). However, most of this work concerns actualistic experiments

with little to no control over different variables, resulting in context-specific data. Therefore, a baseline of more controlled experimental data is needed to link observations with specific heating conditions (e.g., temperature, oxygen availability) and material properties. Some data (and related tools) of this nature already exists for certain types of charred plant material (see **Section 1.4**), but the field would benefit from building similar reference datasets and analytical toolkits for other fire proxies.

Preservation

The Pleistocene fire record is undeniably biased by issues of preservation, affecting the availability of fire proxies at the local and regional scale. Addressing this bias is essential to determine the reliability of the evidence. Accounting for the effect of major post-depositional processes (e.g., sediment movement, surface weathering, frost-action) on the preservation of archaeological materials (incl. fire proxies) is a standard part of the modern archaeological practice, particularly in relation to assessing the integrity of the stratigraphic context of a site. However, the more invisible chemical processes (i.e., diagenesis) that affect fire proxy preservation are less routinely examined (but see Weiner, 2010*;* and work by Karkanas). Diagenesis can cause loss of material, even when that material remained *in situ* (e.g., Weiner, 2010), and thus alter patterns of fire evidence at both individual sites and at a regional scale. Absence of evidence is not always true evidence of absence, but rather an artefact of preservation bias (cf. Roebroeks and Villa, 2011). Not addressing this issue may lead to misinterpretation of (the absence of) fire evidence and hypotheses about the related human behaviour. Furthermore, certain types of fire use may have a higher preservation potential than others, leading to different degrees of visibility in the archaeological record. Therefore, the field would benefit from a more comprehensive understanding of the effect of diagenesis on different fire proxies. This will provide data to predict when and where fire evidence might preserve and help tease apart the effect of behaviour and preservation. In addition, data on diagenesis is needed to test the reliability of the analytical tools, used to identify fire proxies and reconstruct heating conditions, under different preservation conditions. Some data exists on the chemical degradation pathways affecting charred plant material and ash (e.g., Braadbaart et al., 2009; Karkanas, 2021; Weiner, 2010), but more data is needed on other proxies, as well as in relation to different analytical techniques.

Fundamental research approach

Understanding the role of fire use in human evolution and addressing the issues outlined above cannot be adequately done without considering the chemical processes that shape the archaeological fire record (i.e., fire and diagenesis). This thesis proposes to build that

idea into a formal approach for the study of archaeological fire evidence through the use of fundamental research (i.e., science aimed at explaining natural phenomena) and controlled laboratory experiments (for details see **Chapter 2**). Using a fundamental research approach to study the universal processes critical to the formation and preservation of different fire proxies will produce context-independent data and tools that can be used to interpret archaeological material regardless of age, depositional setting, or type of fire signature. The best way to do this is through controlled laboratory experiments where fire and diagenesis are broken down into measurable (primary) variables, which can then be related to changes in the physical and chemical properties of the fire proxy under study. Since the outcome of these processes depends on the characteristics of the starting material, different fire proxies should be studied in detail individually before being integrated into a larger dataset (NB: each fire proxy has its own information potential, see **Section 2.2**). In addition, the suggested approach includes the integration of different analytical techniques to increase the robustness of the data.

This thesis takes one specific fire proxy, heated bone, to start building insight, tools, and reference data on the effect of fire and diagenesis, with the explicit goal to eventually integrate these with comparable results from other proxies. Heated bone is among the most commonly reported evidence of burning and has a high information potential due to its unique composition (see **Section 2.2**), making it a very valuable proxy. Through controlled experiments and analytical chemistry, the work aims to characterise bone heated under different conditions before and after exposure to distinct geochemical conditions. The resulting toolkit and reference datasets will 1) improve our means to identify heated bone, 2) allow the use of its physical and chemical properties to reconstruct fire characteristics, 3) help determine when and where heated bone might be expected to preserve, and 4) improve the reliability of the toolkit by providing data to correct for the effect of diagenesis. Together, these facilitate higher resolution data and a more detailed understanding of the timing and nature of hominin fire use and related biological, behavioural, and cultural developments.

1.6 Aims and research questions

This thesis contributes to the study of fire use in human evolution by using a chemistry-based fundamental research approach to understand bone as a fire proxy. The research is focussed on two parts: heated bone in *pre-* and *post*-burial conditions. This is done by addressing the following aims:

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Part one:

1. To understand the effect of fire on bone.
2. To characterise the heated bone that enters the archaeological record.
3. To develop comprehensive, context-independent reference data and an analytical toolkit for the analysis of heated bone and the characterisation of ancient fire.

Part two:

4. To understand the effect of diagenesis on heated bone.
5. To characterise the heated bone that is recovered from the archaeological record.
6. To develop comprehensive, context-independent reference data and an analytical toolkit for the analysis of heated bone and the characterisation of ancient fire, which explicitly takes the effect of diagenesis into account.

To address these aims laboratory-based experiments were performed in which the effect of fire and diagenesis is broken down into measurable primary variables. The variables that are explored within this thesis are *temperature* and *oxygen availability* for fire, and *pH* for diagenesis (see **Section 2.5**). The following research questions are addressed:

1. What is the effect of heating under reducing conditions (i.e., without oxygen: charring) on the physical and chemical properties of bone?
2. What is the effect of heating under oxidising conditions (i.e., with oxygen: combustion) on the physical and chemical properties of bone?
3. What are the physical and chemical differences between charred and combusted bone, and how do these differences impact the way the two types of bone should be studied?
4. What is the effect of pH-exposure (acidic, neutral, alkaline) on the physical and chemical properties of charred and combusted bone?
5. What is the effect of pH-exposure on the reliability of the analytical techniques used to reconstruct past heating conditions?
6. What is the effect of pH-exposure on the preservation potential of charred and combusted bone?

1.7 Building blocks and structure of the thesis

This thesis is the culmination of countless hours of laboratory work focussed on developing skills, knowledge, and a 'feel' for the processes and materials under study. This process involved developing, testing, and improving experimental protocols, as well as gaining hands-on experience with the different analytical techniques employed within the thesis. To this end, a key component was building a network of specialists and laboratories to collaborate with. This investment, although not explicitly mentioned in the final output, is what enabled primarily chemistry-based research to be conducted within an archaeological framework. That combination of chemistry and archaeology resulted in the following chapters and published papers:

The current chapter (**Chapter 1**) provides the archaeological background needed to contextualise the research presented in this thesis. This includes the main research themes in the study of Pleistocene fire use, an overview of the evidence available in the archaeological record, as well as an overview of how analytical techniques and experimental research were integrated in this field. In addition to the scope, aims, and research questions, Chapter 1 also presents the research framework that informed the different parts of this thesis. This framework has two main pillars: 1) in order to recognise and explain human behaviour in the past, archaeologists first need to understand the natural processes that act upon the material records we study, and 2) the best way to study the aforementioned natural processes is by conducting fundamental research and developing context-independent tools and reference data.

Chapter 2 elaborates on the research framework and presents some of the ideas and concepts underlying the overall approach. This includes the value of different fire proxies, in particular heated bone, the rationale for the chosen variables and experimental approach, as well as the value of integrating different techniques and proxies. In addition, the chapter provides some essential background information to contextualise the fundamental processes under study.

Chapter 3, *Charred bone: physical and chemical changes during laboratory simulated heating under reducing conditions and its relevance for the study of fire use in archaeology*, presents the first systematic study into the effects of heating bone under reducing conditions (i.e., charring). The results show that charred bone has a thermal alteration trajectory that is distinctly different from that of combusted bone (i.e., bone heated in the presence of oxygen). This finding has important implications for the reliability of the analytical techniques that are commonly used

to reconstruct heating temperature of heated bone from the archaeological record. Until the addition of this work on charred bone, all exiting analytical work had exclusively focussed on combusted bone, and in doing so not included the full range of heated bone potentially present in the archaeological record. In addition to exploring the effect of charring on the physical and chemical composition of bone, this chapter also presents comprehensive reference data and an analytical toolkit for the characterisation of charred bone from archaeological contexts. The reference data for charred bone, combined with the data presented in Chapter 4, allows for a distinction to be made between charred and combusted bone. This has important implications for the reconstruction of certain aspects of the taphonomy of heated bones, such as differentiating between direct and indirect heating.

Published as: *Reidsma, F.H., van Hoesel, A., van Os, B.J.H., Megens, L., Braadbaart, F., 2016. Charred bone: Physical and chemical changes during laboratory simulated heating under reducing conditions and its relevance for the study of fire use in archaeology. Journal of Archaeological Science: Reports 10, 282–292.* https://doi.org/10.1016/j.jasrep.2016.10.001

Chapter 4, *Combusted bone: physical and chemical changes of bone during laboratory simulated heating under oxidising conditions and their relevance for the study of ancient fire*, presents a systematic study into the effects of heating in the presence of oxygen (i.e., combustion). While there is quite a body of work that deals with the characterisation of combusted bone, those studies tend to cover a limited temperature range and/or focus on only one or two analytical techniques. This chapter therefore presents the results of more extensive research on combusted bone, covering a broad temperature range and combining multiple analytical techniques. Taking the same experimental approach and using the same analytical techniques as those presented in Chapter 3 allows for seamless comparison of the two datasets. The study provides further details on the difference between charring and combustion, as well as additional parameters to distinguish the two heating conditions. The resulting reference dataset and analytical toolkit can be applied to archaeological heated bones from different ages and contexts.

Published as: *van Hoesel, A., Reidsma, F.H., van Os, B.J.H., Megens, L., Braadbaart, F., 2019. Combusted bone: Physical and chemical changes of bone during laboratory simulated heating under oxidising conditions and their relevance for the study of ancient fire use. Journal of Archaeological Science: Reports 28, 102033.* https://doi.org/10.1016/j.jasrep.2019.102033

Chapter 5, *Laboratory-based experimental research into the effect of diagenesis on heated bone: implications and improved tools for the characterisation of ancient fire*, presents the first systematic study into the effect of diagenesis on charred and combusted bone. The study is

based on extensive experimental work, rather than context-specific archaeological samples, and focusses on the effect of pH conditions (i.e., acidic, neutral, and alkaline) as a proxy for different geochemical settings within depositional environments. The study builds upon Chapter 3 and 4 by taking the reference data presented there as the unexposed (i.e., unweathered) counterpart of the artificially weathered samples. The results show that diagenesis has a substantial impact on the preservation potential of heated bones in the archaeological record, as well as on the reliability of the analytical techniques used to reconstruct past heating conditions. The study provides strong evidence that diagenesis should be explicitly considered in archaeological fire studies, and that existing studies may need to be revised. In addition to insight into the effect of pH exposure and its underlying processes, this chapter provides reference data and an analytical toolkit for the study of archaeological heated bones from different ages and contexts. Since the data itself is not context-specific, a reference dataset can be chosen that best matches the depositional conditions of the archaeological material under study.

Published as: *Reidsma, F.H., 2022. Laboratory-based experimental research into the effect of diagenesis on heated bone: implications and improved tools for the characterisation of ancient fire. Sci Rep 12, 17544.* https://doi.org/10.1038/s41598-022-21622-5

Chapter 6 moves the focus back to the archaeology and presents the discussion and conclusions of the thesis research. The chapter outlines the main contributions to the field. This is followed by a discussion on how the generated laboratory data relates to other variables and fits into real-world scenarios and the reality of the archaeological record. Steps for the application of the thesis results in archaeological research and method development are presented as well. In addition, Chapter 6 discusses the implications of the research for different aspects of the study of Pleistocene fire use, including the identification and preservation of fire proxies, identifying variation in use, and the reliability of analytical techniques. Finally, the chapter presents future directions for the field and some concluding remarks.