‘What to Cook and How to Cook It’: Understanding the Interplay of Subsistence, Ceramic Technology, and Local Environments in the Emergence of Pottery in Northeast Asia Through Residues

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A thesis submitted for the degree of
Doctor of Philosophy
Trinity 2022
Word Count: 75,802

Table of Contents

ACKNOWLEDGEMENTS **4**

TABLE OF ABBREVIATIONS **5**

ABSTRACT **6**

INTRODUCTION **7**

CHAPTER 1 BACKGROUND **11**

1.1 ARCHAEOLOGY BACKGROUND – ARCHAEOLOGY OF NORTHEAST ASIA AND POTTERY EMERGENCE 11

1.1.1 Pottery *11*

1.1.2 Hunter-gatherers *16*

1.1.3 Subsistence and Domestication *21*

1.1.4 The Neolithic of Northeast Asia *24*

1.1.4.1 The Japanese Archipelago 28

1.1.4.2 The Russian Far East 30

1.1.4.3 The Korean Peninsula 32

1.1.5 The Archaeological Record of Northeast China *37*

1.1.5.1 The Palaeolithic Period in Northeast China 37

1.1.5.2 The Neolithic Period in Northeast China 41

1.1.5.3 The Houtaomuga Site 42

1.1.5.3.1 Local Environment 47

1.1.5.3.2 Settlement Patterns and Site Distribution 49

1.1.5.3.3 Subsistence Strategies 55

1.1.5.3.4 Lithics 60

1.1.5.3.5 Human Remains 61

1.1.5.3.6 Pottery 64

1.1.6 Residue Analysis on Neolithic Pottery in Northeast Asia *65*

1.1.7 Summary *69*

1.2 ANALYTICAL BACKGROUND – METHODS FOR THE CHARACTERIZATION OF ARCHAEOLOGICAL LIPIDS 71

1.2.1 Lipids *72*

1.2.2 Preservation of Residues *73*

1.2.3 Extraction *74*

1.2.3.1 Extraction with Liquid Solvents 74

1.2.3.1.1 Traditional Extraction Procedures 75

1.2.3.1.2 Acidified Methanol Extraction 77

1.2.3.2 Supercritical Fluid Extraction 79

1.2.3.2.1 History & Principals 79

1.2.3.2.2 Parameters 80

1.2.3.2.3 Commercial and Industrial Applications 82

1.2.3.2.4 Cultural Heritage Applications 83

1.2.4 Chemical and Isotopic Analysis of Residues *85*

1.2.4.1 Chromatography 85

1.2.4.2 Mass Spectrometry 85

1.2.4.3 Gas Chromatography and Combustion Isotope Ratio Mass Spectrometry 86

1.2.4.4 Liquid Chromatography 87
1.2.5 Radiocarbon Dating and Pottery 88
1.2.6 Organic Residue and Starch Grain Analysis in China 91
1.2.7 Conclusion 94

CHAPTER 2 METHODS AND MATERIALS 95

2.1 Analytical Strategy 95
2.2 Methods 97
2.2.1 Sample Preparation 97
2.2.2 Extraction Methods 98
2.2.2.1 Liquid Extraction 98
2.2.2.2 Acidified Methanol Extraction 98
2.2.2.3 Supercritical Fluid Extraction 99
2.2.3 Analysis 101
2.2.3.1 GC-MS 101
2.2.3.2 GC-C-IRMS 102
2.3 Materials 103
2.3.1 Solvents, Reagents, and Standards 103
2.3.2 Reference Materials for Method Validation 103
2.3.2.1 Modern Experimental Lipids 103
2.3.2.2 Modern Clays Pellets 103
2.3.3 Archaeological Materials of Known Organic Composition Used for Method Validation 104
2.3.4 Houtaomuga Pottery Samples 104
2.3.4.1 Houtaomuga Phase 1 Pottery 105
2.3.4.2 Houtaomuga Phase 2 Pottery 108
2.3.4.3 Houtaomuga Phase 3 Pottery 110
2.3.4.4 Houtaomuga Phase 4 Pottery 112

CHAPTER 3 RESULTS OF ANALYTICAL WORK 115

3.1 Method Development Results and Discussion 115
3.1.1 Acidified Methanol Extraction Procedure 115
3.1.2 Supercritical Fluid Extraction Optimisation 116
3.1.3 Gas Chromatography-Mass Spectrometry Optimisation 122
3.1.4 Testing GC-MS methods on samples from Houtaomuga 136
3.1.5 Conclusions 138
3.2 Results of Lipid Residue Analysis on Pottery from Houtaomuga 139
3.2.1 Houtaomuga GC-MS Results 139
3.2.2 Resinous Substances 141
3.2.3 Houtaomuga GC-C-IRMS Results 142
3.2.4 Preservation 144
3.2.5 Contaminants 145
3.3 Summary 146

CHAPTER 4 DISCUSSION OF ARCHAEOLOGICAL RESULTS 148

4.1 Changes over time 148

4.2 Changes between features 152
4.3 Comparison with previous data 154
4.4 Resins, Hydrocarbons, Diterpenoids, and Lithics 157
4.5 Terrestrial Animals 159
4.6 Aquatic Resources 160
4.7 Plants 161
4.8 Discussion of Results in Broad Archaeological Context 162
4.8.1 Comparison of results at Houtaomuga with Neolithic sites in the Amur River Region of Russia 162
4.8.2 Comparison between Neolithic sites in Mainland East Asia and on the Japanese Archipelago 167
4.8.3 Hunter-gatherer pottery use in relation to results at Houtaomuga 173

CHAPTER 5 CONCLUSION 178

5.1 Implications of Non-destructive Residue Analysis 178
5.2 Overall Implications of Results from Houtaomuga 180

REFERENCES 182

APPENDIX I: LIST OF ALL SAMPLES II

APPENDIX II: ALL COMPILED IRMS DATA FROM NORTHEAST ASIA XIII

APPENDIX III: REFERENCE IRMS DATA FROM NORTHEAST ASIA USED FOR COMPARISON WITH ARCHAEOLOGICAL RESULTS LXV

APPENDIX IV: LIPID COMPOUND AND CONTAMINANT CONCENTRATIONS BY OCCUPATIONAL PHASE LXVII

APPENDIX V: LISTS OF SAMPLES WITH INADEQUATE PRESERVATION FOR ANALYSIS LXXII

APPENDIX VI: IRMS DATA SPLIT BY FEATURE AND OCCUPATIONAL PHASE LXXVI

APPENDIX VII: R INTERFACE CODE LXXVIII

APPENDIX VIII: GC-MS METHOD TESTING CALCULATIONS LXXXII

APPENDIX IX: DETAILS OF QUANTITATION AND BLANKS PROCEDURE FOR GC-MS LXXXVI

APPENDIX X: TABLES FOR FIGURES 3.16-3.22 LXXXVIII

APPENDIX XI: SAMPLES ANALYSED IN THIS THESIS XCIV

Acknowledgements

This thesis could not have been possible without St Cross College and the Division of Social Sciences at the University of Oxford for their generous financial support of my research. I would also like to thank the School of Archaeology and Santander (through academic travel awards at the University of Oxford) for funding various stages of travel and lab expenses required to undertake this research

To my dear supervisors, Anke Hein and Thibaut Devièse, thank you so much for your continuous support from the initial stages of this work to the final product. Your personal and professional advice and guidance will always be remembered and appreciated. I could not have asked for a better team to work with during my DPhil and I appreciate you every day.

Many thanks as well to Wang Lixin 王立新 and Pauline Sebillaud for all your gracious support with this research and with your help during my time in China. Isotopic analysis undertaken in this work would not have been possible without generous assistance from Sven Isaksson and the archaeological science lab at the University of Stockholm. Thank you to Pauline A. Duval for helping me with this work and for being a lovely friend as well.

I would like to specifically thank Vincent J. Hare and Roger Wilson for supporting my work with method validation and testing using archaeological samples. I would also like to thank much of my DPhil cohort and co-workers from the Oxford Radiocarbon Unit and Research Lab for Archaeology and the History of Art. Many of you helped me navigate this process, hunt down literature, fix equipment, and keep my sanity for many years.

Lastly, I would like to thank all my friends and family. To my wonderful parents, I would have never gotten here without everything you have given me, including my work ethic, my ambition, my passion, and initiative. I appreciate everything you do for me from every phone call of mine you have answered to every plane ticket home you have bought me and the long list of everything in-between. I would like to thank my siblings Sara and Ben for giving me my sense of humour and the ability to laugh through hard times. I would also like to thank my partner, Phil Ford. Without you I would never have gone to Oxford and would not be where I am today. Thank you for always loving me when I am struggling and supporting me in everything I do. I am a stronger person because of all of you, and without you all this thesis simply would not exist.

Table of Abbreviations

Abbreviation	Explanation
AMS	Accelerator mass spectrometry
BF_3	Boron trifluoride
BP	Before present
BSTFA	N,O-Bis(trimethylsilyl)trifluoroacetamide
CH_2Cl_2	Dichloromethane
$CHCl_3$	Chloroform
CO_2	Carbon dioxide
DAG	Diglyceride
EFA	essential fatty acids
ESI-Q-TOF	Electrospray-ionisation quadrupole time-of-flight mass spectrometry
FAME	Fatty acid methyl esters
FFA	Free fatty acid
GC-C-IRMS	Gas chromatography combusions isotope ratio mass spectrometry
GC-MS	Gas chromatography mass spectrometry
H_2SO_4,	Sulfuric acid
HPLC	High performance liquid chromatograpgy
IS 1	Internal standard 1 - Tridecanoic acid TMS
IS 2	Internal standard 2 - Hexadecane
KOH	Potassium hydroxide
MeOH	Methanol
MS	Mass spectrometry
P_C	Critical pressure
RT	Retention time
SFC	Supercritical fluid chromatography
SFE	Supercritical fluid extraction
TAG	Triglyceride
T_C	Critical temperature
TLC	Thin layer chromatogprahy
TMS	Trimethylsilyl group

Abstract

Since the early 2000's, archaeological excavations in the Nenjiang River Basin have led to the discovery of some of the oldest pottery assemblages in northeast China. The main objective of the DPhil research presented here is to understand what role vessel technology played in human lives over an extended time span regarding diets, food preferences, and human environmental interactions in this region. Pottery analysed in this thesis comes from the site of Houtaomuga 后套木嘎 located in the Lower Nen River Valley on the Song-Nen Plain, Jilin province northeast China. Using molecular and isotopic analysis, this thesis characterised lipids extracted from archaeological pottery using advanced analytical methodologies to understand pottery use at the initial implementation of the technology and over the course of the Neolithic. Results of lipid residue analysis presented as a part of this research were also described in context with other regions in northeast Asia, such as Russia's Far East and Japan, which also have pottery bearing hunter-gather sites from the Neolithic which are contemporaneous.

Before turning to the analysis of the archaeological material from Houtaomuga, this thesis undertook a method optimisation to understand the efficiency and limitations of current techniques. This method development ensures that the method of analysis was adequate to answer proposed research questions prior to sampling and will improve upon methods for lipid residue analysis which are currently standard practice within archaeological science. As a result, this thesis presents the first extensive application of non-destructive supercritical fluid extraction (SFE) of ancient lipid residues followed by isotopic characterisation. Results indicate this novel method workflow is comparable to current methods which are highly destructive to ceramic artifacts. By using this workflow, it was determined that pottery at Houtaomuga was primarily used for cooking aquatic molluscs and C_3 plant resources. While other proxies indicate that human diets at Houtaomuga were highly diverse, pottery use was highly specified for all four phases of Neolithic occupation of the site, suggesting people preferentially chose certain foods or dish types to prepare in vessels.

Introduction

Food is an integral thread in the fabric of human life. What people eat reflects the relationship between metabolic necessity and the external world; it is the intersection of biological imperatives and cultural habits (Hedges, 2009). Food is limited by what is physically available in the surrounding environment, however, biological availability and environmental carrying capacity can be drastically altered by human intervention (Chen, 2012). Food and food preferences have "embedded political, socioeconomic, and cultural meaning", as concepts of what consumers and producers determine as "good to eat" are socially constructed (Chen, 2012, p. 1; Smith, 2006). Eating together, sharing food, provides important "social clue", and cuisines are closely connected to group identities with shared preferences connecting both individuals and groups. Food can also be shared with ancestors, ghosts, gods, and the dead during various types of religious ceremonies such as funerals and offerings, giving food preparation and presentation an additional and even deeper meaning in certain contexts (Tierney and Ohnuki-Tierney, 2012). During much of prehistory and history and to a certain extent into the present, pottery vessels have been crucial containers for preparing, storing, and/or serving food on a broad range of occasions. Therefore, it is vital to understand pottery use as it relates to subsistence to better understand past diets, human-environment, human-human, and even human-non-human relationships, and cultural choices.

Food and natural environments are tightly intertwined and together determine cooking techniques develop in tandem with both. Certain resources that may be biologically available may not be edible to humans without processing, the most prominent example identified in archaeological contexts being acorns. Meanwhile, humans may not be able to absorb certain nutrients from food in its uncooked form, for instance, certain roots and leafy greens. Developing a special tool for cooking such as pottery containers, can allow people to better adapt to their environment and use more resources available to them (Grillo, 2014). Understanding pottery function can thus aid researchers in understanding how humans interact with their surroundings and adapt to various climates. It is important to note that pottery is not a necessity for cooking and a wide range of resources can be cooked over an open flame or through other methods. Therefore, researchers need to be mindful of a broad range of potential food preparation methods when considering the reasons for the emergence of pottery and its

potential use in relation to cooking and eating. Nevertheless, in the majority of places around the world, pottery is crucial in food preparation and presentation. Thus, investigating pottery in the archaeological record is vital for understanding past human diets and their relationship with their environments.

Pottery function has been defined by Rice (1996, p. 139) as the broad roles or activities or capabilities of ceramics as containers (for storage, processing, and transport) or structural materials (i.e. bricks), while "use" refers to the specific ways in which a vessel was brought into service for a particular purpose. Understanding the previous use of a vessel through empirical means can help archaeologists understand it as a means for acquiring a nutritional resource (Hedges, 2009). Moreover, food can be used as a proxy to better understand human cultural affiliations because ideas surrounding food and consumption are socially constructed (Chen, 2012; Mintz, 1996), and this approach may be more straightforward than the relationship between pottery style and identity in some regions and contexts.

Pottery appeared in northeast Asia at the beginning of the Holocene. Thereafter, its use by hunter-gatherers quickly became widespread for reasons which are not fully understood yet (Wang and Sebillaud, 2019). This innovation in technological application suggested a significant change in human economic behaviour, subsistence, and human-environment relations. Since 2013, archaeological scientists have attempted to understand these changes in human behaviour in the region by using residue analysis to study how this new pottery technology was used. These scholars have focused on hunter-gatherer pottery use in Japan, Russia, and Korea. Northeast China also saw the advent of pottery use amongst hunter-gatherers at this time, but researchers have not previously used residue analysis extensively to study this pottery.

Geographical gaps in our understanding of pottery use within a region as large as northeast Asia is not uncommon. Indeed, there are some limitations of residue analysis that make its applicability in many areas difficult or impossible. This thesis is one of the first extensive applications of organic residue analysis of samples from prehistoric China. Previous research on neighbouring regions with similar hunter-gatherer pottery such as Japan and southern Siberia have explored the function of similar pottery types, however, the relationship between these regions and northeast China is not fully understood (Lucquin et al., 2016; Shoda et al., 2020). While this research will by no means explicitly define interactions between these

regions or unequivocally determine their cultural affiliations, it will shed light on similarities and differences between these interactions regarding pottery function. This is particularly useful for our understanding of how pottery use can change or persist across varying environments, and thus inform future interdisciplinary work.

To examine pottery use, scholars often examine vessel forms; however, this can only indicate potential intended uses. Insights into actual use can be gleaned from use-wear in combination with experimental and ethnographic work, and especially residue analysis. The latter technique provides insights into vessel function (e.g. its usage as a vessel for liquid storage, boiling, or roasting), but also into the types of foods prepared or stored. Residue analysis is, therefore, an incredibly powerful tool that can allow inferences not just on object usage but also on subsistence practices and food preparation techniques and customs. Despite its general potential, there are still concerns with this technique of analysis, meaning that much work still needs to be done in method development and actual analysis alike. This thesis, therefore, combines two main aims: first, method development in residue analysis; and second, the application of this technique on a specific body of material from early Neolithic northeast China. In so doing, it sheds light on the usage of the earliest pottery vessels in the region and thus some of the motivations behind developing pottery technology in the first place.

Archaeological researchers have long recognised the importance of diet and pottery and, in the last twenty years, have used residue analysis to investigate these issues (Roffet-Salque et al., 2017). A selection of this research has focused on small sample sizes or specific specialised pottery forms (Dunne et al., 2019b; Leclerc et al., 2018), and in some instances, extensive residue analysis has been conducted on pottery to understand broad dietary changes and its relationship to changes in pottery use. This work has been undertaken selectively in certain regions – primarily, Europe and Asia (Craig et al., 2013; Cramp et al., 2014; Hendy et al., 2018; Shoda et al., 2017), but is continually expanding (Grillo et al., 2020). The method remains underutilised in some regions because of a variety of factors. Primarily, the method remains destructive and expensive. To help mitigate some of these negative factors of residue analysis, this thesis addresses methodological advantages and limitations for analysis to test such hypotheses about ceramic form/function relationships, cultural affiliations, and diet.

This thesis characterised lipid residues from Neolithic pottery using molecular and isotopic analysis to understand pottery use at the initial stages of the technology and over the course of

the Neolithic. Focusing on the site of Houtaomuga 后套木嘎 in northeast China, this research sampled pottery from all phases of Neolithic occupation at the site to understand what role vessel technology played in human lives over a long time span regarding diets and human environmental interactions. Additionally, this research took into account surrounding areas of northeast Asia where similar analysis has been undertaken to aid in future comparative work looking at pottery use at a larger scale. The formal report on the site of Houtaomuga (3 volumes book in prep) has not been published yet and includes ongoing research regarding the faunal and plant remains, as well as lithic research.

Before turning to the analysis of the archaeological material in question, this thesis conducted a method optimisation to understand the efficiency and limitations of current techniques. This method optimisation was necessary because powdering pottery samples was not authorised for this study. To overcome this, this study harnessed non-destructive residue analysis using supercritical fluid; a method that is still in its infancy and has never been applied to pottery of this age. Method development will ensure the method of analysis is adequate to answer proposed research questions. A variety of lipid classes and ceramic sherds with different fabric types, porosities, and residues must be examined, as the contents of pottery from northeast China are unknown and previous research in the area has been limited to a small amount of material (Kunikita et al., 2017). This process will ensure residue analysis methods chosen are appropriate for the complete extraction and characterisation of former ceramic contents.

Chapter 1 Background

This section presents background information about the archaeology in the research area as well as the currently applied chemistry methods used and developed in this thesis. A brief background of the theoretical considerations as part of this research are also presented.

1.1 Archaeology background – Archaeology of Northeast Asia and Pottery Emergence

1.1.1 Pottery

This thesis focuses on the initial and continued use of pottery by hunter-gatherers in northeast Asia. The emergence of pottery and its initial usage have long been debated. Until the late twentieth century, archaeologists largely believed that pottery originated from a single place in southwest Asia as a product of the agricultural revolution and theorized that it spread within a wave-of-advance model (Jordan et al., 2016). This theory arose from early discoveries in the Levant where pottery production began one thousand years after the development of settled agricultural villages and has been repeatedly cited as evidence of the inevitable coexistence of these technologies (Shelach-Lavi and Tu, 2017). However, since the association of early pottery on the Japanese archipelago with non-agricultural groups in the late twentieth century, interest in hunter-gatherer pottery has come to the forefront of archaeology (Shelach-Lavi and Tu, 2017). Since this discovery, archaeologists have identified evidence for pottery making among hunter-gatherers at a considerable number of sites across East Asia.

Based on such discoveries, we now understand the Neolithic period globally, including in East Asia, to be a "patchwork" of regional variations, rather than a single universal process of set innovations (Yanshina, 2019). As a result, scholars often define Neolithic East Asia—including Japan, Korea, Russia's Far East, and northeast China—as beginning with the emergence of pottery, regardless of the presence or absence of additional "Neolithic" defining features denoted by those working on other regions of the world; including sedentism, animal husbandry, and agriculture (Kuzmin, 2006; Underhill, 1997). While it has been accepted that

these characteristics do not often develop simultaneously, pottery is often still identified by archaeologists as a marker for social change (Hommel, 2018). When using the term Neolithic in this research, I am referring to the defined time period between about 13,000 cal BP to 5,000 cal BP of human behavioural changes in northeast Asia, rather than discussing broader trends of changing lifeways across Eurasia or imposing the idea of a universal "Neolithic revolution" and all of its implications. This specific time frame is the defined time period previously established by scholars working in the region and defined as the Neolithic. I choose to follow Barnes' (2019) notion that Neolithic and Neolithization are not useful for cross-cultural comparisons, and I do not aim to undertake any such comparisons here.

Pottery in East Asia appears around a similar time in many regions, but due to regional variations in decoration and form, the overall consensus among researchers is that this pottery was developed independently among several hunter-gatherer groups (Hommel, 2014; Jordan et al., 2016; Jordan and Zvelebil, 2010; Kuzmin, 2017; Wang and Sebillaud, 2019). Unfortunately, there is not enough information to confirm this hypothesis at present. The earliest of these sites are in southern China, where excavations at cave sites have yielded Late Pleistocene deposits of thick low-fired ceramic vessels. One of the most famous among these sites is Xianrendong 仙人洞 in Wannian district, Jiangxi province, where researchers found pottery dating to 20,000 years cal BP (Cohen et al., 2017; Jiangxi Province Museum, 1976). At the end of the 1970s, pottery dating over 10,000 BP was discovered in Russia's Far East and Japan, followed by similar discoveries across southern and northern China (Wang and Sebillaud, 2019; Yanshina, 2017). Early pottery in North China differs from the South. The earliest pottery in North China is younger and dates from around 11,500 cal BP to 11,000 cal BP (Shelach-Lavi and Tu, 2017). Some notable sites in North China are Nanzhuangtou 南庄头 in Hebei Province and Donghulin 东胡林 in the Beijing area, both of which have similarities to pottery sites in northeast China, the main focus of this thesis (Hao et al., 2008; Hebeisheng Wenwu et al., 2010). This pottery is fired below 900 degrees and often comb decorated and persists at sites in the North until the emergence of larger sedentary regional communities (~8,000 BP) including those around Xinglongwa 兴隆洼, Peiligang 裴李岗, Cishan 磁山, Houli 后李, and Dadiwan 大地湾 sites (Shelach-Lavi and Tu, 2017). Due to the large discrepancy in dating, archaeologists do not consider these pottery styles from hunter-gatherer pottery in northern and southern China to be from the same horizon, and there is little extant evidence regarding their association.

Scholars have suggested different hypotheses for the driving forces behind the emergence of pottery in various regions of the world and East Asia specifically. In North Africa, early pottery in the Green Sahara or the Sahel is associated with the boiling of newly abundant, wild cereal grains to make them more digestible (Huysecom et al., 2009). As new data emerges, it will be interesting to see similarities and differences in motivations behind developing and adopting pottery technology in this region versus East Asia, where cultural, economic, and environmental factors differ greatly. In East and Central Asia, the most common hypothesis is that the technology was used to cook food types that may require long cooking periods, including acorns and shellfish (Jordan et al., 2016). Using a pot for cooking is less labour intensive than using baskets with a hot stone boiling method and in some instances, requires less skilled labour and time than woven baskets (Aikens and Lee, 2013). Researchers have also suggested that pottery possibly emerged as a tool for past people to distinguish between communities, groups, and/or individuals or as a prestige good which signifies social rankings (Hommel, 2014). Regardless of which hypotheses individual researchers subscribe to, the majority agree that the emergence of pottery was essential for subsequent agricultural and social changes in East Asia through population growth by processing a diverse set of foods, including more plants, that were abundant after the beginning of the Holocene (Aikens and Lee, 2013, p. 47).

Understanding initial innovation or adoption of ceramics sheds light on these later developments; however, we must remember the inherent link between technology and other cultural and economic factors, which may determine if a technology is useful and/or worthwhile to create or adopt (Jordan et al., 2016). For example, we cannot determine a group of people adopted pottery technology solely because it made one of their cooking processes easier. This is because there could be other considerations a group must take into account when making this decision on technology use, i.e., they might prefer the taste of the more difficult cooking process or might not have easy access to raw material for pottery production. The adoption or advancement of pottery technology is the result of a series of decisions made by a group of people with multiple contributing factors.

The functional aspect of pottery is a central part of this decision-making process; however, the function has yet to be deeply explored based on empirical evidence in northeast China. For the most part, scholars have linked the emergence of pottery in this area to the changing ecology

during the Pleistocene to Holocene transition. Generally, climate warming during this period saw a decline in large mammal prey and the further exploitation of alternative plant and animal food resources including fish and acorns, which in turn led people to explore new methods of food preparation (Li et al., 2017; Liu and Chen, 2012; Zhushchikhovskaya, 2009). However, evidence for acorn processing is limited to sites in the Japanese Archipelago and is not found in mainland areas of Russia and northeast China, meaning the reason for use likely differs between these regions (Wang and Sebillaud, 2019). The presence of fish hooks and net sinkers at sites in this area suggest a substantial reliance on fishing, and this is supported by carbon and nitrogen stable isotope residue analysis of charred food residues across the Japanese archipelago (Craig et al., 2013; Shoda et al., 2020; Wang and Sebillaud, 2019). Overall the relationship between climate change, ceramic technology, decreased mobility, and population growth needs to be investigated further (Hommel, 2014). Such investigations are incomplete without ceramic residue analysis.

There is evidence of charring in early pottery in northeast Asia, which has been assumed to be associated with cooking (Wang and Sebillaud, 2019), however, while useful for long-term simmering and boiling, pottery is not essential to cooking. Hot-stone technology – the process of adding hot rocks into a basket of water to raise the temperature and cause it to simmer – is a common and easy cooking technique employed around the globe (Driver and Massey, 1957; Nelson, 2010). This is also known as indirect boiling. The main benefit of a ceramic pot in cooking is its ability to sit on a hot fire for prolonged periods (Hommel, 2012). Under certain circumstances where long-duration cooking is required, such as gelatinizing indigestible starches, denaturing toxic chemicals, rendering fat, oil, and grease, the time-saving benefit of pottery technology would be substantial (Arnold, 1985; Hommel, 2014; Reid, 1984; Rice, 1999). Hot-stone boiling is also less energy effective than direct boiling in a pot (Shelach-Lavi and Tu, 2017); however, it would be simplistic to assume past humans only strove for energy efficiency in their everyday activities.

Ceramic vessels are also beneficial for “moist” culinary technologies, namely cooking and steaming. These food preparation methods also require more time and allow for juice/fats to be preserved in the dish instead of being lost in the fire and enable the cooking of scraps and leftovers from previous meals (Hommel, 2014). This also means it extracts the maximum nutritional value and facilitates bulk cooking of small food resources such as small fish or millet and allows for the extraction of nutrient-rich marrow. Moist technology also creates a

nutrient-rich broth for the elderly and infants (Hommel, 2014). Lean meats often consumed by hunter-gatherers are deficient in essential fatty acids (EFA), especially Omega 3 fatty acids like linoleic acid (Speth and Spielmann, 1983). Adults usually have more stored up fats in their body which can provide these nutrients, however, infants lacking these fat stores need to obtain EFA through other means (Speth and Spielmann, 1983).

Indeed, forager communities could have preferentially chosen pottery, over other cooking methods, because of its ability to retain animal fats through moist cooking technologies for calorie retention benefits. Cooking increases the energy gained per calorie while increasing the digestibility of lipids specifically (Groopman et al., 2015). An analysis of human teeth from northeast China has shown a low frequency of dental caries, meaning diets were high in protein and low in carbohydrates (Zhang et al., 2016). Lack of fat in high protein diets causes significant health issues, which can be fatal (Speth and Spielmann, 1983). Any increase in fat procurement would have been beneficial to these hunter-gatherer populations significantly. Moreover, perhaps people preferred to retain fats through moist cooking methods for taste purposes, however, this cannot be ascertained archaeologically using residue analysis alone.

Additionally, unglazed ceramics retain fats and thus repeated use of the pot can also provide essential fat to the diet. These fats retained in the pottery may also add to the overall flavour of the dish; for instance, not cleaning a pan between cooking meat and creating a sauce or cooking starches and other vegetables for example. Scholars have demonstrated the impact of fat on flavour and how fats interact with aromatic compounds to alter the taste of most dishes in a way that is difficult to reproduce with other compounds (Hatchwell, 1996). However, this is difficult to identify archaeologically. With these ideas in mind, the adoption of pottery may be understood as the result of human food preferences and choices. There are multiple ways to cook most food types, however, pottery is specifically advantageous in some specific ways regarding fat retention, and lipid residue analysis can support our understanding of such choices.

Archaeologists broadly have discussed this notion of food choice; however, those researching hunter-gatherer subsistence practices have focused less on this for several reasons. First, some archaeologists hold antiquated assumptions about hunter-gatherers and their lives, which does not include people undertaking activities outside of those necessary for survival. In this instance, choosing a cooking process for taste preferences rather than food bioaccessibility and

nutrition might be seen as an activity that is beyond the necessary. As Sherratt (1999, p. 13) noted, the use of the word 'subsistence' by archaeologists often unintentionally implies the bare minimum needed by people to survive. Secondly, archaeological research in recent years has attempted to be more scientific and consistent in analysis, and ideas of cultural preference are often impossible to test using scientific methods. Despite these inherent limitations, researchers have considered food choice in certain contexts. Fuller and Rowlands (2011) have discussed food preferences as the result of ritual and cosmological ideas, food selection, and technologies available for preparation and consumption resulting in long-term continuity of "food-zones" which have developed over long periods. Residue analysis on pottery aids our understanding of these food zones' development through insight into food choice and technology for production and consumption, as cooking likely is heavily influenced by technology available for the process (Fuller and Rowlands, 2011). Overall, the way we research food and pottery in the past and in northeast Asia specifically is heavily influenced by underlying assumptions about hunter-gatherers and the historical context through which the field of archaeology developed.

1.1.2 Hunter-gatherers

In the Neolithic period of northeast Asia, people mainly relied on a hunting-gathering-fishing economy and were highly mobile. Those researching hunter-gatherers in archaeology and anthropology have approached lifeways, including pottery use, in different ways, which it is necessary to introduce here. The term hunter-gatherer is used to provide a basic means of "classifying human cultural diversity according to economic and subsistence criteria" (Jordan and Cummings, 2014, p. 2). I use the term in this way to describe only a forager type economy. Many scholars have fairly criticized this definition for being reductive and not capturing the wide diversity within this classification. Unfortunately, there is not a term that accounts for this diversity that is widely accepted within the field and the terms hunter-gatherer or hunter-gatherer-fisher are used here for clarity and continuity with other research.

Until about 12,000 years ago, human populations only used a form of hunting and gathering as a mode of subsistence (Lane, 2014). While well-defined research and targeted analysis of hunter-gatherers did not become popularised in archaeology until the 1960s, the concept emerged slowly earlier and was fully realised in the 18th century (Garvey and Bettinger, 2014). In this period, western cultures considered the economy to be a primary driving force of human

interaction, and thus they drew links between subsistence activities and other ways people lived their lives (Jordan and Cummings, 2014). Following the 19th century, unilinear evolutionary theories consisting of highly generalized and finite categorizations with unilateral movement described hunter-gatherer groups in archaeology as belonging to the lowest rung of an often three-tiered system, i.e. the "savagery" stage of L. H. Morgan's schema of evolution (Jordan and Cummings, 2014; Morgan, 1877). The primary notion was that more "advanced" groups from the highest form of society, "civilization", were unconstrained by environmental uncertainties influencing daily food procurement freeing time for intellectual and recreational pursuits (Bettinger, 1991). This notion has been called into question as research shows hunter-gatherers often have more leisure time than agriculturists in some cases (Dyble et al., 2019). This concept of an "advanced" society directly implies growth from a "primitive" one – a highly problematic colonialist notion. Unilinear evolution has been heavily criticized (i.e Barnard, 1983), since its emergence for its ridged structure, one-way movement along a predetermined trajectory, and lack of accounting for human variability; however, its importance in developing the ideas and terminology used in hunter-gatherer studies requires mention, particularly considering that narrow understandings of the term rooted in this framework persist.

In the 20th century, archaeologists and anthropologists began to consider the importance of the environment for hunter-gatherers' lives and livelihoods. This change in thinking was likely brought on by reports of hunter-gatherers in the Americas thriving in harsh environments which settlers struggled to overcome (Bettinger, 1991). This first school of American anthropological thought, historical particularism, reflected the influence of the environment on cultural adaptions with the recognition of such groups being ecologically savvy (Garvey and Bettinger, 2014). Headed by Franz Boas, this framework considers cultures in the context of their own histories and environments and does not equate cultural similarities with comparable development (Boas, 1896; Garvey and Bettinger, 2014). This framework encouraged an understanding of human groups in isolation and in its own historical and environmental context in contrast to previous ideas of evolutionary development which could be applied to any human group in any context.

As a result of historical particularism, Julian Stewart's cultural ecology approach aimed to study human adaptions to their physical environment, with a particular emphasis on subsistence technology, where the environment is seen as limiting the trajectory of cultural adaptions

(Stewart, 1972). Stewart viewed technology as a result of historical circumstances and as a powerful counterbalance to environmental constraints determined. However, in practice, cultural ecologists often interpreted technology as adaptive responses to their environment, which is rightfully critiqued as environmentally deterministic (Garvey and Bettinger, 2014). In contrast with some of his predecessors, Stewart believed fully understanding these adaptions required cross-cultural comparisons, which resulted in cultural ecology producing a contentious model for cultural evolution (Bettinger, 1991). The framework has been highly criticized in archaeology and anthropology for being environmentally deterministic and suggesting culture evolves along a predictable trajectory, nevertheless it has had a huge impact in the field, specifically within our understanding of hunter-gatherers. It led to the majority of research on hunter-gatherers in the 20th century focusing on adaptions in this way. Cultural ecology gave rise to neofunctionalism[1] in hunter-gatherer research, which emphasizes group over individual selection, and failed to properly support links between adaptions and the function they supposedly performed (Garvey and Bettinger, 2014). The wave of new archaeology in the mid-20th century directly drew on themes of cultural ecology and aimed to understand humans' relationships and adaptions to their environments, specifically through scientific hypothesis testing. New archaeology aimed to describe the benefits of cultural change without researching the mechanisms or results of such changes.

The influence of cultural ecology and processual archaeology is noticeable in research on the Palaeolithic and Neolithic in East Asia. Most research in the field suggests the fluctuating climate during this time period resulted in dietary changes and technological innovation (e.g. Cohen, 2013). Critics might suggest this interpretation is environmentally deterministic and clouded by ideas of the supreme environmental adaptability of hunter-gatherers held by most researchers in the field as a direct result of historical approaches of cultural ecology and processualism. I find it important to mention such concerns, however, I think this is the result of little additional evidence rather than a deliberate attempt to describe past peoples in a reductive way. Compared to most time periods in many regions in the world, the Neolithic of northeast Asia has been less systematically researched and there are still new sites and surveys being completed. We know less about human burials and settlement patterns throughout this period than we do about climatic fluctuations because environmental data is abundant while

[1] All such discussions of human adaptions are inherently functionalist in nature. See Garvey and Bettinger (2014) for further discussion beyond the scope of this work.

archaeological data is still catching up. This thesis will add to the discussions about past human behaviour and technological changes in the context of their surrounding environment. I aim to avoid cultural ecological and environmentally deterministic undertones which are present in current research, as I do not believe there is enough evidence at present to support the hypothesis of a correlation between climatic shifts and pottery adoption. While we know the climate changed significantly after the Holocene, we may not know enough about what people were eating before this time to comment on how diets changed in light of this climate shift and with the development of pottery. I instead aim to describe pottery function along with human-environmental relations instead of these developments as adaptions or reactions to environmental changes.

My research in the following chapters further discusses prevailing hypotheses among scholars working in northeast Asia regarding the driving forces behind pottery introduction among these hunter-gatherer groups. These hypotheses are not made using faulty or incomplete evidence; however, I do not want to present their findings uncritically. Current scholars suggest that hunter-gatherers begin using pottery extensively due to increased use of aquatic resources in their diet and a decrease in large mammals. I believe this notion rests (unintentionally) on a causal fallacy where a sufficient condition for pottery adoption is being mistaken for a necessary condition. While an increased reliance on a different food type than previous years, like fish in this example, or broadening resource economy might result in the adoption of pottery technology, it alone does not provide sufficient cause. These hypotheses inherently imply that pottery is required to cook these aquatic products, however, there is little evidence to support this notion.

There are numerous incidences of hunter-gatherer groups that significantly changed their diets and ways of living when they change to a fishing-based economy and often do not use pottery. In the Upper Palaeolithic (~40,000-15,000 BP), fishing equipment appears in larger quantities on a global scale, and shell middens become more prevalent (Yesner et al., 1980). Binford (1968) noted a global trend at the end of the Pleistocene of human groups diversifying their diets and increasing the types of food resources utilized including aquatic resources. I often do not prefer to mention these types of global generalizing notions, as I do not find them particularly useful to discuss past populations when so many local variations affect human behaviour and diets. However, in this instance, I think it is useful to mention this suggestion of a global change in aquatic food use here to illustrate that the dietary shift we see in northeast

Asia during this period is not an isolated incident, but pottery production is. When reading most scholarly articles about this phenomenon, one might get the impression this change in northeast Asia caused this unique hunter-gatherer pottery adoption when this is far from the case. Specifically, the later portions of the Middle Stone Age (300,000 and 130,000 BP) in southern Africa is marked by microlithic use and reliance on aquatic resources very similar to northeast Asia, however, pottery technology is missing (Henshilwood and Lombard, 2014). Hunter-gatherer-fisher groups living in British Columbia did not adopt pottery technology in Antiquity (e.g. Prentiss et al., 2011). The North coast of modern-day Peru had a long period of an aceramic Neolithic, specifically with the Caral-Supe civilization, which relied very heavily on ocean and river fishing (Shady, 2006). At the end of the Pleistocene, hunter-gatherer groups on the Iberian Peninsula and southern coast of modern-day France without pottery also begin to rely more heavily on aquatic resources and their diverse diets include terrestrial mammals and marrow extraction (Bicho and Haws, 2008; Straus, 1991). This is a similar broad dietary spectrum as observed for people living in northeast China during the Neolithic, but again, pottery technology is absent (Kunikita et al., 2017; Merrett et al., 2016; Song et al., 2016; Wang et al., 2017). I am not presenting these examples to directly relate these different cultural groups to each other but instead suggesting that hunter-gatherers in northeast Asia are oversimplified in comparison.

I believe this faulty reasoning occurs partly due to the use of reductive terms in archaeology like hunter-gatherers. Hunter-Gatherers as an economic "type" show great amounts of diversity in socioeconomic practices (Yesner et al., 1980). The way we discuss and think about these groups might be different if they were classified in a more useful manner or we avoided classification altogether. Classifications of human groups based on economics can simultaneously highlight differences while concealing diversity within classifications (Grier, 2017). The development of the term "complex hunter-gatherers" in the late 20th century aided discussions of diversity within hunting and gathering groups, particularly those who relied on forager economies but had some type of more complicated social arrangements or lived in productive rather than marginal ecological niches (Arnold, 1993; Grier, 2017). However, Grier (2017, p. 17) points out that complex hunter-gatherers as a classification themselves are highly diverse and are not best characterised in such general and restrictive ways. Hunter-gatherers who use pottery have also been classified as complex hunter-gatherers (Rice, 1999, p. 1), and if we are going to use this term within the discipline, we should aim to further describe and highlight these complexities.

One might argue this reasoning is acceptable in archaeology due to the high variability and unpredictability of human behaviour across time and space. While this is important to take into consideration, we should aim to describe and understand such differences in detail rather than accept them as a necessary difficulty. For example, we could look at what cultural and environmental factors could be at play in northeast Asia during the time of pottery adoption that are different in other hunter-gather-fisher populations from various regions. There are clear differences in human behaviour between northeast Asia during the Holocene-Pleistocene transition and the regions mentioned above, and it should be our goal to understand and describe these phenomena. I do not intend to make any direct comparisons to these very different culture groups from an extensive geographic range, but I do wish to highlight the discrepancies in the conclusions many archaeologists are making regarding different variations of hunter-gatherer-fisher groups in the archaeological record. This research sheds light on some of the specific intricacies of hunter-gatherer-fisher populations living in northeast China at the end of the Holocene and will hopefully contribute to improved means of describing the unique phenomena of ceramic foragers in this region.

1.1.3 Subsistence and Domestication

A great deal of research has considered the process and drivers of domestication, which often focuses on northeast China, specifically the Liaoxi region, as it is one of the early centres of millet domestication. Scholars have proposed three main hypotheses for the transition from hunting and gathering to agriculture for this region (Shelach-Lavi et al., 2019). First, the stress or "oasis" hypothesis where environmental conditions deteriorated, or overpopulation, or both, caused innovation in food procurement. The second suggests that transition occurred during a time of enriched resources, allowing for experimentation of the environment to improve resources. The third posits that domestication was brought about by socio-economic competition and is not influenced by external forces such as the environment. Since this research possibly includes innovations in food procurement and processing alongside innovative adaptions of hunter-gatherers to a changing environment, it is necessary to discuss these hypotheses together.

Current and previous research often analyses the potential relationship between the emergence of pottery and the eventual domestication of millet in northeast Asia. There are two primary reasons for this. Firstly, scholars identified warming in the region after the late Pleistocene and early Holocene as a driver of both eventual domestication and the emergence of pottery production (Aikens et al., 2010; Derevianko et al., 2014; Li et al., 2020; Yang et al., 2018; Yanshina, 2017). Second, pottery in this region has long been assumed to have been used primarily for cooking newly popularised foods resulting from such climatic fluctuations, which does not include cereals but could include more wild plants (Wang and Sebillaud, 2019). This in turn relates to changes in diet and relationships between people and their environment and thus issues of domestication. However, this change in climate resulting in environmental richness alone likely did not bring about agriculture (Aikens and Lee, 2013).

Importantly for pottery emergence and domestication, there is evidence that Pleistocene hunter-gatherers began to use grinding stones for processing *Triticeae* and *Paniceae* grass species. Liu et. al (2013) identified that for starches on grinding stones revealing the cultivation of these plants at Shizitan 柿子滩 in North China as early as 23,000 years ago by hunter-gatherers. Since *Paniceae* grasses were exploited about 12,000 years before their domestication, further illustrating the long process which occurred over many years during which pottery becomes prominent. Using starch grain analysis, archaeologists can trace the points in time where *Paniceae* grass species start to be favoured more heavily than *Triticeae* species, and this period also coincides with environmental data showing a changing climate (Yang et al., 2018).

Unsurprisingly, there is evidence for cultivation eventually leading to domestication much earlier than physical evidence for domesticated plants. Plant domestication began with hunter-gatherers and involved a long process of cultivating plants and the selection of domestication traits “varied not only by taxa and region but also according to when, where, and how different tillage and harvesting techniques were employed”. These changes occurred slowly over many millennia from the late Pleistocene into much of the Holocene (Harris, 2014, p. 2). Despite the slow domestication process, fully domesticated millet appeared rapidly in the archaeological record of the Liaoxi region of northeast China during the early Holocene (Lee, 2016). Domestic millet was evident in a broad region over North China by 8,000–7,500 cal. BP, suggesting its domestication was initiated far earlier (Lee, 2016). Some argue millet spread from northeast china northward into Russia’s far east through Manchuria via an inland route (Li et al., 2020).

This potentially highlights the presence of already established links between these two regions and plant use/diet, which differed from coastal communities in East Asia. Researchers often examine sedentary living in northeast China in relation to plant/animal domestication and the emergence of pottery, however, this evolved rapidly at a later date (Shelach-Lavi et al., 2019). Sedentism is not be discussed extensively in this work because there is no evidence of permanent occupation at the sites analysed as they were likely seasonal camps (Wang and Sebillaud, 2019).

Shelach-Lavi and Tu (2017) argue that early pottery production may have no connection with eventual domestication and transition to agriculture in this region, or their interconnection is at least more complex than previously assumed. This argument is based on evidence that wild plants and animals are prepared in these containers, alongside the wild versions of future domesticated plants, such as millet. Shelach-Lavi and Tu (2017) argue that since resources produced in these vessels—such as acorns and deer—were never domesticated, the relationship between vessel emergence and agriculture are not inherently related. This argument is problematic for two reasons. Primarily, it implies that humans have the ability to domesticate every wild plant and animal. This notion is false, as some plants and animals do not carry traits that make them predisposed to domestication (Gifford-Gonzalez and Hanotte, 2013; Harris, 2014). Second, it removes human agency and the influence of additional factors by suggesting the process of domestication had no other socioeconomic or cultural considerations. For example, there is a possibility that humans preferred the taste of millet to acorns, or millet had a more beneficial growing season for the type of environment, as it thrives in cold climates and has a short growing season (d'Alpoim Guedes, 2011). Furthermore, there are additional considerations in acorn processing, as they are bitter and poisonous to humans before a long process of tannin leaching (Driver, 1952). This convoluted processing could have given humans another motivation for abandoning acorns as a food source in favour of something like millet. Finally, this notion suggests that the use of pottery for wild resources had little to no influence on people's relationship to their environment which would develop people's understanding of how and when these wild resources grow, making future domestication is more feasible. Many scholars agree that domestication is a long process that requires an in-depth knowledge of one's environment, and intensive exploitation of wild recourses processed in pottery vessels would likely contribute to this knowledge (Crawford, 2008). Overall, the separation of domestication from early pottery use and development removes the technology from its social and economic context. Such innovations do not occur in a vacuum, and such

conclusions cannot be drawn without direct empirical evidence about products produced in ceramic vessels.

This thesis aims to understand the role of hunter-gatherers in the process leading to plant domestication and sedentism, a role which has been less frequently studied in the past (Hommel, 2012; Renfrew, 2007). I aim to contribute to this discussion by studying hunter-gatherer diets and their change over time using pottery as a proxy. The present study also aims to understand subsistence in context with the surrounding environment. The association between food and environments are detrimental to the eventual domestication of millet in the region, particularly considering the shift in preference from *Triticeae* grass species to *Paniceae* grass species (Yang et al., 2018). Changes in eating habits and ceramic technology are likely heavily intertwined with environmental adaptions and other cultural changes and must be considered in context.

1.1.4 The Neolithic of Northeast Asia

Based on pollen data from Japan, we know deglaciation occurred in northeast Asia 20,000 years ago causing the return of temperate biota previously limited to lower latitudes (Aikens and Lee, 2013). Along with these changes in ecology, we see changes in human behaviour, and hunter-gatherers living in this area began to adapt by broadening subsistence strategies to include new resources (i.e., acorns, nuts, and annual seeds) after the extinction of megafauna (Aikens and Lee, 2013). These changes in human activity are reflected not only in palaeobotanical and zooarchaeological remains but also in changes in lithic technologies and the emergence of grinding stones and the use of body ornaments (Qu et al., 2013; Shelach-Lavi and Tu, 2017; Sun et al., 2014; Yang et al., 2018; Yue et al., 2020a; Zhang et al., 2011). These changes in human activity likely coincide with changes in human behaviour, possibly including social structures, food consumption and preparation, divisions of labour, etc. These changes developed gradually but gained significant momentum during the late Pleistocene and early Holocene, where they were accompanied by the emergence of pottery (Shelach-Lavi and Tu, 2017). Together, these changes in technology suggest a significant change in human economic behaviour, subsistence, and human-environment relations as different plants and animals are being exploited for consumption than in previous years.

At the end of the Holocene, East Asia sees a boom in pottery production across Japan, Lake Baikal, the Russian Far East (primarily the Amur River Region), Korea, and Northeast China (Yanshina, 2017). While each region displays variations in pottery production and style, many of these contemporaneous ceramics have common characteristics, particularly those on the mainland, including rim notching, stamping, organic and/or grog temper, and hand-building (Figure 1.1) (Wang and Sebillaud, 2019; Yanshina, 2017). It is unknown if these characteristics developed independently, and Yanshina (2017) argues there is currently not enough evidence to determine their exact relationship to one another. Conceivably, these vessels could have been created to replace woven baskets for cooking, eating, and storing new plant species being exploited in abundance at the beginning of the Holocene, benefiting from relatively fast and unskilled[2] production compared to weaving, and longer cooking periods (Aikens and Lee, 2013). The following sections offer a discussion about the Neolithic in the different parts of northeast Asia respectively (Figure 1.2 and 1.3). The state of current research regarding pottery use and residue analysis in this region is outlined in section 1.1.6. This discussion is a combination of all the individual regions of northeast Asia because many residue studies have included samples from multiple regions, and they are presented together for clarity and to avoid repetition.

[2] I use the term "unskilled" even though it is subjective because this is what is being argued by Aikens and Lee (2013). Other researchers may disagree that pottery is easier to make than baskets. Such a discussion is outside the scope of this work.

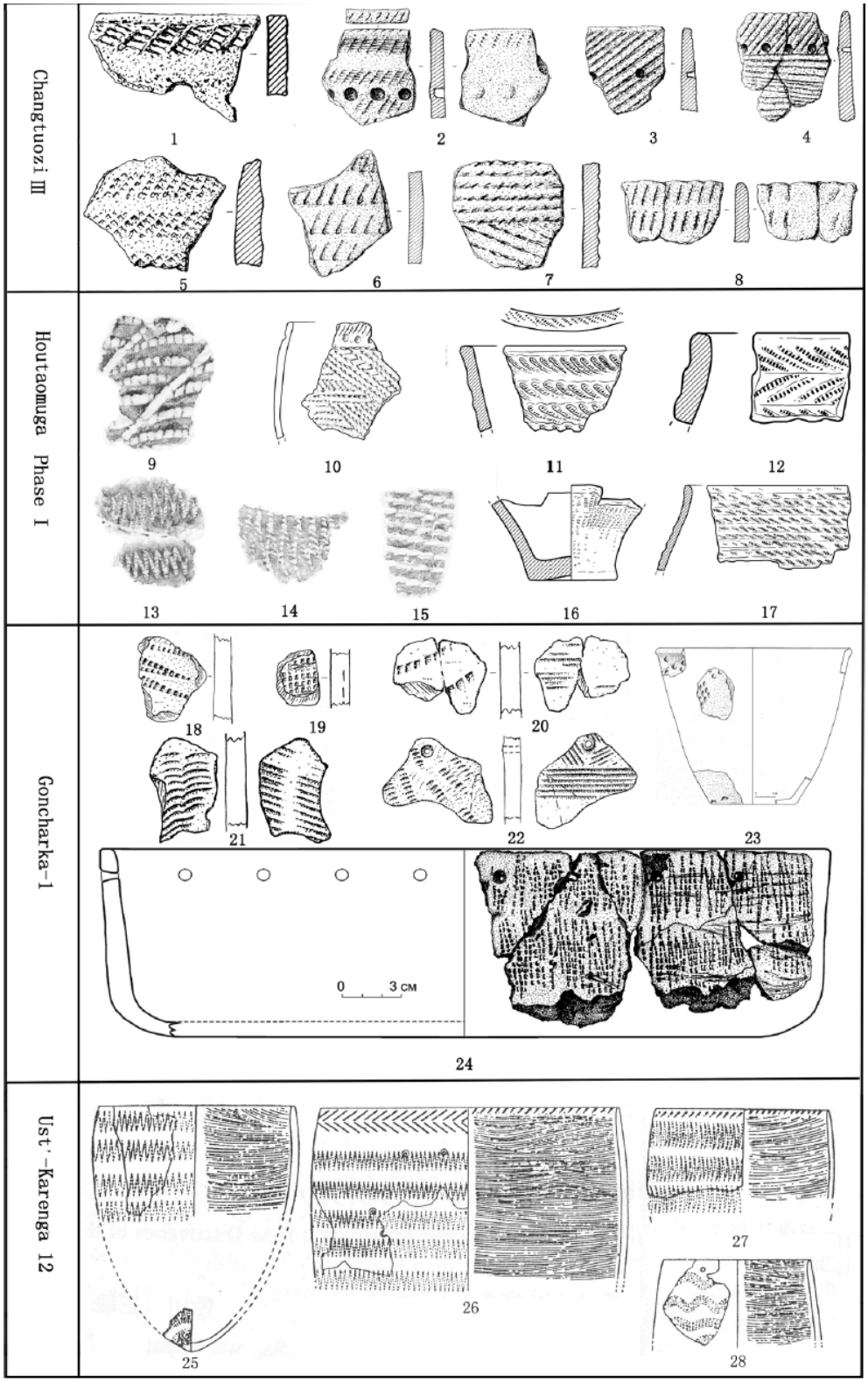


Figure 1.1. Visual comparison of pottery from northeast Asia from Wang and Sebillaud (2019:81). Changtuozi III and Houtaomuga Phase 1 come from sites in northeast China. Goncharka-1 is a site located near the Lower Amur in Russia's Far East. Ust'-Kareng pottery comes from the eastern side of Lake Baikal. The exact locations of each site can be seen in figure 1.2.



Figure 1.2. Map of early pottery sites in East Asia (Wang and Sebillaud, 2019, p. 75).

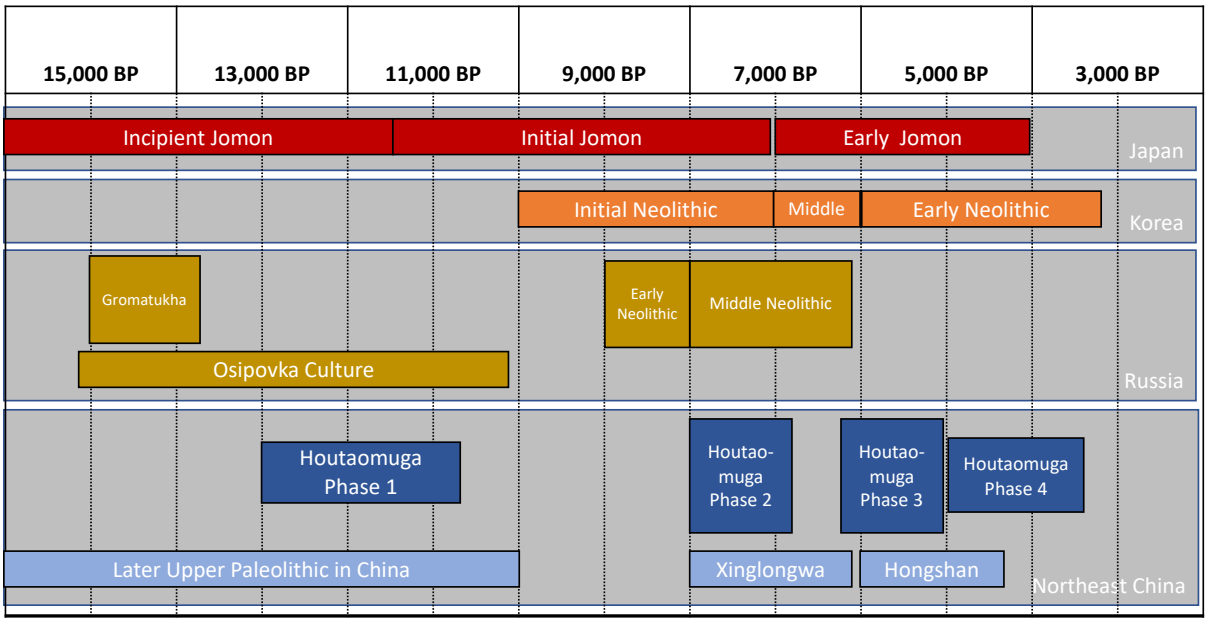


Figure 1.3. Compilation of dates and time periods of northeast Asia mentioned in the text of this thesis. Exact dates and their references can be seen in appendix II.

1.1.4.1 The Japanese Archipelago

The Japanese Archipelago was the first region of northeast Asia with an early Neolithic occupation discovered by archaeologists. Specifically, the emergence of the Jōmon period (15,000 to 5,000 BP)[3] in this area received significant scholarly attention, because, at the time of its discovery in the 1970s, this culture horizon was significantly older than any other previously known Neolithic phase (Yanshina, 2019). Jōmon 縄文 means "cord marked", similar to the term for the Neolithic in Korea, Chulmun 즐문 (Jeulmun; "comb-marked"), and this phrasing comes from the surface treatment on Neolithic pottery (Figure 1.4) (Kuzmin, 2006; Lee, 2017). According to settlement data, people living during the Jōmon Period on the Japanese Archipelago lived with changing degrees of sedentism, which can vary by region and time period significantly (Matsumoto et al., 2017). Regardless, they are often referred to as sedentary hunter-gatherers in most literature and they trend more toward sedentary settlements from the Incipient to the Late Jōmon (Matsumoto et al., 2017). Subsistence practices during the earliest phases involve a forager-fisher economy, with certain regions focusing on more extensive plant use than others (Habu, 2004). Along with the adoption of pottery during this period, people during this time move away from microblade and composite tool technologies

[3] The end date for the latest Jōmon period is debated by archaeologists.

towards a less labour intensive flaked stone tool industry, possibly also implying a change in economic activity (Iizuka and Izuho, 2017). People living during this period begin to favour partially polished axes and adzes, bifacial tools, new types of cutting and scraping tools, abraders, and grinding stones, however, their appearance throughout the region is inconsistent (Habu, 2014; Yanshina, 2019, 2017). These changes in tool technology suggest significant changes in human activities. This change along with the adaption of pottery is accompanied by a shift in settlement location from caves in the Palaeolithic towards regions near lake and river margins during the Neolithic and changes in diet which focus on marine resources (Yanshina, 2019). This contrasts with the main site of study in this thesis, Houtaomuga 后套木嘎, where microblade and composite tool types are preferred, however settlement patterns during the Neolithic also clustered around lakes and river margins (Sebillaud et al., 2021).

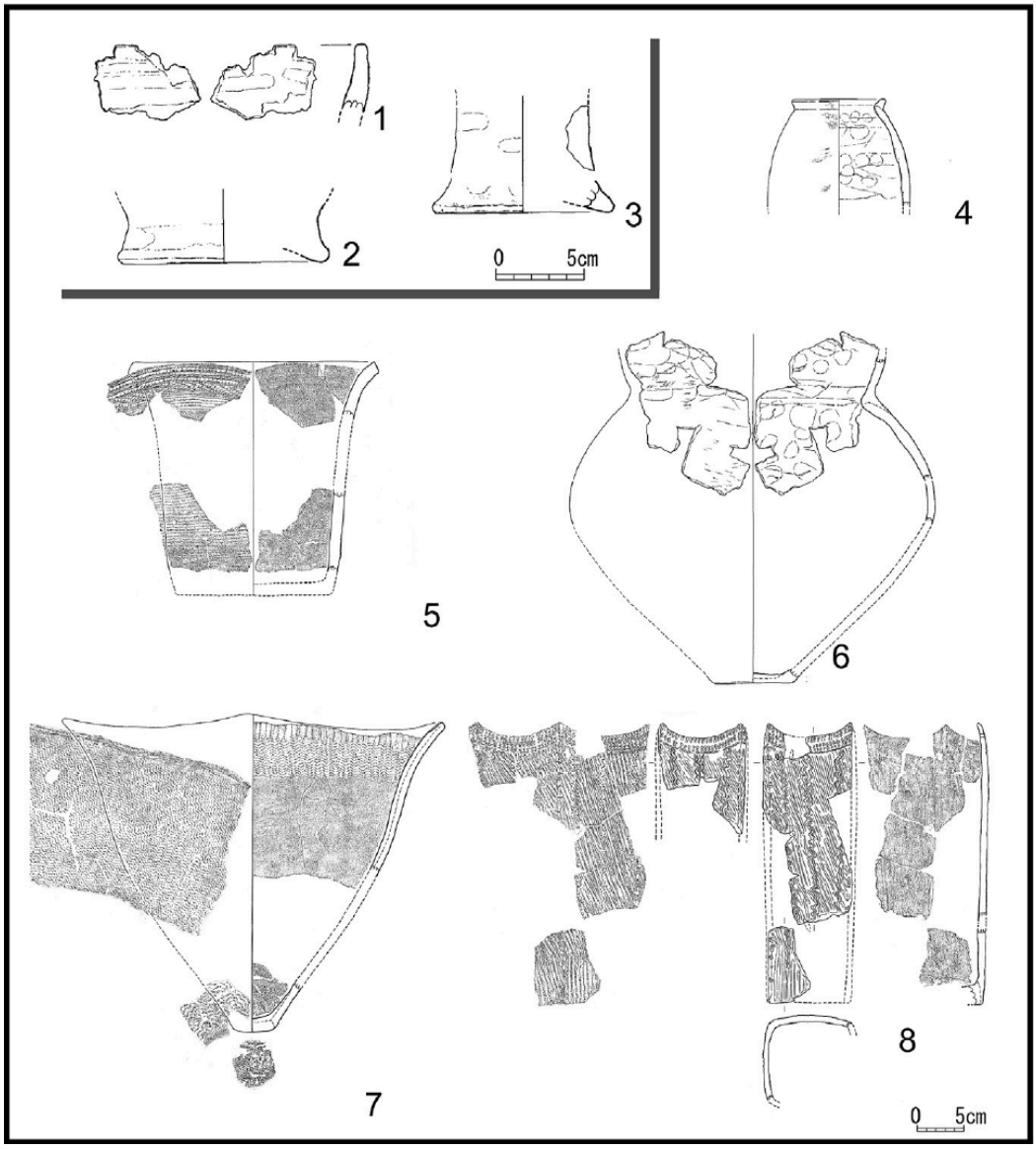


Figure 1.4. Vessel forms from the Kenshojo-Ato site in Japan. 1 to 3: forms from the Incipient Jōmon period, 4 to 8: forms from the Initial Jōmon (Iizuka and Izuho, 2017, p. 109).

1.1.4.2 The Russian Far East

The Neolithic of Russia's Far East began at the earliest with the Okhotsk culture in Sakhalin Island between 13,000-10,000 BC (Zhushchikhovskaya, 2009). The Neolithic on mainland Russia spans from at least 8,000 to 3,500 BP and includes stone industries with abrasive and bifacial retouching techniques, early textile technology, and pottery with straight walls and flat bottoms which seem to imitate baskets (Figure 1.5). Settlements do not include structures and are only identified often by hearths (Yanshina and Kovalenko, 2022). Evidence of faunal assemblages and stone tool kits suggest that subsistence practices during this time mainly

included fish and small game (Kaner and Taniguchi, 2017). However, it is important to note that faunal remains do not preserve well in acidic soils around the Amur river (Yanshina and Kovalenko, 2022). The same changes in stone tool technology seen in the Jōmon period of Japan are present in mainland Russia, and lithic technologies include a wide range of bifacial tools used for a large variety of small game and fish (Kaner and Taniguchi, 2017). It was originally believed that this pottery type which appears in the Neolithic in Russia might have been moulded in baskets, however, recent research has shown these vessels were hand-built using coils (Wang and Sebillaud, 2019). While the exact firing temperatures is unknown, these vessels were likely fired at low temperatures (>900ºC) due to their texture (Zhushchikhovskaya, 2009). When this pottery initially appears in the archaeological record, its appearance is infrequent and in small numbers in this area, however, it becomes widespread and more frequent in later Neolithic hunter-gatherer-fisher communities (Shoda et al., 2020; Zhushchikhovskaya, 2009). Sites with early ceramics are located in large numbers along the Amur River, and evidence for intensive fishing and the beginnings of sedentary living in this area is present (Kuzmin, 2006). It has been hypothesised that this change in diet and lifestyle is the result of this change in settlement patterns and particularly favourable environmental conditions with warming temperatures and abundant river resources, in combination with rich clay deposits in the Primor'e and Priamur'e regions (Zhushchikhovskaya, 2009).

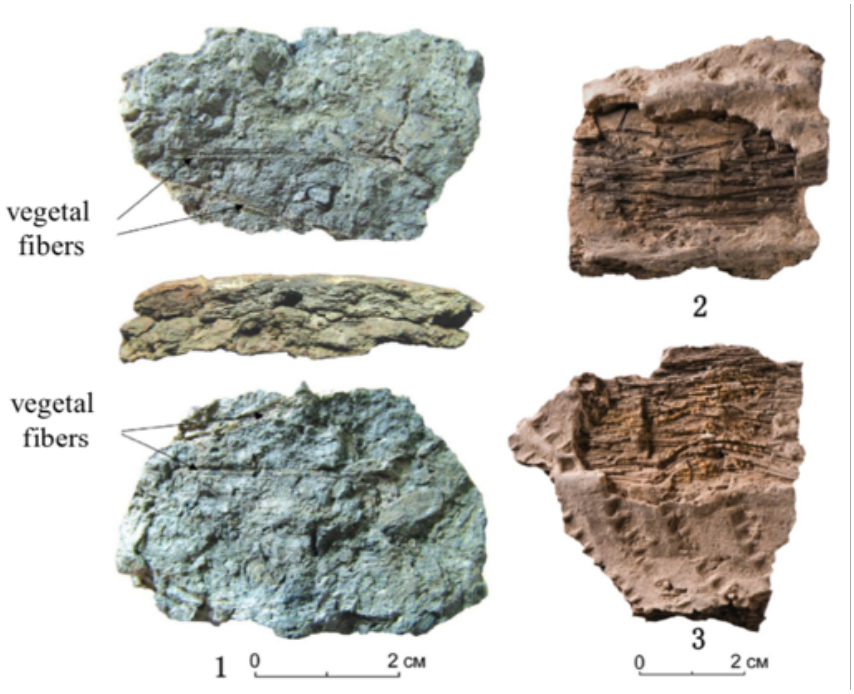


Figure 1.5. Detail of vegetal fibres used as tempering material in the clay of the pottery of Goncharka-I site (Russia, 1) and Houtaomuga Site from Phase 1 (2 & 3) (after Wang and Sebillaud, 2019, p. 26).

The further inland Circum-Baikal region of eastern Russia is different from the Russian Far East. The presence of ceramics in the later Pleistocene is not certain, and the shift towards plant and animal domestication is complex (Hommel et al., 2017; Hommel, 2012). Fishing develops in this region during the Early Neolithic period and fishing tools and bones appear in archaeological assemblages in larger numbers (Yanshina, 2019). However, many of the early pottery-bearing sites in the region do not have clear evidence for fishing and their initial relationship remains unclear (Yanshina, 2019). Due to this uncertainty in pottery development and the large geographic distance from the research area in Northeast China, this region of Russia will not be discussed in greater detail.

1.1.4.3 The Korean Peninsula

Similar to many archaeological traditions in East Asia, Korea follows a definition of "Neolithic" which begins with the advent of pottery and ground stone artifacts (Kuzmin, 2006). Neolithic sites in Korea have been approached in the past as "fossil records" and classified as Palaeolithic archaeology which influenced the ways they were excavated and catalogued, and the dating of these sites often rely on shell middens which are infrequent and difficult to

precisely date, making research on this period more difficult in the region (Bae and Kim, 2010). These dating and classification complications in the early years of archaeological research in Korea have resulted in early ceramics being removed from their social and economic context, and this relationship is still being fully understood today (Cho and Ko, 2009). The earliest pottery from Korea is nearly identical to the earliest known pottery from far eastern Russia, specifically Osipovka and Gromatukha types with flat bases and fibre tempering, thick walls, and little to no surface decoration (Kuzmin, 2006). The earliest possible date for ceramics in Korea discovered in context and with adequate dating is 10,500–10,000 BP, and this sample was located at the Gosanni site on the island of Jeju, off the southern coast of Korea (Cho and Ko, 2009; Im, 1995; Kang, 2002). The pottery at the site of Gosanni is the same comb-marked type found at coastal and inland sites. These samples, however, are only relatively dated with tephrochronology, and no ceramics have been found in context with a more specific radiocarbon date (Cho and Ko, 2009). Direct dating of ceramic has been undertaken, however, thermoluminescence dates tend to have significant error ranges (Kuzmin, 2006). The remaining sites on the peninsula with early pottery show a slightly later introduction date of 8,000 BC.



Figure 1.6. Raised-design ware from Osanni indicative of the Initial Neolithic period in Korea (SNUM 1984: 92, plate 35-1) (Cho and Ko, 2009, p. 154).

The majority of what is defined as Neolithic sites on the Korean Peninsula with pottery dated between 8,000 BC to 1,500 BC, a period referred to as the Chulmun (comb-marked) period based on the dominant ceramic decoration pattern (Bae and Kim, 2010; Cho and Ko, 2009). This Neolithic period is generally split into three further subperiods in this area of the Initial (8,000-5,000 BC), Early (5,000-4,000 BC), and Middle (4,000-1,500 BC) Neolithic or Chulmun (Cho and Ko, 2009). Lithics associated with this pottery include microblades and ground stone tools suggesting similar hunting and subsistence strategies to the other regions of northeast Asia (Lee, 2017). The Initial Phase is marked by raised design wares (Figure 1.6) mainly appearing in the south of the peninsula. The ceramic vessels are beaker-shaped with narrow, flat bases, and there are decorative appliqué bands around the rim. Isotopic evidence shows diets become more diversified during this Initial Period with an emphasis on coastal food resources and this may relate to the introduction of the technology (Kwak et al., 2020). Pottery from the Early Neolithic (Figure 1.7) is smaller than later periods and includes beaker vessels with impressed motifs located around the vessel rim, a style referred to as *abinmun* – meaning nail impressed, as this decoration type is made using sharp incising tools (Cho and

Ko, 2009). Sites dating to both the Initial and Early Neolithic periods have overlapping distribution patterns, mainly distributed on the southern and eastern coasts and pottery assemblages are accompanied by fishing hooks and harpoons (Cho and Ko, 2009). Sites from these periods do not include permanent structures and are mostly comprised of shell middens (Aikens and Lee, 2013; Nelson, 1990). Scholars note the possibility of pottery being introduced into Korea initially from Japan, where pottery was directly connected to adapting economies to fishing and gathering (Aikens et al., 2010a; Aikens and Lee, 2013; Cho and Ko, 2009; Lee et al., 2019). This change in technology in Korea may be related to climate warming and sea-level changes seen in the region in the early Holocene, where a wide variety of food resources enabled the emergence of "complex" hunter-gatherer-fisher economies (Habu, 2014).

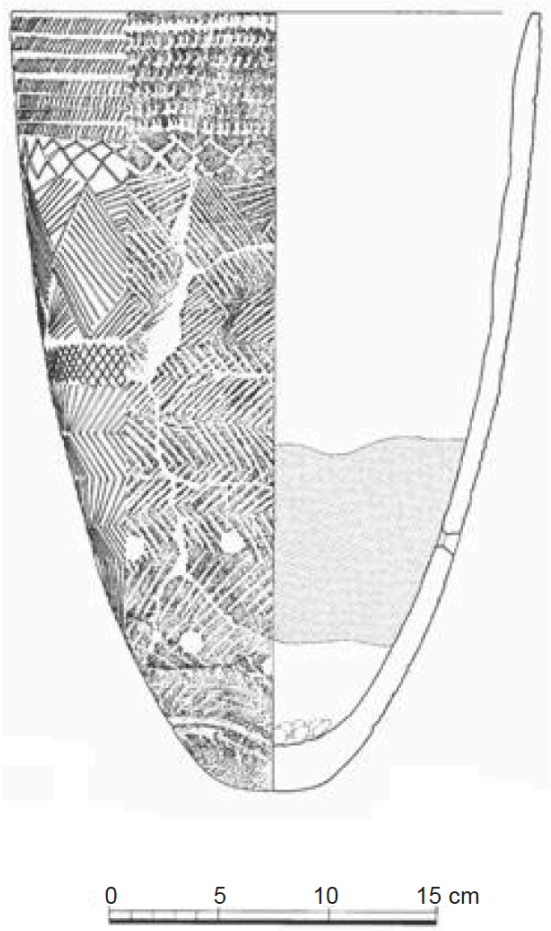


Figure 1.7. Comb-pattern ware from the site of Amsadong highlighting comb stamping pattern common in the Early Neolithic period in Korea (National Museum Korea NMK 1994: 298, figure 116 from Cho and Ko (2009:158).

The Middle Neolithic sites expand further inland than Initial and Early sites and begin producing the defining comb patterned pottery representative of this period in Korean archaeology (Cho and Ko, 2009). There is a significant increase in the number of sites during this period, and it has been hypothesised that an environmental change or population increase, or both, lead to resource stress, and an inability to feed the large populations present with the available shellfish and gathered plant resources (Cho and Ko, 2009). This hypothesis is supported by evidence for broadening flora and broadening subsistence spectrums coinciding with this period (Lee, 2011; Lee et al., 2019).

1.1.5 The Archaeological Record of Northeast China

Northeast China includes the provinces of Heilongjiang, Jilin, Liaoning, and the eastern part of Inner Mongolia, encompassing 350,000 km^2 (Jia, 2005; Wang and Sebillaud, 2019). Anatomically modern humans appear in the archaeological record of North China around 40,000 years ago during the Palaeolithic period (Mao et al., 2021; Shang et al., 2007). Archaeological investigations in northeast China have been particularly interested in the Liaoxi region and areas north of Beijing and its connections with the Chinese Central Plains. A significant portion of research has centred around northeast China, specifically the Hongshan Culture (ca. 6,000-4,700 BP) in the Liaoxi region, with evidence of jade production and large scale agriculture (Shelach, 2000). However, research has broadened substantially in the last 30 years. Until the 1990s, cultural periods that predate Hongshan were not known, identified, or named, with some archaeological material before this period being referred to vaguely as pre-Hongshan (Shelach and Teng, 2013). By now it is clear that the Xinglongwa culture (ca. 8000-6800 BP) with distinct pottery production and sedentary settlement sites predates Hongshan in the southern portion of northeast China (Shelach and Teng, 2013). Extensive research on settlements of the Xinglongwa period and those predating them in this region are expanding, and our understanding of the early Neolithic and contemporaneous culture periods nearby is developing. Less research has focused on the region North of the Liao River valley and the present study is contributing to closing this research gap.

1.1.5.1 The Palaeolithic Period in Northeast China

Before the Neolithic and beginnings of pottery production, the focus of this work, humans living in northeast China began utilising microblade technology and body decoration in the form of shell beads, and bone and shell tools appear in the archaeological record (Bar-Yosef, 2002). This period is known as the Later Upper Palaeolithic in China (21,000-10,000 BP) (Bar-Yosef and Wang, 2012; Qu et al., 2013). Human fossils from this period are rare, and this causes research limitations for this period (Qu et al., 2013). Bone tools also first emerge during the Upper Palaeolithic and include bone harpoons (Qu et al., 2013), tools particularly useful for people living in regions with abundant freshwater resources such as northeast Asia. Socioeconomic organization changes during this time, which leads to dramatic cultural and social changes in the early Neolithic (Bar-Yosef, 2002; Qu et al., 2013). Discussion of this period is important for this research, as these changes in the archaeological record mark a

significant change in human behaviour that lays the foundation for the Early Neolithic. To understand the shift of hunter-gatherers to using pottery technology, we need to discuss what came before in terms of behaviour, tools, and subsistence.

One of the main changes in human lifestyles occurs during the Late Upper Palaeolithic in northeast China and there was an emergence of microblade industries,[4] ending the dominance of previous core-and-flake industries around 30 ka cal BP (Lu, 1998). This use of microblades continues in North China from this period well into the Neolithic, and stone tools at Houtaomuga belong to the same industry (Chen et al., 2005). Researchers define this technological shift in tools as a marker for the beginning of the Late Upper Palaeolithic period in North China (Lu, 1998; Qu et al., 2013). Typologically a microblade is defined by archaeologists as a small blade with parallel edges whose length is more than twice its width (Qu et al., 2013).[5] In China this definition has been limited further by scholars to blades that are 20–60 mm long and less than 10 mm wide, however, they are often only 4–7 mm wide and 15–50 mm long (Lu, 1998). Microblade lithic industries do not exclusively consist of microlithics and include flake components, as bladelets alone are not sufficient for all tasks (Qu et al., 2013). A majority of scholars agree that fine-grained raw materials are a necessity for microblade lithic assemblages, such as flint (Chert) or obsidian (Qu et al., 2013).

Microblades were often produced as a part of composite tools either hafted in wood, bone or antlers as effective specialist weapons for hunting (Elston et al., 2011; Lu, 1998; Qu et al., 2013). Due to their organic components, composite tools do not survive in a complete state in the archaeological record except in exceptional circumstances, for instance in the Siberian permafrost (Zhang, 2021). Archaeologists have suggested microblade technology on its own or hafted as composite tools, is an animal-oriented technology used for intensifying hunting while minimising risk and labour (Elston and Brantingham, 2002). Using microblades as specialist hunting tools supports prevailing notions that humans intensified exploitation of resources in the environment during this period while environmental conditions are less

[4] Sometimes referred to as microlithic industries in western Eurasia.

[5] Different schools of archaeology have varying criteria regarding microblades. Such differences result in the subdivisions of prehistoric cultures across many regions to be disjointed and do not coincide chronologically. For clarity in this work, I refer to the microblade definition and chronological indices used most frequently by scholars working in North China specifically.

favourable than the Neolithic. Microblade industries occur during the Palaeolithic and continue well into the Neolithic in various regions across northeast Asia, including Japan, the Korean Peninsula, Mongolia, and Russia, however scholarly work understanding the dispersion and relation of these technologies is still ongoing (Qu et al., 2013; Yue et al., 2020b; Zhang, 2021). The rapid and wide-ranging spread of this stone tool technology has been mentioned as significant by archaeologists because the motivations behind the adoptions and the way the technology rapidly spreads are still not well understood (Bar-Yosef and Wang, 2012). This spread of technology during the Palaeolithic is worth discussing for this research, as in later years similar pottery technology spread through the same regions. The spread of microblade technology could suggest that there was an established network of trade routes for raw materials and the exchange of techniques and ideas. Later, the concepts of pottery technology could have travelled around the region using the same web of established interlinked networks, meaning one or more centres of pottery development could be possible, and the technology could have spread from these regions quickly over time.

Microlithics have also been found in the region around Houtaomuga, the main site of focus in this research. The earliest evidence for anatomically modern human occupation and microblade lithic industries in northeast China near Houtaomuga was found at the site of Xishantou 西山头 in context dating to around 28,000 BP (Jilin University et al., 2019). The site is located at an environmental transitional zone extending from the foot of the middle section of the Greater Khingan Mountains to the Song-Nen plain (Jilin University et al., 2019). The lithic assemblage contains blades, drills, scrapers, choppers, and microblades similar to Houtaomuga (Jilin University et al., 2019). Only one animal skeleton was found at Xishantou 西山头, however, it was too fragmentary be identified (Jilin University et al., 2019). The first microblade sites of the Late Palaeolithic in the Nenjiang River Basin are Dabusu 大布苏 and Guanyao 缸窑, and these stone types share similarities with other Palaeolithic stone tool industries in northeast China (Li and Chen, 2007). Since the last glacial period, northeast China has experienced cold and warm fluctuations, however, this reached its peak between 21,000 and 18,000 years ago, resulting in the region becoming an ecological transition zone between forest and grassland at the time (Jilin University et al., 2019). During the conditions of the Late Palaeolithic, the raw materials for microblade production were easy to obtain, and the mircoblades themselves efficient to produce and transport, leading archaeologists to infer that their production is a result of environmental adaption (Elston et al., 2011; Jilin University et al., 2019). Microblades are

more portable and easier to repair than their core-and-flake predecessor making them advantageous for people living in northern China as the climate cooled and the steppe expanded (Elston et al., 2011; Elston and Brantingham, 2002). While I find this hypothesis about microblade use as an adaption possible, it is noteworthy that the technology persists after an additional environmental change during the Early Holocene. Some additional research further explaining the reasons behind this would be beneficial here. Further research in the Nen River Valley between the first human occupation and the initial introduction of microblades to at Neolithic sites where pottery technology is first utilised would be vital to further understand motivations behind the adoption or development of the technology. This work aims to look at pottery use, which can shed some light on the reasons for use of the technology, but any interpretation can only go so far without further research into the Palaeolithic period.

Many scholars have argued that subsistence during the Palaeolithic is centred around hunting land animals based on microlithic technologies as discussed here and large mammal bones found at various sites (Bar-Yosef and Wang, 2012; Elston et al., 2011; Qu et al., 2013). They argue that these same animals were hunted using chopper technology, and an intensification of this activity occurs when people switch to microblades (Bar-Yosef and Wang, 2012). Elston et al. (2011) argues that this Late Palaeolithic adoption of microblades associated with early pottery in North Asia during the Neolithic is further evidence of a forager economy focused on land animals because plant intensification can be aceramic. My concern with much of these data is that they are primarily focused on the preservation of certain activities like plant and small aquatic animal exploitation. These activities are much more difficult to identify archaeologically, if they preserve at all, meaning the majority of the evidence we have to interpret human activity during this period is large mammal bones and lithics. In this instance, the absence of other evidence seems to be associated with evidence of absence. Additionally, increased animal exploitation can also be aceramic, as this occurs in other regions of the world during the Late Pleistocene, and Elston et al. (2011) assessment fails to mention this. Understanding subsistence practices during this period is vital to understanding pottery use in the Neolithic as many scholars have suggested the use of pottery signifies a significant shift in subsistence practices, however, it is difficult to understand a shift from one period to another if the initial period is not well understood.

Currently, there is no evidence for Palaeolithic pottery in North China. Pottery during the Late Palaeolithic is unique to South China and does not occur anywhere else during this time (Qu et

al., 2013). Pottery from the Late Palaeolithic is mostly round-bottomed vessels with uneven surfaces of all over surface treatments of thick cord marks or woven patterns, often with soot marking implying their use for cooking or boiling (Wang and Sebillaud, 2019; Zhang, 1999). A wide range of hypotheses regarding what food types were cooked in these vessels have emerged and include primitive cultivated rice (Zhao and Wu, 2003), bone marrow (Cohen, 2013; Patania and Jaffe, 2021), and aquatic animals including fish and clams (Fu, 2012). This phenomenon of Palaeolithic forager pottery however occurs in southern China, making its in-depth discussion out of the scope of this work (Cohen et al., 2017; Kuzmin, 2017). It would be interesting in the future if residue analysis is extensively applied across this period in China to compare results to those presented here and the rest of North China to shed light on similarities and differences in pottery use to better understand motivations for the production and use of them by hunter-gatherers at a large scale on the continent.

1.1.5.2 The Neolithic Period in Northeast China

Located in central northeast China, the Song-Nen Plain is an inland low elevation region where the Songhua River (a tributary of the Heilongjiang (Amur) River) and the Nen River join (hence the name Song-Nen). The region lies in a transitional zone between two environments; the arid high altitude of the Mongolian Plateau to the northwest and the humid forested environments of the Changbaishan Mountains in the southeast (Sebillaud et al., 2021). This region in Jilin province has an elevation of less than 200 metres and is full of lakes and marshes (Jia, 2005). The region is surrounded by three mountain regions: the Changbaishan Mountains to the southeast, the Daxing'an Mountains in the Northwest, and the Xiaoxing'an Mountains in the northeast (Figure 1.8)

During the Neolithic, North China becomes a centre of domestication for millet, and extensive research has covered this region. While there is little evidence of formal and extensive agriculture in the Neolithic in northeast China around the Song-Nen Plain, a discussion of the transitions to agriculture is warranted here because the long extensive process of domestication is thought to have begun far before evidence of domestication is present in the archaeological record. Moreover, the development of domesticated plants involves discussion of relationships between subsistence and human relationships with the environments, which is also a central focus of this work.

There are major differences in the ways people transition to agriculture between northeast China and the rest of North China, and scholars have suggested that the reason lies in the vast differences in environments (Lee, 2011; Shelach-Lavi, 2015). This suggests that even before the beginnings of formal agriculture, people in northeast China had a different relationship to their environment and differences in subsistence practices than areas where millet agriculture comes to dominate in later years, particularly in the Liaoxi region. Many argue that people living in the Song-Nen Plain during the Neolithic were more dependent than previous Palaeolithic groups on fishing, hunting, and plant foraging based on different stone tool technologies, net weights, settlement patterns, and palaeobotanical research (Jia, 2005; Sebillaud et al., 2021; Tang et al., 2020). The Song-Nen Plain is home to a series of sites that have the earliest evidence for pottery production in northeast China. One of these sites – Houtaomuga – will be the focus here. This site was chosen for this study because of the long periods of occupation, thorough documentation, and abundance of ceramic material.

1.1.5.3 The Houtaomuga Site

The Houtaomuga site (后套木嘎, N 45º 39’ 27.5’’, E 123º 47’ 15.1’’) is located in Da’an City on the southeast shore of Lake Xinhuang, Jilin Province in the Nenjiang River basin (Figure 1.2 and 1.8) (Wang and Sebillaud, 2019). The site was excavated from 2011to 2015 by the Research Centre on Chinese Frontier Archaeology of Jilin University, Jilin Province Archaeological Research Institute, the French Ecole Pratique des Hautes Etudes, the University of Oregon. The site stretches for a total area of 1,410,000 m^2 with a high-density artifact zone of about 550,000 m^2 (Wang and Sebillaud, 2019). Archaeologists have identified seven phases of human occupation at the site based on pottery styles spanning from the late Palaeolithic to Early Neolithic transition (13,000 cal BP) to the Liao-Jin period (9^{th} to 13^{th} century AD; Figure 1.8) (Wang and Duan, 2013; Wang and Sebillaud, 2019; Wang et al., 2016).

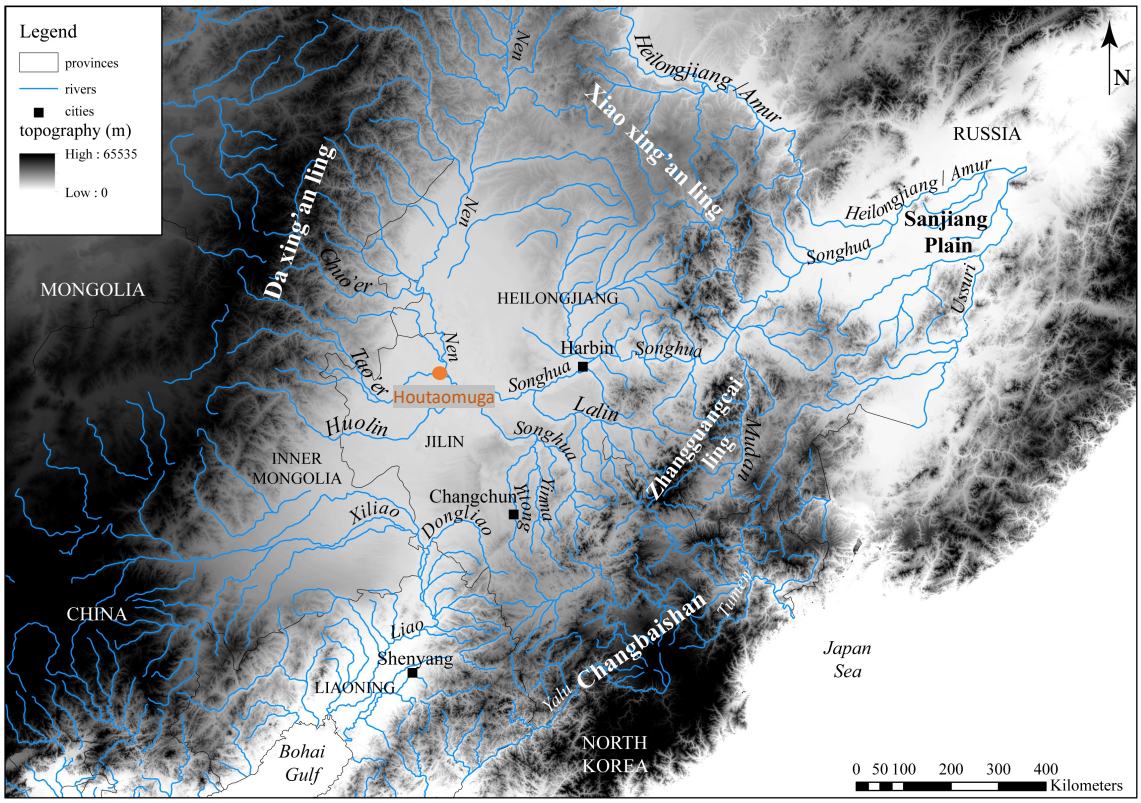


Figure 1.8. Topographic map of northeast China and the location of Houtaomuga. Map credits Pauline Sebillaud.

Over 2,000 sherds from a transitional period between the Late Palaeolithic and Early Neolithic have been identified from Houtaomuga Phase I. Kunikita et al. (2017) did an extensive series of radiocarbon dates at Houtaomuga and the nearby site Shuangta for the initial Neolithic phases (Figure 1.9). Additional radiocarbon dates which are unpublished are presented in table 1.1[6]. Phase 1 Houtaomuga (H-I) was dated to 10,820-9,770 BP (13,000-11,000 cal BP); Phase 2 Houtaomuga (H-II) was dated to 7,085-6,755 BP (8,000-7,500 cal BP); Phase 3 Houtaomuga (H-III) was dated to 5,380-4,920 BP (6,300 -5,500 cal BP), and Phase 4 Houtaomuga (H-IV) was dated to 4,420 BP (5,000 cal BP). Houtaomuga is similar to sites in the South-East Coast region of Russia, Japan, and the Korean Peninsula during this period based on similarities in pottery and other artifacts such as microblades (Wang and Sebillaud, 2019). Obsidian artifacts

[6] These radiocarbon dates are presented in BC as dates in this format are the preferred calibration for most archaeologists working on Neolithic China. In archaeological science, BP dates are preferential, and I aim to use these dates as often as possible in this research. Other published research on residue analysis in northeast Asia is often presented in BP, and my goal is to present some continuity with much of this work and my own.

are present at Houtaomuga, suggesting contact with these neighbouring regions is likely, however, the degree of contact is not fully understood yet (Cho and Ko, 2009). The local environment at Houtaomuga is different from many of these surrounding regions and warrants further description and discussion, specifically considering the implications changing environment have on altering human activities.

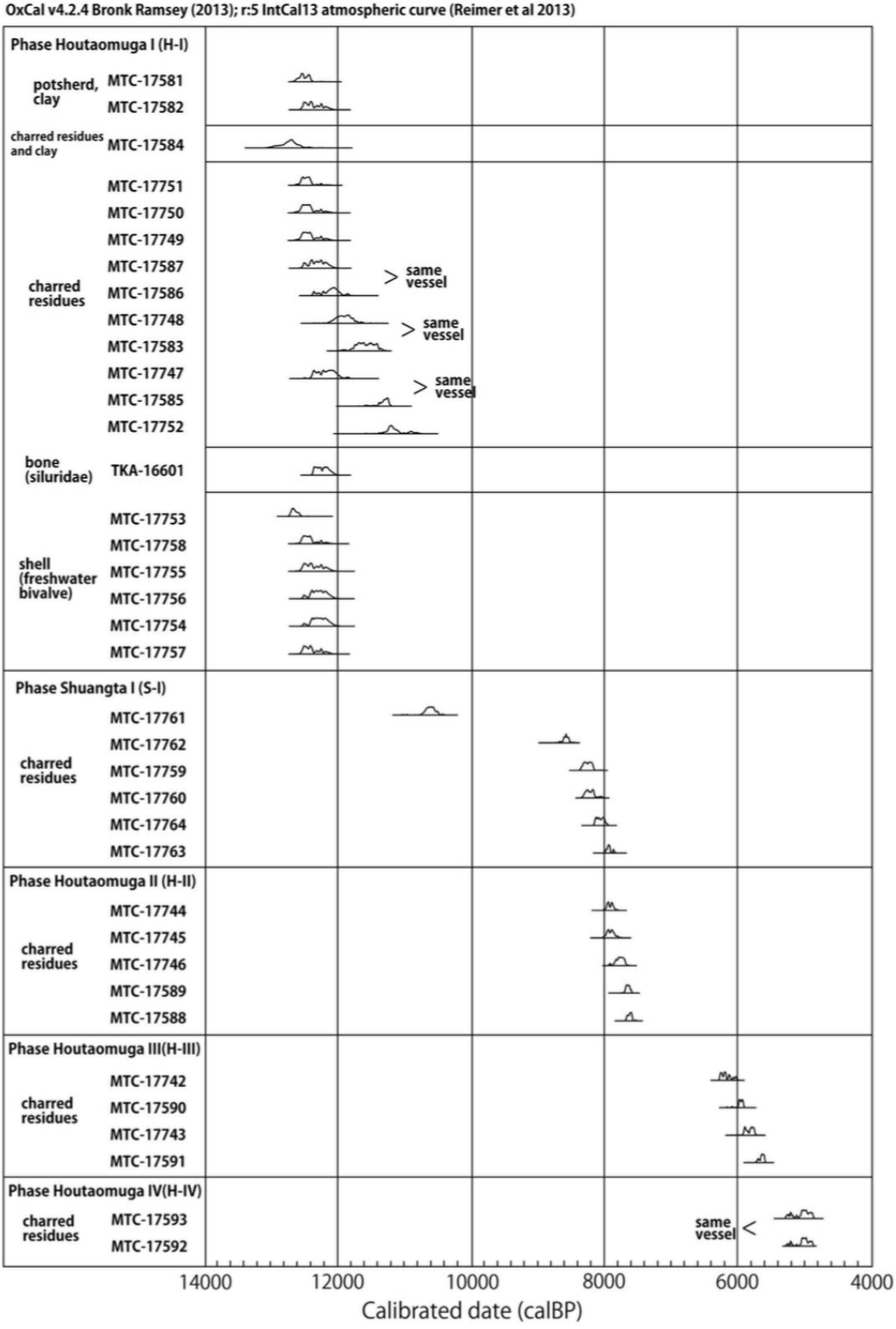


Figure 1.9 Calibrated ages of the Houtaomuga and Shuangta Sites (Kunikita et al. 2017, p. 65).

Table 1.1. Table of radiocarbon dates from the site of Houatomuga

Phase	Feature number	Sample	Calibrated date (cal. B.C.)	Laboratory
I	12DHAIIIG22	Charred organic residue	11 082–10 582	Tokyo
I	11DHAIIIG18	Vegetable fiber tempered shard	10 990-10 900	Beijing and Xi'an
I	12DHAIIIG22	Shell	10 773-10 618	Tokyo
I	12DHAIIIH143	Vegetable fiber tempered shard	10 716–10 446	Tokyo
I	12DHAIIIH146	Vegetable fiber tempered shard	10 716-10 446	Tokyo
I	12DHAIIIG21	Charred organic residue	10 682-10 421	Tokyo
I	12DHAIIIG21	Charred organic residue	10 669-10 425	Tokyo
I	12DHAIIIG21	Charred organic residue	10 642-10 182	Tokyo
I	12DHAIIIG22	Shell	10 641-10 272	Tokyo
I	12DHAIIIG21	Shell	10 618-10 184	Tokyo
I	11DHAIIIT1006(3)	Vegetable fiber tempered shard	10 608–10 183	Tokyo
I	12DHAIIIG21	Shell	10 608-10 172	Tokyo
I	12DHAIIIG22	Charred organic residue	10 581–10 153	Tokyo
I	12DHAIIIG21	Shell	10 490-10 125	Tokyo
I	12DHAIIIG21	Shell	10 480-10 107	Tokyo
I	12DHAIIIH189	Charred organic residue	10 460-10 000	Tokyo
I	12DHAIIIG22	Os de poisson chat	10 450-10 131	Tokyo
I	12DHAIIIG22	Charred organic residue	10 296–9 986	Tokyo
I	12DHAIIIH233	Vegetable fiber tempered shard	10 182-9 756	Tokyo
I	12DHAIIIH233	Charred organic residue	9 875–9 381	Tokyo
I	11DHAIIIH248	Dog bone	9 810-9 320	Beijing and Xi'an
I	12DHAIIIH189	Charred organic residue	9 463–9 257	Tokyo
I	12DHAIIIG22	Charred organic residue	9 448-9 112	Tokyo
I	11DHAIIIM45	Human bone	9 285–9 195	Beijing and Xi'an
II	12DHAIIIH246	Charred organic residue	6 056-5 880	Tokyo
II	11DHAIIIH132 :1	Charred organic residue	6 053-5 843	Tokyo
II	11DHAIIIM56	Human bone	6 020-5 890	Beijing and Xi'an
II	11DHAIIIG8:2	Charred organic residue	5 911-5 721	Tokyo
II	11DHAIIIH93	Charred organic residue	5 752-5 632	Tokyo
II	12DHAIIIG13	Charred organic residue	5 727-5 617	Tokyo
II	11DHAIIIM32	Human bone	5 660-5 540	Beijing and Xi'an
II	11DHAIIIM54B	Human bone	5 520-5 460	Beijing and Xi'an
II	11DHAIIIM54A	Human bone	5 480-5 370	Beijing and Xi'an
II	11DHAIIIM54C	Human bone	5 480-5 340	Beijing and Xi'an
III	11DHAIIIF2	Charcoal	4 340-4 220	Beijing and Xi'an
III	15DHAIF22(1) :67	Charred organic residue	4 334-4 222	Tokyo
III	11DHAIIIF4	Charcoal	4 270-4 050	Beijing and Xi'an
III	11DHAIIIF3	Charcoal	4 260-4 040	Beijing and Xi'an
III	11DHAIIIF4	Charcoal	4 260-4 040	Beijing and Xi'an
III	12DHAIVH44	Charred organic residue	4 068-3 957	Tokyo
III	11DHAIIIG6	Charcoal	4 045-3 965	Beijing and Xi'an
III	12DHAIIIH157(2):17	Charred organic residue	3 971-3 797	Tokyo
III	11DHAIIIM50	Human bone	3 950-3 700	Beijing and Xi'an
III	11DHAIIIF6	Charcoal	3 950-3 760	Beijing and Xi'an

III	11DHAIIIG1	Auroch tooth	3 900-3 700	Beta Analytics
III	11DHAIIIF6	Charcoal	3 810-3 700	Beijing and Xi'an
III	11DHAIIIF6	Charcoal	3 800-3 690	Beijing and Xi'an
III	11DHAIIIF6	Charcoal	3 800-3 690	Beijing and Xi'an
III	11DHAIIIT1307(2)	Charred organic residue	3 770-3 644	Tokyo
III	11DHAIIIG1	Charcoal	3 720-3 630	Beijing and Xi'an
III	11DHAIIIF1	Charcoal (hearth)	3 660-3 510	Beijing and Xi'an
III	11DHAIIIM79	Charcoal	3 650-3 520	Beijing and Xi'an
III	11DHAIIIF1	Charcoal (hearth)	3 640-3 490	Beijing and Xi'an
III	11DHAIIIF1	Charcoal (hearth)	3 640-3 490	Beijing and Xi'an
III	11DHAIIIF1	Charcoal	3 540-3 360	Beijing and Xi'an
IV	14DHAIG1	Auroch tooth	3 500-3 300	Beta Analytics
IV	14DHAIG1	Auroch tooth	3 500-3 300	Beta Analytics
IV	11DHAIIIH85	Charred organic residue	3 127-2 917	Tokyo
IV	11DHAIIIH85	Charred organic residue	3 118-2 919	Tokyo

1.1.5.3.1 Local Environment

The area around Houtaomuga is located at the northern boundary of the modern East Asian Monsoonal system and is highly sensitive to rapid environmental changes (Yue et al., 2019). Environmental reconstruction through pollen analysis of varved lake sediments in northeast China sheds light on the past climatic conditions during various phases of occupation at Houtaomuga (Stebich et al., 2015, 2009; Teng, 2010, 2004; Wu et al., 2016). Additional pollen records from the site of Taoshan near the southern Lesser Xing'an Mountains also provide climatic insight and are discussed here (Yang et al., 2017; Yue et al., 2019).

During the late Pleniglacial period (16,700 - 14,450 BP), the environment was dominated by steppe and open taiga-like woodland landscapes, along with grasses and sedges characteristic of cold and dry conditions (Stebich et al., 2009; Wu et al., 2016; Yang et al., 2017). During this time, the population in the region increased and the density of archaeological sites increased as well (Yang et al., 2017). Overall, the early Neolithic in Northeast Asia occurred at a significant environmental period after the Last Glacial Maximum or LGM (21,000 BP). Northeast Asia was primarily a cold steppe environment during the LGM, and conditions continued to improve and warm between 10,500 and 8,000 BP (Li et al., 2017; Shelach, 2000). Additionally, this increased temperature lead to an increase in monsoon precipitation in Northeast China, which may have resulted in an improved plant load during this time (Li et al., 2019).

Beginning with the late glacial amelioration (~14,450 BP) and into the early Holocene (11,600 BP), the environment mainly consisted of dense deciduous forests predominately consisting of thermophilus broadleaf taxa (Stebich et al., 2009; Teng, 2004; Xia and Wang, 1987). This is a significant shift that can be identified by changes in plant resources, signifying an improvement in the productivity of the landscape (Yue et al., 2019). Yang et al. (2017) and Yue et al. (2019) suggest that during this period these environmental factors and population increase directly contributed to later sedentism and agricultural practices, aiding local populations to change subsistence strategies and behaviours through decreasing mobility and exploiting locally available resources at increased rates. Archaeologists suggest this signifies that the processes towards later settled agricultural groups began with these earlier conditions and can be seen in technologies indicative of resource intensification including pottery, axes, and adzes (Yue et al., 2019). This resource intensification and technological and lifestyle changes following a shifting environment was also observed in the adjacent region of northeast Asia, (Yue et al., 2020b). Two short term climate reversals to colder and drier conditions occurred at 13,900 – 13,800 BP (Oldest Dryas/Greenland Interstadial (GI) 1d event) and 13,1000 – 12,900 BP (Gerzensee/Killarney/GI-1b oscillation) (Stebich et al., 2009; Wu et al., 2016). A climatic reversal that is compatible with the Younger Dryas (12,900 – 11,600 BP) event in the circum-Atlantic region was present and marked by a decrease in broadleaf trees and reappearance of deciduous conifers indicative of colder conditions including *Picea* and *Larix* (Stebich et al., 2009). At the beginning of the Holocene (around 11,600 BP), dense deciduous forests consisting mainly of thermophilous broadleaf taxa become established and widespread in the region (Stebich et al., 2009). Pollen analysis has shown that despite an increasing vegetation coverage rate, the environment during this period was wet and cold, with temperatures decreasing further during the end of this Phase 1 at Houtaomuga (Tang et al., 2016). Pollen from gramineous plants increases during Phase 1, and this has been attributed partially to environmental conditions and human use of these plant types in pottery production (Tang et al., 2016).

During the first millennium of the Holocene (around 11,650 – 10,700 BP) the regional boreal vegetation was replaced by broadleaf deciduous forests during a transitional stage, and cold tolerant conifer rich areas changed into a warm and moist interglacial Holocene forest (Stebich et al., 2015; Yang et al., 2017). Shelach et al. (2019) hypothesize these wetter conditions facilitated the complex transitional process to sedentism and eventual millet domestication in

some areas of northeast China, as sedentary living practices evolve rapidly and domestication appeared to take several millennia in North China. From 10,700 to 7,800 BP, boreal conifers virtually disappear, and vegetation shifts to oak and walnut rich temperate broadleaf deciduous trees and shrubs. This suggests the environment was warmer than current conditions while being overall mild and dry (Stebich et al., 2015; Teng, 2010).

Around 8,500 BP, northeast China's environment was altered by the Holocene Climate Optimum resulting in warming temperatures and increased precipitation lasting until about 5,000 BP; during this period, the climate was warm and humid except for individual locations which were dry due to increased temperatures or elevation (Teng, 2010). The climate during this time shifted from cold and wet to warm and wet, likely being more comfortable for human occupation (Tang et al., 2016). During the middle Holocene (7,800 – 5,200 BP) humid conditions persist, forest coverage reached a maximum, and walnut trees became more prominent (Stebich et al., 2015). The region shifted to drier conditions from 5,200 to 3,600 BP, resulting in mixed conifer and hardwood forests. A rapid increase of birch pollen at the expense of pine species suggests decreased forest density and drier conditions, signalling substantial ecosystem disturbance (Stebich et al., 2015; Teng, 2004). This disturbance may be in part related to human disturbances due to the increased presence of Chenopodiaceae pollen (Stebich et al., 2015). This is substantiated in the archaeological record, as we know humans had a large impact on the environment in northern China from 5,400 BP due to intense "dry" agriculture – i.e. foxtail and broomcorn millets (Li et al., 2006). The local environment at Houtaomuga and the surrounding region likely influenced how and where people chose to live and perform activities, and a discussion of the settlement patterns in the region is needed.

1.1.5.3.2 Settlement Patterns and Site Distribution

The Houtaomuga site is located on the southeast shore of Lake Xinhuang, in Jilin Province (Figure 1.8). During the Neolithic period in northeast Asia, settlements are often located near water bodies, which provides abundant freshwater lakes, marshes, and rivers. In south-eastern Russia, the majority of Neolithic pottery bearing sites were located on the Amur river, such as Goncharka 1, the closest Russian site to Houtaomuga (Figure 1.10) (Shoda et al., 2020). This association between bodies of water and these sites is part of the reason archaeologists have associated people living here with an aquatic-based economy.

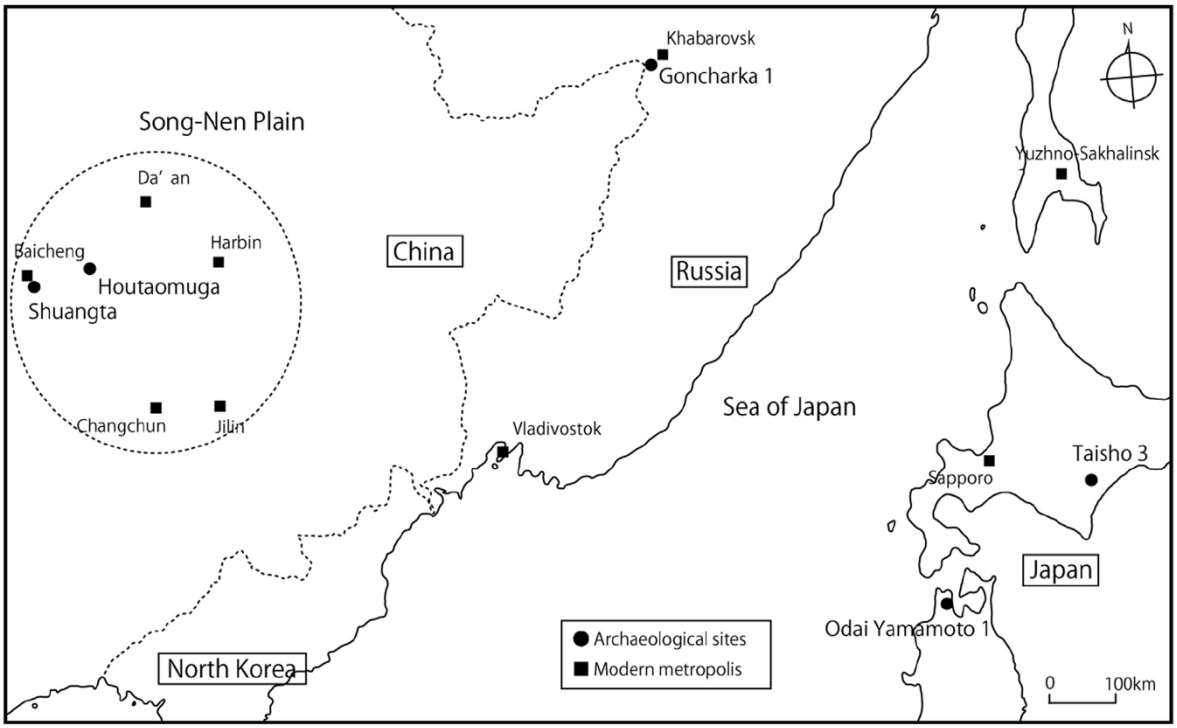


Figure 1.10. Map showing the distribution of early Neolithic pottery sites in northeast Asia mentioned in this thesis, including the main site in this research, Houtaomuga and the Goncharka-1 site (Kunikita et al., 2017, p. 63).

Houtaomuga is located on a flat plain with large marshlands and lakes. This location is suitable for hunter-gatherers who exploit aquatic resources. The extent of human occupation and exploitation of this aquatic environment is still not fully understood, but some research has shed light on the use of the surrounding area. Results of systematic regional survey around the Xinhuang and Yueliang Lakes near Houtaomuga show there were no housing features during Phase 1 of occupation at Houtaomuga or any of the surrounding sites, and no regional settlement organization had formed (Sebillaud et al., 2021). Phase 11 is likely a collection of scattered seasonal camps which formed much earlier than other settlements in the northeast China region (Sebillaud et al., 2021). Phase 1 features are distributed over 22,500 m^2 and have not been fully excavated outside of the Houtaomuga site in the region (Liu et al., 2016). Human settlement intensifies beginning in Phase 22, and the first architectural features appear between 4,300 and 3,500 BC During Phase 3 (Sebillaud et al., 2021). Phase 3 begins the first emergency of a simple settlement hierarchy without agriculture, while sites in the Liaoxi region to the south begin to show evidence for sedentism and agriculture around the same time (Sebillaud et al., 2021; Shelach and Teng, 2013). Any evidence for agriculture does not appear on the Song-Nen Plain until Houtaomuga Phase 3, and this evidence is still suggesting that domestic plants

were not a major contribution to the diet (Tang et al., 2020). Without agriculture like the southern regions, Phase 3 and 4 show evidence of established structures that can be formally described as a village and more elaborate large structures emerge (Sebillaud et al., 2021; Wang et al., 2017). Increases in pottery of various types suggest that occupation was more long-term than in previous periods, and settlements are located on higher elevation areas (Sebillaud et al., 2021). Phase 4, the Late Neolithic, was a time of marked social change around northeast China, and the Houtaomuga population and population density increases significantly during this time (Sebillaud et al., 2021). During all phases of occupation, water was a restrictive factor in the distribution of human activity areas, and people are likely selectively choosing higher ground locations for settlement for protection from other humans, animals, or flooding (Sebillaud et al., 2021).

A map showing the distribution of features excavated at Houtaomuga can be seen in figure 1.12 with each zone by occupational phase in figures 1.12 to 1.15 and those selected for the study can be seen in table 1.2. 6450 m^2 of the site have been excavated and includes 127 graves, 635 pits, 56 ditches, 43 semi-subterranean houses (Tang et al., 2020). Results of residue analyses will be compared between each phase but also between feature types and between different areas of the site in an attempt to further shed light on specific activity areas within the site. Residue analysis in other regions has been successful at marking particular areas of production and specific activity (Drieu, 2018). If such selective spaces exist at Houtaomuga, this dissertation will deepen our understanding of the activity and spatial organization of the Houtaomuga site during each of the early phases of occupation



Figure 1.11. Plan of all the features from all the phases excavated between 2011 and 2015 at the Houtaomuga site (figure via Pauline Sebillaud)

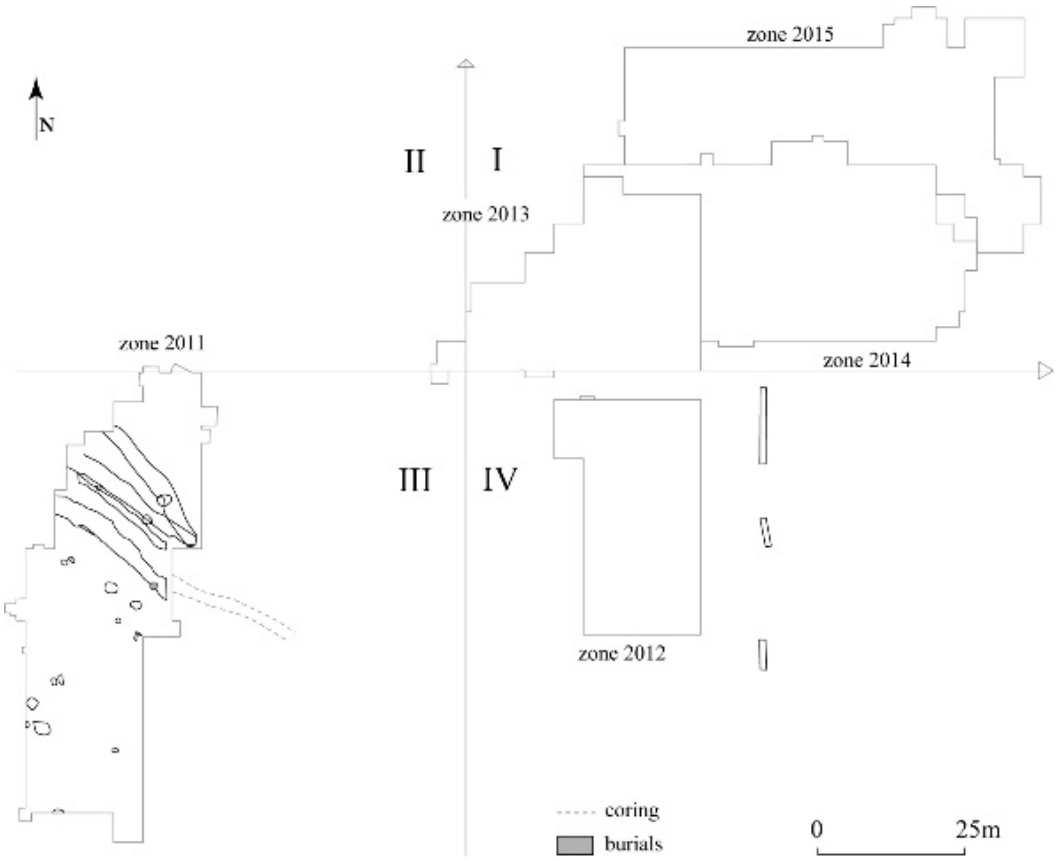


Figure 1.12. Plan of the phase 1 features in the excavation zones at the Houtaomuga site (figure via Pauline Sebillaud)

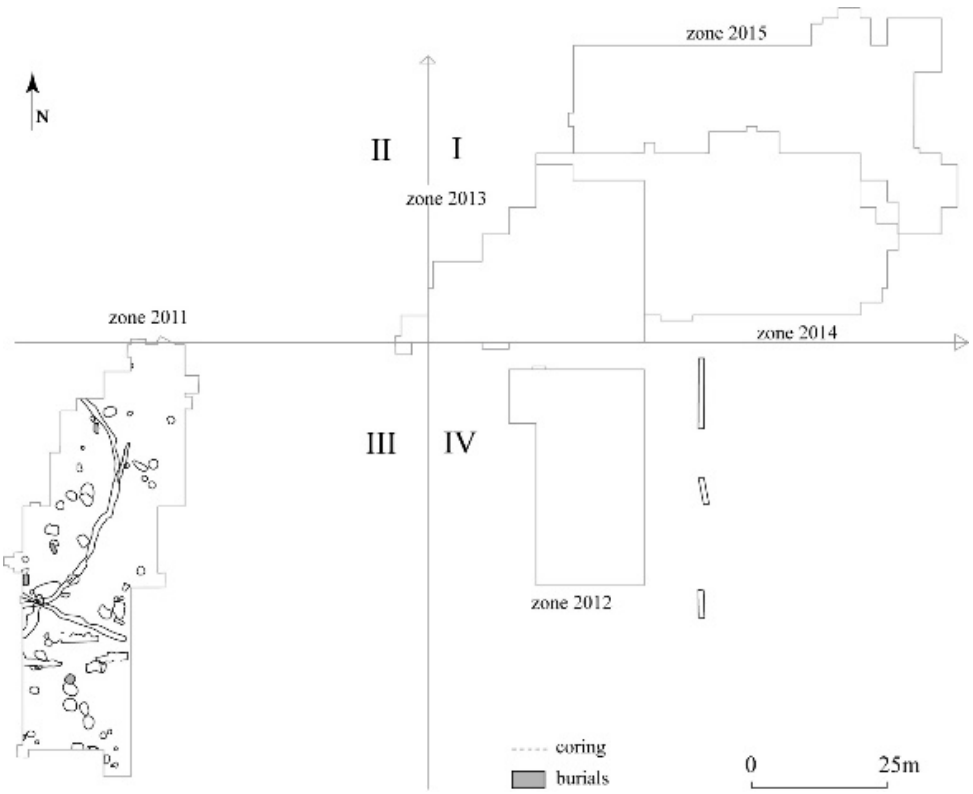


Figure 1.13. Plan of the phase 2 features in the excavation zones at the Houtaomuga site (figure via Pauline Sebillaud)

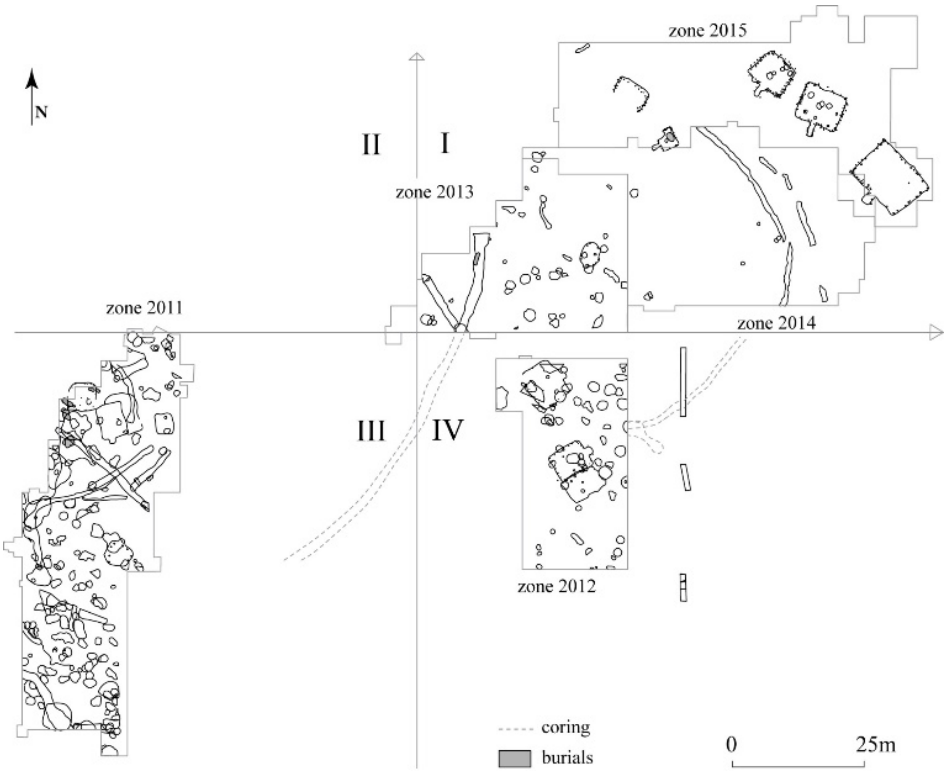


Figure 1.14. Plan of the phase 3 features in the excavation zones at the Houtaomuga site (figure via Pauline Sebillaud)

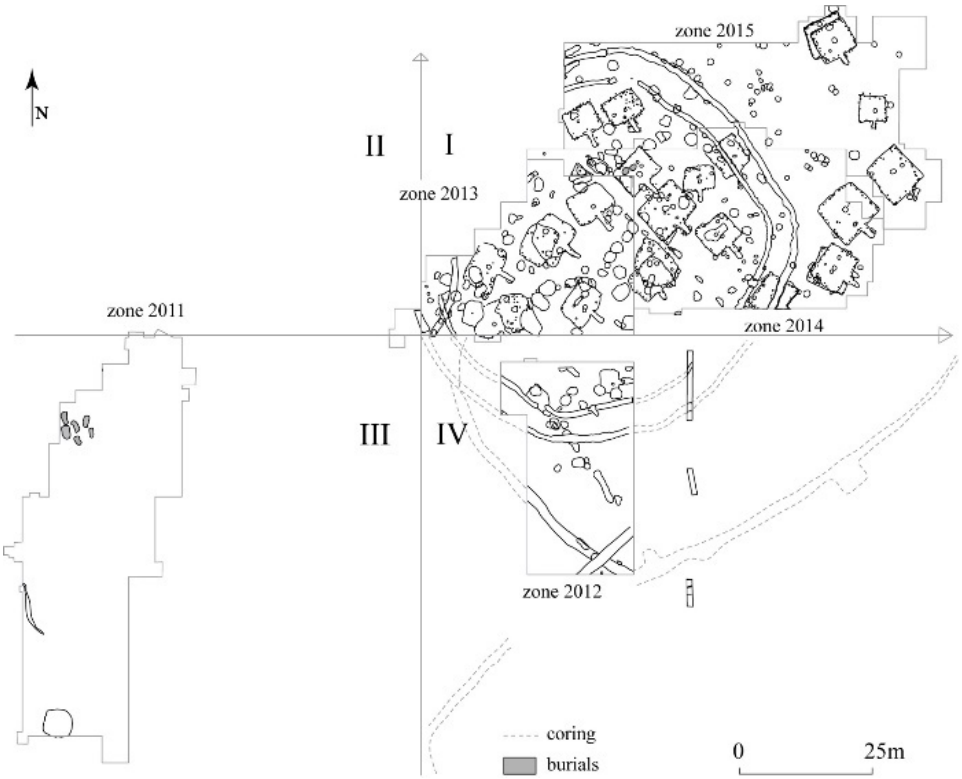


Figure 1.15. Plan of the phase 4 features in the excavation zones at the Houtaomuga site (figure via Pauline Sebillaud)

Table 1.2. Table of features yielding samples for this study

Feature	Phase	Zone	Feature Type
G18-1	1	III	Ditch
G18-2	1	III	Ditch
G21-1	1	III	Ditch
G22	1	III	Ditch
H146	1	III	Pit (Ash Pit)
H248-1	1	III	Pit (Ash Pit)
G002	2	III	Ditch
G013	2	III	Ditch
H131	2	III	Pit (Ash Pit)
G001	3	III	Ditch
F008-1	4	I	House Floor
F010-1	4	I	House Floor
F013	4	I	House Floor
F016-1	4	I	House Floor
F016-2	4	I	House Floor
F016-3	4	I	House Floor
F017-D1-2	4	I	House Floor
F018-2	4	I	House Floor
F021-2	4	I	House Floor
F023-2	4	I	House Floor
H085	4	III	Pit (Ash Pit)

1.1.5.3.3 Subsistence Strategies

Subsistence strategies at Houtaomuga were broad-based, being largely dependent on fishing and hunting, with plant cultivation based on palaeobotanical analysis, faunal analysis, microlith stone tool technologies, bone and shell tools, and evidence for aquatic resource procurement found at the site (Chen et al., 2011; Tang et al., 2020; Wang and Sebillaud, 2019). Tools, some of which have previously been associated with agricultural activities, including grinding stones and large chipped stones, are infrequent but present in the archaeological record of the Song-Nen Plain, where they were used by hunter-gatherers (Chen et al., 2011). Analysis of starches from grinding stones at other Neolithic sites in northeast China shows evidence for a broad-spectrum subsistence economy, as these tools were multifunctional and used for a variety of resources including both wild and domestic plants (Tao et al., 2011). This suggests these tools could have been used mainly for wild resources and useful for multiple applications on the Song-Nen plain. Analysis of plant remains from Houtaomuga suggests the use of upland and

wetland plant resources in early phases at Houtaomuga (Tang et al., 2020). One grain of broomcorn millet was found in a ditch where a pottery encrustation was dated to 7,675–7,567 cal. BP in Houtaomuga Phase 2 (Tang et al., 2020). However, without further evidence, such as AMS dating and more millet grains, the presence of formal millet agriculture at the site cannot be further demonstrated. Examination of teeth from the Late Neolithic at Houtaomuga show little evidence for dental caries, similar to that of other hunter-gatherer-fisher groups throughout history (Zhang et al., 2016). Populations that have an increased reliance on domestic crops, such as millet or rice, have increased frequencies of dental caries (Temple and Larsen, 2007). Additionally, teeth from Houtaomuga show evidence for a high protein and low carbohydrate diet through a high prevalence of calculus (Zhang et al., 2016). This dental evidence is consistent with the zooarchaeological evidence at the site, showing limited numbers of domestic animals (Chen et al., 2011; Zhao, 2007).

The earliest evidence for domesticated millets in Northeast China dates to at latest 8,700 cal BP (Houyuan et al., 2009). This has been assessed via starch grain analysis showing evidence for foraging groups preferentially using millet and *Triticeae* from the LGM to the mid-Holocene (Yang et al., 2018). Yang et al. (2018) put this evidence into its environmental context, showing that climate fluctuations may have increased the yield stability, abundance, and availability of these wild plants, and growing conditions continued to favour millets and were more unfavourable to *Triticeae* over time. The domestication process was likely lengthy, and the presence of millets on processing tools likely means they were ground into a flour or meal and cooked in pots as early as 10,000 cal BP (Yang et al., 2012). However, we do not know the extent of this association between millet domestication in northeast China and the people who lived at Houtaomuga. Currently, there is little to no evidence for millet as a staple food at the site, and subsistence is instead focused around a forager-fisher economy for all of the Neolithic (Sebillaud et al., 2021; Tang et al., 2020; Wang and Sebillaud, 2019).

An extensive of the fauna remains from a ditch belonging to the Phase 3 of Houtaomuga (4300 to 3500 BC) allows a glimpse at the hunting and subsistence strategies during this period. Overall, the people who lived at Houtaomuga had very diverse diets and mixed economy (Lai, 2016; Song et al., 2016; Wang et al., 2017). Fish and mammal remains account for the highest proportion of animals at the site (Song et al., 2016; Zhang, 2015). In this phase 3 ditch, the identifiable animal bone specimens include 358 molluscs, 4726 fishes, 10 reptiles, 6 birds, and 1,418 mammals (Song et al., 2016). Four species of mollusc and six fish species were identified

and include varieties of snakehead, catfish, and carp, all of which live in freshwater lakes, rivers, reservoirs, and ponds (Song et al., 2016). Carnivorous fish bones far outnumber herbivorous fish, and Song et al. (2016) argue this could either indicate they were more numerous in the region's freshwater resources or signify a human preference for them. I also want to add this could be caused by the durability of larger fish bones in the archaeological record being that carnivorous fish bones are often larger and thicker than herbivorous fish and might be overrepresented as a result. Some fish bones from this context have grill marks, suggesting they were cooked over a fire (Song et al., 2016). Grill marks are also found on some mollusc species including shredded clams, which can be used as food as well as raw material for pottery and tool production (Song et al., 2016). Regarding pottery use, this could imply pottery was not used for cooking fish and other freshwater species, or more than one cooking style or dish (i.e. soup) including such animals, may have been employed or existed. Residue analysis can further our understanding of these practices.

Auroch (*Bos primigenius*) is the most abundant identifiable species in the faunal assemblage at Houtaomuga during Phase 3 (Song et al., 2016). The majority of these animals are juveniles, signifying these animals were easier to hunt or there was a specific preference for the young individuals (Song et al., 2016). A slight change can be seen between Phase 3 and 4, where fewer fish and molluscs and more auroch were identified in the fourth phase compared to the third, suggesting the beginnings of a shifting economy towards the Late Neolithic (Luan, 2016). During Phase 4, people expanded the scope and intensity of auroch hunting, and this change can be seen in differences in auroch genotypes between Phase 3 and Phase 4 (Luan, 2016). This shift is also present in the faunal analysis of pit features from Phase 4, which reveal a large number of auroch bones are being buried during a short period of time, indicating either an increase in the auroch population or an expanding and improving hunting technique (Luan, 2016). The majority of faunal remains at the site have been subject to weathering, however cattle bones bear the brunt of this damage, leading faunal experts to hypothesize these remains were placed in the open air for some time before burial (Song et al., 2016). Domestic cattle are first present in northeast China during Houtaomuga Phase 4 around 5,000 BP, around the same time domestic cattle appear in northwest China (Luan, 2016). This has led Luan (2016) to suggest there could have been two routes in which domestic cattle were introduced in China: one being the Xinjiang-Hexi Corridor into northwest China, and the other being through the Eurasian steppe and Northeast Asia into northeast China. While cattle domestication begins to show effects during the Late Neolithic, particularly genetically between species in Phase 4,

there are no signs of intensive livestock feeding at Houtaomuga and evidence indicates hunting and fishing are still the primary economic activity (Song et al., 2016; Wang and Sebillaud, 2019).

While there is evidence of human activity on the faunal remains, including evidence of chopping, sawing, cutting, grilling, grinding and drilling, the most common are smashed bones, particularly among auroch (Song et al., 2016). The foremost evidence of this activity occurs on the long bones, which are chopped at the proximal and distal ends, which suggests this action is a part of processing bone marrow or sucking marrow directly from the bone (Figure 1.16)(Luan, 2016; Song et al., 2016). Auroch mandibles unearthed from the site are smashed at the bottom edges, a thinner wall of the bone, which may also be related to marrow acquisition (Zhang, 2015). In addition, there was one bear femur identified in Phase 3 which is also shattered in a manner that Song et al. (2016) argued could be explained by marrow extraction. This specific activity is noteworthy as it has been suggested for other regions in China that pottery was heavily utilized by hunter-gatherer groups for cooking bone marrow either for bone grease production or soups (Patania and Jaffe, 2021).

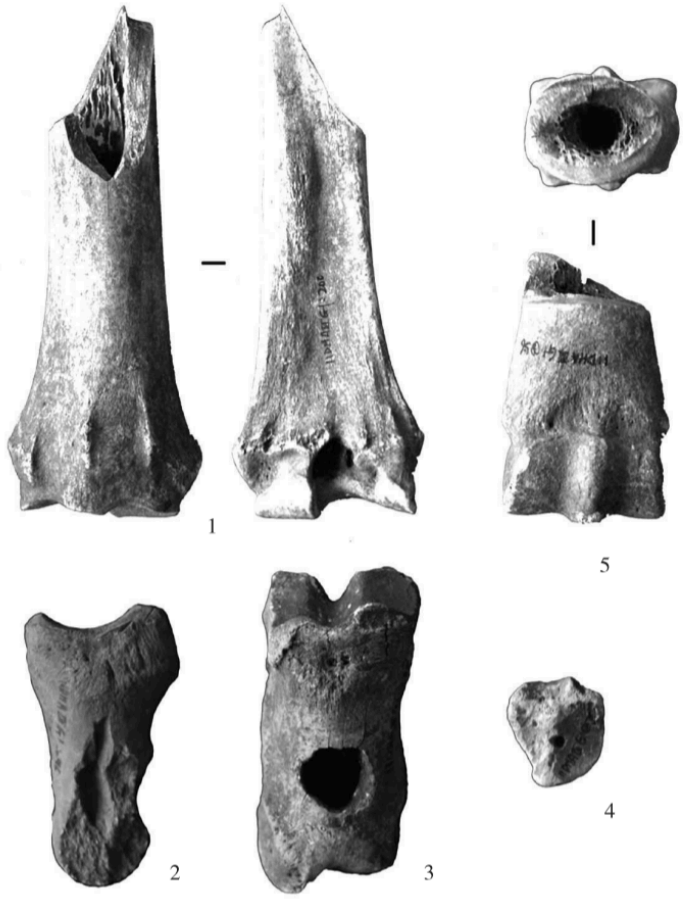


Figure 1.16. Auroch (sp.) long bones with smashing marks and holes (Song et al., 2016, p. 373)

Other evidence of human activity on bones includes burning, which either suggests people roasted food at the time or were using bones as fuel (Song et al., 2016). If the primary method for cooking was roasting, this could have implications for pottery use regarding food production, as this activity might not always be conducted using pottery technology or this technology could have been used only for special occasions or specific circumstances. Such specific circumstances might be socio-cultural, or practical for cooking certain dish types that may require a pot such as soups and stews. A large portion of these burned bones include those not often used for meat such as vertebral bodies and pelvic acetabulum, further supporting the hypothesis these bones are used for fuel, however, grill marks on mammalian long bones and bones from large carnivorous fish suggest otherwise (Song et al., 2016). There is a possibility for multiple cooking techniques and bones used as fuel at the site, and residue analysis can shed light on these practices. In southern and central China, scholars have suggested that hunter-gatherers use pottery for cooking bone marrow either for bone grease production or soups which facilitated group cohesion through feasting (Patania and Jaffe, 2021). It is unknown what implications this may have in northeast China, but the possibility of bone

marrow extraction using pottery either for grease production or moist cooking technologies cannot be ruled out considering the significant evidence for the practice archaeologically. Results from this work could show pottery was used for bone grease rendering, or it could highlight that marrow extraction was undertaken using separate means.

1.1.5.3.4 Lithics

The lithics assemblage at Houtaomuga is a typical collection of north China microlith industry type as defined in section 1.1.5.1. and includes flakes, microblades, cores, bipolar core and wedge-shaped microblade cores, chunks, scrapers, points, polished stone axes, and celts (Figure 1.17) (Chen et al., 2005). The Houtaomuga assemblage is similar to those of the Late Palaeolithic in North China, and some differences may be attributed to regionality (Chen et al., 2005). Lithics are comprised of a wide range of raw materials including felsite, chert, shale, rhyolite, silicic volcanic rock, andesite, agate, obsidian, quartzite, metamorphic mudstone, and sandstone (Chen et al., 2005). The most prominent stone implement group is flakes, followed by microblades, which consist of 16.54% of the total assemblage (Chen et al., 2005). Over. 80% of microblades at the site have been retouched by pressure techniques, but there are also instances of soft hammer percussion retouching (Chen et al., 2005). These blades may have been made for composite tools using bone or antler, similar to other nearby regions utilizing microblade lithic industries (Li and Chen, 2007). Later in the Neolithic, these tool types were sometimes hafted as knives as composite tools in North China (Cui, 2010; Lu, 1998).

What is missing in this research is wear analysis on lithics from the Houtaomuga site. This analysis is currently being undertaken and will be included in the final published report of the site. by This type of analysis has become more common at sites in China in the last few decades and has been beneficial to further understanding subsistence economies (Chen, 2020). Use wear analysis on stone tools would be highly complementary to the research presented here, as it would shed light on technology use and subsistence practices. Additionally, it has been suggested that microblades are evidence for a hunting-based economy that is intensifying its exploitation of animal resources. It would be valuable to confirm this suggestion with use wear or to further highlight the use of these lithic tools for plant exploitation. Plant exploitation is much more difficult to identify in the archaeological record, and residue and use-wear studies

can shed further insight. It would be specifically advantageous at Houtaomuga as we know plants and aquatic resources were extensively exploited (Tang et al., 2020).

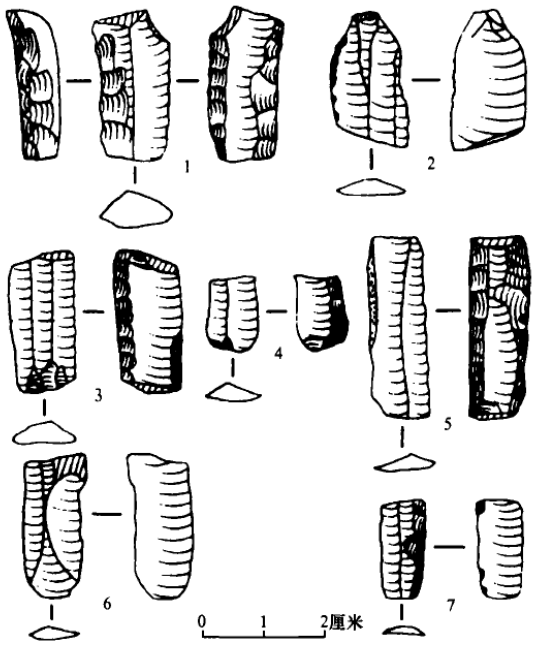


Figure 1.17. Drawing of stone microbaldes from Houtaomuga, surface collection (Chen et al., 2005, p. 10)

1.1.5.3.5 Human Remains

Human burials at Houtaomuga do not have fixed burial practices, and there is no fixed pattern in the direction, posture, or objects in the burials (Xiao, 2014). The average age of death was generally low, with an abnormal sex ratio between men and women, with male burials at a higher occurrence (Xiao, 2014). Xiao (2014) suggested that this abnormal sex ratio could be an indicator that males at Houtaomuga were taking up more dangerous roles or tasks. This is plausible; however, I think it is important to note that burials overall are not numerous at the site during the Neolithic, making the sample size likely unrepresentative. Moreover, it has been suggested that the site was only occupied seasonally (Luan, 2016; Sebillaud et al., 2021), further reducing our confidence in the representativeness of the burials overall. We can assume there is likely another location where people from Houtaomuga lived and buried individuals which are missing from the analysis of the population as a whole.

Phase 1 includes one burial of an adult male with a height of 1.6m with no pathological phenomena (Xiao, 2014). Teeth from this individual (tomb M45) exhibit severe signs of dentition wear which can be linked to the consumption of shellfish containing too much sand

(Xiao, 2014). Phase 2 contains burials of 4 individuals and pathological observations found an instance of premature sagittal suture healing (Xiao, 2014). Significant tooth wear appears and coexisted with the appearance of dental calculus and alveolar bone resorption (Xiao, 2014). These instances along with ubiquitous enamel hypoplasia possibly suggest a poor diet among individuals living during this period (Xiao, 2014). An increased number of burials belong to Phase 3, with 13 earthen shaft tomb of 14 individuals comprised of 4 adult males and 10 immature individuals, and this increased along with an increased number house features during this phase (Sebillaud et al., 2021; Xiao, 2014). Generally, pathological phenomena include osteoma, early healing of skull sutures, abrasions of the teeth, enamel hypoplasia, and suspected tumours (Xiao, 2014). These conditions, especially teeth conditions related to diet, could have implications for pottery use at the site. However, it is again important to remember the unrepresentative sample size. Furthermore, the snapshot of human lifeways we see through human remains is skewed towards individuals likely in the worst health of a population. This is particularly important to remember at a site that was not consistently occupied and many individuals belonging to this population were not buried here.

There is no obvious consistency in burial practices, as there are many burial orientations and a broad range of object combinations (Xiao, 2014). This great range of variation may also be related to the small and likely incomplete sample size of human burials. 8 of 13 tombs of the Neolithic periods do not contain burial objects, while two immature individuals were buried with clay pots, and stone tools in the co-burial and one adult burial (Xiao, 2014). However, there is one burial of an individual around 3 years of age (M92) which included a large number of atypical burial objects, suggesting a difference in socioeconomic status (Xiao, 2014). While no dental caries were identified in individuals from Phases 1 and 2, a larger number of individuals exhibit this condition in Phase 3 (Xiao, 2014). This suggests a change in diet, as groups whose diets are composed of a large portion of carbohydrates will have a higher incidence of dental caries however it is unclear if this change suggests a change in diet during Phase 3 or if this is the result in changes in sample size (Temple and Larsen, 2007; Xiao, 2014). The overall degree of wear on teeth during this phase increases, but it is speculated that this may be related to other dental diseases rather than diet shifts (Xiao, 2014). Phase 4 burials were located in a part of the site where remains are dense, and some bones are displaced (Xiao, 2014). The four burials from this phase are all earthen shaft tomb with burial objects including pottery and shell ornaments, bone artifacts and stone tools (Xiao, 2014). The amount of objects is small at about one of each type per individual (Xiao, 2014).



Figure 1.18. Example of ICM from burial M90 (Xiao, 2014, p. 48)

Intentional cranial modification (ICM) is present in 11 skulls from Houtaomuga belonging to Phases 1, 3, and 4 burials (Figure 1.18), dating between 12,000 and 5,000 BP (overall incidence 58%) (Zhang et al., 2019). This denotes the earliest incidence of the practice and is the longest *in situ* duration at a single Neolithic site (Zhang et al., 2019). This practice is not fully understood, but we can determine it had a long existence and was selective in some way (Zhang et al., 2019). The practice has no sex-specific selectivity and was present in both males and females (Zhang et al., 2019). However, there is a possibility of the practice being present or lacking within families, as highlighted in shared tomb M54 from Phase 2 containing three individuals with no modifications and contrarywise shared tomb M87 in Phase 3 containing two individuals suspected of being of the same family both have signs of ICM (Zhang et al., 2019). Moreover, it is feasible the practice was selective based on privilege/class, as burial M92 containing a 3-year-old child with ICM also contains a substantial number of funerary objects, suggesting a significant social status of some kind (Xiao, 2014; Zhang et al., 2019). Tomb M91 (Phase 4) contained an adult female with ICM and strips of red pigment on the top of the frontal bone and parietal bone along the sagittal direction which are described by the bioarcheologist as “cinnabar-like” (Xiao, 2014). The tomb also contains a large number of shell ornaments found around the ankles making it a very unique burial among the population (Xiao, 2014). This also further supports the notion that the practice of ICM was implemented on

individuals selectively, possibly depending on family affiliation or socioeconomic conditions (Xiao, 2014; Zhang et al., 2019).

1.1.5.3.6 Pottery

Understanding human remains at the site sheds light on past lifestyles including diet. This thesis aims to understand pottery use with human diets in the context of these Neolithic lifeways over the first four phases at Houtaomuga. To fully understand pottery development and continued use, I will also discuss what we know about pottery production and decoration. This will also aid in our understanding of the relationship (or lack thereof) of Neolithic pottery in northeast China and surrounding regions which also see drastic changes in diet and pottery technology during the same time.

Early Neolithic pottery from Houtaomuga Phase 1 displays some similar characteristics to early pottery in China and the Russian Far East, with regional variation in object shapes, decoration, and paste types (Wang and Sebillaud, 2019; Yanshina, 2017). Current research on the production techniques of these vessels is being undertaken by Pauline A. Duval, and the results of this work will provide further insight. At present, it is believed vessels are handmade by coiling seemingly with local clay and have various thicknesses throughout. They are generally coarse wares with a brittle consistency, indicating low-temperature firing (all well below 900ºC), likely in pits or on surface rather than kilns, and yellow-brown, greyish-brown, or grey with decoration covering the majority of the vessel (Wang and Sebillaud, 2019). This decoration is described by archaeologists as horizontally orientated chevrons and parallel lines of comb-shaped points made by an instrument (Figure 1.19; Wang and Sebillaud, 2019). The impressions are deep (2-3mm) and some of the designs are seemingly uninterrupted as if made from a rolling tool (Wang and Sebillaud, 2019, p. 79). It has been hypothesized this all over treatment was intentional to reduce the vessel's vulnerability to thermal shock relating to use as cooking pots (Wang and Sebillaud, 2019). Inclusions/tempering often includes sand and organic plant fibres and shell in Phase 2, 3 and 4. Currently, more research on clay provenance, firing temperature, and production techniques are ongoing (Wang and Sebillaud, 2019). Houtaomuga Phase 1 remains yielded three main vessel shapes; large cylindrical jars, large-opening curved-belly jars, and bowls (Wang and Sebillaud, 2019). More details on the specific

differences between pottery Phases at the site and accompanying illustrations can be seen in materials section 2.3.4.

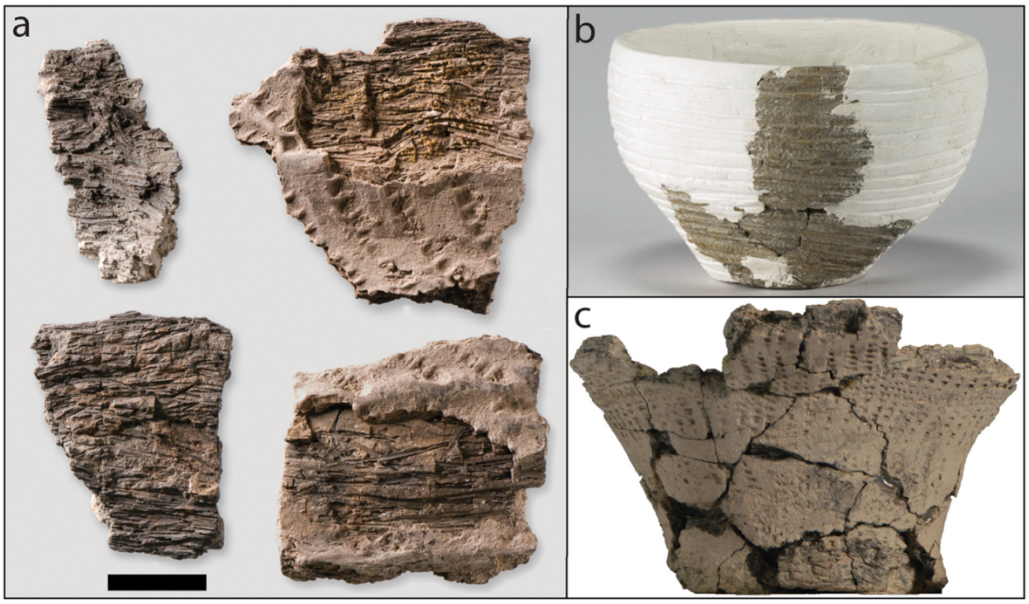


Figure 1.19. Pottery from Ditch 18, dating from Phase I at Houtaomuga. (a) plant fibres impressions of cattail embedded in clay (b) a reconstructed flat-bottom bowl; and (c) a bottom part of a vessel (Tang et al., 2020).

1.1.6 Residue Analysis on Neolithic Pottery in Northeast Asia

Early Neolithic pottery in northeast Asia emerged during a period of shifting environments and lifeways. Knowing the function of these vessels, including those at Houtaomuga, helps us understand motivations for their production and widespread adoption. This section will outline results from residue analysis in the region and what we currently do and do not know about pottery use during the Neolithic. An overview of this research in northeast Asia is presented here as it relates to the current study of pottery at Houtaomuga. Descriptions of the types of methods used for residue analysis including their limitations can be found in section 1.2. Specific sites and details as they directly compare to the results of this thesis are discussed in sections 4.8.

Scholars have argued that these vessels in northeast Asia during the Neolithic were used as cooking pots, suggesting that people needed to cook more plant and/or aquatic resources than in previous years rather than large game (Wang and Sebillaud, 2019). The current prevailing hypothesis is that following initial climatic shifts at the Last Glacial Maximum – a time where

large animals were the predominant prey for hunter-gatherers – the inhabitants of northeast China began progressively relying more on fish and novel plant resources. However, we have little data confirming plant and aquatic resources were lacking in Palaeolithic diets before this period. It is hypothesized that the need for acorns to be soaked and boiled for long periods to remove tannins and be edible to humans drove the production of pottery in this area (Liu and Chen, 2012). However, little to no evidence for acorn exploitation has been found in Northeast China and Russia's Far East, leading some to hypothesize the importance of acorns was mainly limited to the Japanese archipelago while people further inland relied more heavily on freshwater fishing (Kuzmin, 2006; Wang and Sebillaud, 2019).

Several types of residue analysis have been conducted on early pottery from Japan and Russia's Far East, however, few similar studies have been undertaken in China so far. Such research has been extensive in Japan, and a complex picture of pottery use through deep time has been established and may provide additional insight into the mainland regions (Craig et al., 2013; Lucquin et al., 2016). This research began when Craig et al. (2013) successfully demonstrated that viable lipid residues can be obtained from ceramics dated up to 15,000 cal BP. Pottery from this study dates to Japan's Incipient Jōmon Period (15,000 to 11,800 cal BP) at the end of the late Pleistocene epoch, and it has been hypothesized that at that time humans had to adapt to changing climates. Lipid residues show evidence for dietary shifts towards more marine resources, possibly as an adaption to shifting environmental conditions (Craig et al., 2013; Lucquin et al., 2016).

Similar residue analysis work has been undertaken on material from the Korean Peninsula. The earliest pottery here dates to the early Holocene (~7,900 – 6,800 BP), slightly later than the surrounding regions. This pottery was investigated using chemical and isotopic residue analysis and results indicate that they were mostly used to process marine resources, similar to results from the Japanese Archipelago (Shoda et al., 2017). This is in contrast with palaeobotanical and zooarchaeological evidence indicating that a variety of food resources were exploited, leading scholars to hypothesize about the processing or storage of aquatic oils (Shoda et al., 2017). Results from the area are comparable to other hunter-gatherer pottery use studies in Japan, Europe, and Alaska (Admiraal et al., 2019; Craig et al., 2011; Shoda et al., 2020). The wide geographic range of this practice is notable, as it may suggest this practice among hunter-gatherer fishers is a pattern. This could suggest that the practice is expected in the region of

northeast China in question due to the early association of pottery presence and marshland environment.

Like on the Japanese Archipelago and the Korean Peninsula, the emergence of ceramic vessel technology in modern-day Russia and China during the Terminal Pleistocene and early Holocene has been associated with hunter-gatherer strategies for collecting, processing, and consuming new plant food which resulted as an adaption to the shifting climate. During the Upper Palaeolithic, humans were assumed to be more reliant on megafauna than plants, which go extinct at the end of this period (Barnes, 2015). Residue analysis of ceramics from Russia's Far East has been undertaken, and mainly show evidence for the processing of aquatic resources, but also ruminant animal fats in many circumstances (Gibbs et al., 2017; Lucquin et al., 2018; Shoda et al., 2020, 2018).

Lipid residue analysis results from the Lower Amur in Russia's Far East has shown that early pottery from the Osipovka Culture (16,200-10,700 cal BP) indicate that pots were used to process aquatic oils, not unlike those analysed in Late Glacial Japan, and Sakhalin Island (Gibbs et al., 2017; Shoda et al., 2020). Geographically, these samples are closest to the site of Houtaomuga and both regions shared environmental characteristics from the Late Glacial Period to the Early Holocene (Shoda et al., 2020). Early pottery from northeast China was likely used for a similar purpose, considering the clear importance aquatic resources had at the time (Kunikita et al., 2017; Sebillaud et al., 2021; Wang and Sebillaud, 2019). However, early pottery sites in the Lower Amur have very few ceramic samples, and sites were not occupied for long periods. This has led Shoda et al. (2020) to suggest that pottery in Russia's Far East was used for restricted purposes, such as for novel or ceremonial foods. This is not the case at Houtaomuga, as the site was possibly occupied seasonally during phase I, however then has a long and extensive history of permanent occupation with a large ceramic assemblage (Sebillaud et al., 2021). There is a possibility that early pottery at Houtaomuga served a similar purpose initially, and this may change over time, or an entirely different ceramic function was employed from the beginning. This study will shed light on this question by analysing samples from all Neolithic periods at Houtaomuga to trace changes over time. Moreover, early Holocene pottery from the Amur region in Russia has distinct regional variability in vessel forms and decorations, with northeast China also having its own distinct differences (Yanshina, 2017). Molecular and isotopic characterization of lipids from these various ceramics within Russia has shown that these distinct pottery traditions had different use patterns, with those in the

Middle Amur being used less for aquatic products and more for ruminant animal fats (Shoda et al., 2020). The pottery in northeast China could also maintain its own distinct use patterns, and residue analysis will shed further light on this phenomenon by comparing results from northeast China to current research in other regions of northeast Asia.

Little analyses have been undertaken in mainland China on material from the early Neolithic, and work that has been published is based on a very small number of samples (Kunikita et al., 2017; Shoda et al., 2018). Recent research by Shoda et al. (2018) analysed 36 pottery samples from four archaeological sites in South Korea, Sakhalin Islands, Japan, and China: an immense geographical range with an average of only 9 sherds per site with several of the sites being multi-phased. The Chinese site in question, Tianluoshan, is located in Zhejiang province and dates to 5,000 – 4,700 cal BC (Shoda et al., 2018). The date range for these samples is far later than many of the sites used for comparison and significantly further south, making a comparison with the research area of this thesis not particularly useful.

A small selection of ceramics from the Houtaomuga has been analysed for former contents via $\delta^{13}C$ and $\delta^{15}N$ measurements (Kunikita et al., 2017). Four samples tested at the University of Tokyo gave values of carbon isotope $\delta^{13}C$ were all in the −25.8‰ to 31.3‰ range, which coincides with the range of change of $\delta^{13}C$ in the C_3 type plants. In combination values of carbon isotope, $\delta^{13}C$ were all in the −25.8‰ to −31.3‰ range likely coming from the fibre tempering of the vessels (Kunikita et al., 2017; Wang and Sebillaud, 2019). In carbonized food crust from two samples, the nitrogen $\delta^{15}N$ isotope value is higher than that for modern C_3 plants, showing that these samples contained animal protein, most certainly coming from carnivore animals or freshwater fish (Kunikita et al., 2017). With the methods employed, it is not possible to determine the difference between animal and freshwater fish though this analysis and further characterization and modelling would be required. Additionally, this type of analysis would only be able to distinguish between C_3 and C_4 plants, providing little information on individually exploited plant resources. However, a procedure that can identify polyunsaturated fatty acids or ω-(o-alkylphenyl)alkanoic acids (APAA's) from fish or biomarkers of plants along with monounsaturated fatty acids would be ideal. These components are highly susceptible to thermal degradation, and current methods for their analysis may be damaging. This thesis will investigate methods to mitigate these negative effects and include isotopic characterisation of residues to better determine aquatic processing. It is worth mentioning that

other proxies for subsistence would be useful to include in this research, though, some remain unavailable this time. Late Palaeolithic to Early Neolithic transition pottery sites in northeast China were not discovered until 2011, meaning there has not been a long period of time for scientific analysis such as isotopic analysis on human bones, but this is ongoing. Such research can take many years as there are certain ethical and sampling considerations. Additionally, data from northeast China have not been researched and published to the same degree as other regions (i.e., Eastern Europe or Japan) leaving some gaps in the archaeological record and making comparisons to nearby sites from the same period more difficult. I have included research on subsistence practices, as palaeobotanical and faunal analysis which has already been undertaken, however more research in the future using additional lines of evidence here would be useful.

1.1.7 Summary

Research on the Neolithic in East Asia places particular emphasis on pottery technology rather than economic practices. This period, beginning around 13,000 cal BP in northeast China, sees changes in human behaviour and technology specifically regarding lithics, grinding stones, and pottery vessels. The shift in lifeways involving ceramic use by hunter-gatherers in this area has been noted to be of interest by many scholars (Wang and Sebillaud, 2019). This thesis intends to shed light on the use of these vessels from northeast China from their onset and throughout the Neolithic at the site of Houtaomuga to better understand the process of this long-term transition. This research will discuss specific lifestyles of foragers who have economies based on aquatic resources and who use pottery during a period when many similar peoples do not. After this period, northeast China becomes a centre for millet domestication, and we know human relationships with these plants begin far before in the Neolithic (Shelach-Lavi and Tu, 2017; Tang et al., 2020). Results from this thesis will be put in context with residue analysis research at sites in neighbouring regions, where pottery is similar in decoration but with regional variability.

While it seems highly likely that pottery in northeast China was used for cooking fish and other aquatic resources similar to neighbouring regions in Russia and Japan, it is also conceivable that pottery vessels were used for primarily plant exploitation – particularly considering changing environmental conditions during this time leading to a warmer and wet environment

and there is evidence for plant exploitation at the site (Stebich et al., 2015; Tang et al., 2020). It is plausible that the adoption of pottery technology in this region could be linked to the exploitation of plant species such as sedges and wetland grasses not exploited during the Palaeolithic which may require the technology for long periods of boiling to be desirable for consumption. Additionally, pottery technology may be a driving force for more extensive plant cultivation, allowing for semi-sedentary lifeways (Rice, 1999). Pottery can be seen as an adaptive tool, allowing people to utilize more resources from the environment, aiding in food security (Grillo, 2014). In addition to extensive exploitation of freshwater fish and various plant resources through vessel technology, people in northeast Asia could thus have thrived in this region more permanently and relied less on mobile routines. However, without additional residue analysis of this pottery in China specifically, the functions remain unknown and many of these hypotheses remain untested for the area.

This thesis aims to close some gaps in our understanding of the Neolithic period in northeast China through pottery residue analysis at Houtaomuga. Pottery is seen as a marker for significant dietary and social changes, and this technology change was preceded by another significant technology change in lithic technology during the Palaeolithic. This change in lithics is also seen to suggest significant changes in lifeways, possibly in the form of intensified hunting, however, this lithic use also continued when people transition to using pottery. The implications and relationships of these changes are still not fully understood. Elston et al. (2011) believes the change in lithic technology indicated intensified animal exploitation and early pottery use is thus interpreted as a result of this continued trend, and vessels were used for bone grease rendering. Residue analysis on pottery can shed further insight into this hypothesis. Faunal remains at Houtaomuga show evidence for bone marrow extraction, and residue analysis here can provide insight on the processing of this material.

Settlement patterns at Houtaomuga show people were focused on remaining on high ground near lakes and marshlands. This is likely related to the protection or to avoid flooding. Residue analysis can shed light on specific activity areas and resource exploitation within the site. Results of this research will be compared to similar studies in nearby sites in Russia and further sites in Japan to understand how pottery use changes or stays the same over different locations. Subsistence practices show wild plants were an important part of the diet at Houtaomuga, and human teeth show evidence of wear associated with plant use. Residue analysis will provide further insight into the processing of these resources to see if they were broken down through

cooking. It is important to consider the small sample size of human remains at the site however, other proxies for diet are much needed, here residue analysis can provide further insight. Large mammals, fish bones, and shells at the site show evidence of being cooked over an open flame. Pottery residues will determine if this was the primary method of cooking, or if these resources were also cooked in pottery containers. It has been suggested that Houtaomuga was used as a seasonal camp for auroch hunting based on faunal remains and settlement patterns in Phase 4. This research could shed light on these processes, if pottery is shown to be primarily used for such resources.

Residue analysis needed to answer the archaeological questions proposed here about northeast China has been successfully applied to the archaeological record in neighbouring regions and around the world. A further description of these methods, their limitations, and their ability to teach us about past populations are described below.

1.2 Analytical background – Methods for the characterization of archaeological lipids

Archaeologists have long questioned how people used ceramics. This is partially due to the desire to understand diets and subsistence practices.[7] Archaeologists are frequently concerned with how people engage with their environments through animal and plant management and how, where, and when agricultural economies emerge and what effects they have on populations and environmental carrying capacities, and what dietary factors affect health through prehistory (Kintigh et al., 2014). Among artifacts often preserved in archaeological contexts, pottery can provide information on various human activities including subsistence. To understand how pottery was used in the past, chemical methods have been applied to former pot contents, encrusted remnants, and absorbed organic material trapped within the ceramic walls (Heron and Evershed, 1993). This type of applied chemistry is not a recent development. In the early 20th century, Gill (1906) analysed the still intact contents of a Mycenaeans vase found in Egypt housed at the Museum of Fine Arts, Boston. At the time, such applications were still in development and produced results without a high degree of specificity. Since the 1970's,

[7] This is also to understand the motivations for ceramic production; however, this is not a main theme discussed here and beyond the scope of this work.

significant developments in the field of analytical chemistry – namely in chromatographic and mass spectrometric instrumentation – have driven the application of organic residue analysis in archaeological pottery (Devièse et al., 2018; Heron et al., 1991; Heron and Evershed, 1993; Roffet-Salque et al., 2017). Modern analytical instrumentation is particularly well suited for archaeological applications due to their high sensitivities; being compatible with low concentrations of preserved organic material from archaeological contexts. These applied chemistry techniques are highly useful to answer a wide variety of archaeological questions about human subsistence, vessel technology, and food preferences for example.

1.2.1 Lipids

Much of the published literature in archaeological and conservation science focuses on either proteins, lipids, polysaccharides, carbohydrates, and amino acids. Lipids are hydrophobic and insoluble in water making them less prone to leaching through rain or groundwater when buried at archaeological sites than other organic components (i.e. proteins). These biomolecules are soluble in nonpolar solvent, making them easily extractable with liquid solvents for identification and quantification. They include fatty acids, waxes, sterols, fat-soluble vitamins, monoglycerides, diglycerides, and triglycerides (Gunstone, 1996).

Lipids are preferred for pottery analysis, as they are well preserved over extensive time scales, and can be indicative of the foodstuffs produced or prepared [8] and stored in ceramic vessels with a high degree of specificity (Evershed, 2008a; Roffet-Salque et al., 2017). Lipids are common in most foodstuffs, making them ideal proxies for specific research regarding pottery use, human diet and subsistence in the past, and these studies have gained popularity in the last 20 years as highlighted in figure 1.20 (Barnard et al., 2005; Evershed, 2008a; Malainey et al., 2007). Lipids are high-density energy sources and are used for both cellular functions and energy storage in the human body (Hedges, 2009). Certain specific lipids cannot be synthesised by humans and must be obtained from food, including mainly unsaturated fatty acids from plants such as linoleic acid ($C_{18:2}$) and polyunsaturated acids from fish including eicosapentaenoic acid (EPA, $C_{20:5}$) (Hedges, 2009). Ancient lipids do not only shed light on

[8] While this research focuses primarily on the relationship between food and pottery, I use the phasing "foodstuffs produced" as pottery contents may not exclusively be food related. Additionally, I avoid the term "cooking pots" unless there is additional empirical evidence to support the pots use for cooking.

ancient food practices, but also technological innovations. For instance, lipidic organic materials such as resins and waxes can serve as waterproofing agents for ceramic containers for liquid transport (Colombini et al., 2003; Robinson et al., 1987) and adhesives for repair (Charters et al., 1993). Additionally, lipids can be indicative of non-food related vessel use i.e. oil lamps (Heron et al., 2013). Lipidic substances have also been used as varnishes, binders, glues, and pitches in works of art (Charters et al., 1993; Colombini et al., 2010; Drieu, 2018; Duce et al., 2015; Fox et al., 1995; Regert, 2004).

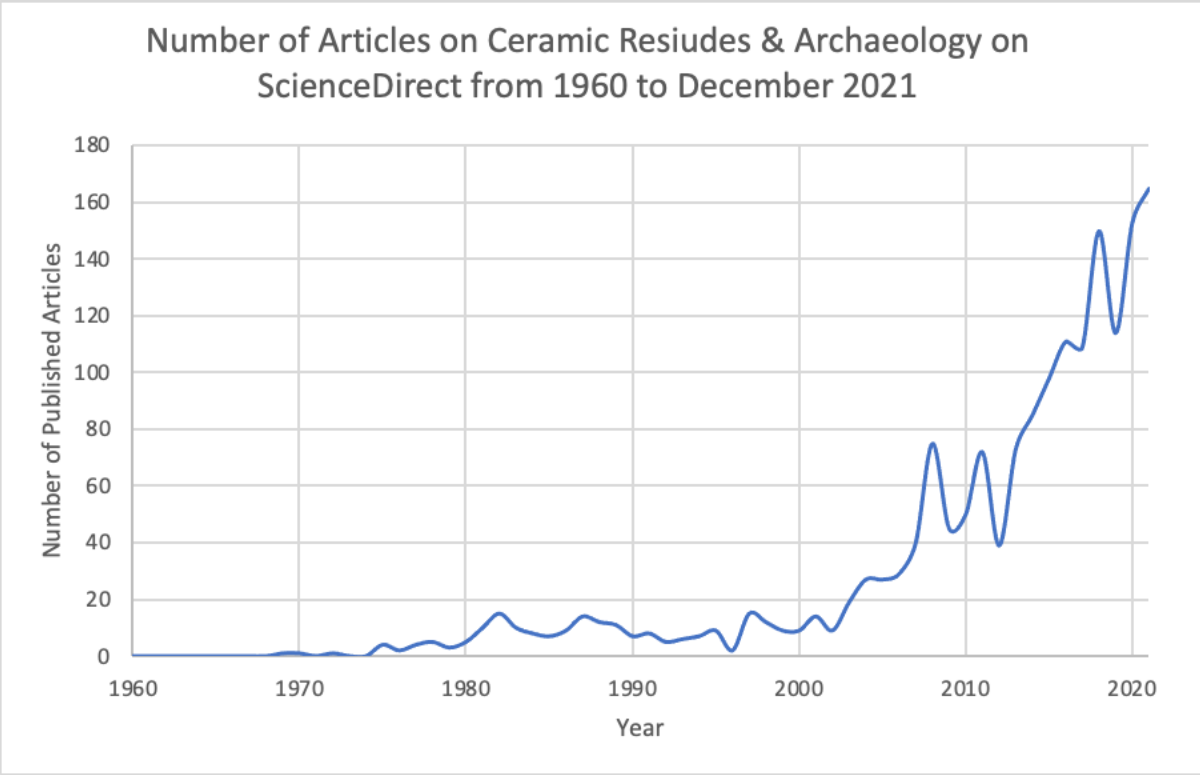


Figure 1.20. Popularity chart of published articles on organic residue analysis of archaeological pottery.

1.2.2 Preservation of Residues

Organic residue preserved on and/or in archaeological pottery falls into two main categories: 1) carbonised encrustations on the surface of the vessel, likely food remnants from a cooking event or 2) microscopic organic remains which have been absorbed into the porous ceramic body of the vessel. Absorbed residues are preferred over surface residues as the latter is more susceptible to excavation and curation contamination than the former (Roffet-Salque et al., 2017). The bulk of lipid material is preserved at the top of vessels close to the rim and halfway through the ceramic matrix based on experimental studies (Evershed, 2008b). However,

absorbed lipid recovery is also influenced by the porosity of the ceramic, the surface finish/treatment (as ceramics lined with a resin or burnished will be less permeable), and the chemical composition of the ceramic – perhaps affecting the chemical interactions that occur between the organic lipid material and the inorganic matrix. It has been discovered by the empirical observation that lipid material is better retained in cooking pots over storage pots, but this phenomenon has yet to be explored in-depth (Correa-Ascencio and Evershed, 2014).

A potential third category would be vessels that have their former contents fully intact, however, preservation of intact food within a pot are few and far between. This is primarily because vessels enter the archaeological record this way infrequently, and ancient food is only preserved under special conditions. For example, due to immaculate preservation conditions, the German archaeological institute of Istanbul discovered an underground storage facility in Hattusha, north-central Turkey containing enough carbonised grain to feed at least 20,000 people for a year (Bogaard, 2017: Seeher, 2006). Direct evidence for kefir dairy in the Early Bronze Age in Xinjiang, China has been identified through proteomic analysis of preserved cheese in a pottery vessel from the Xiaohe Tombs (Yang et al., 2014). Preservation at this site is unprecedented, with over 330 tombs naturally mummified due to the dry environment. Additionally, there are a few instances of favourable preservation conditions from waterlogged contexts (Colombini et al., 2003; Stern et al., 2008).

1.2.3 Extraction

All organic remains from archaeological contexts are complex mixtures that often include inorganic components including sand, dirt, and ceramic powder. To analyse these remains, the organic fraction must be extracted and isolated from these inorganic impurities. Various types of procedures to achieve this are currently used in the field and a summary of their mechanisms and limitations is presented below.

1.2.3.1 Extraction with Liquid Solvents

Extraction of lipids from archaeological ceramics is most often done using liquid solvents in current research practices. The process uses a polar solvent to extract lipid material from the ceramic matrix and remove any inorganic material from the sample prior to GC-MS analysis or any other characterisation method. A summary of these practices and their limitations is presented here, as this work focuses on improving current protocols for this type of analysis.

1.2.3.1.1 Traditional Extraction Procedures

To learn more about ancient vessel use and diet, analytical methods to characterise ceramic residue have been developed over the last 30 years. Initial procedures for the extraction of lipids from their ceramic environment were second nature in the field of analytical chemistry and similar to those used for identifying organic components in art objects such as paint binders. Frequently, and still being applied in current research, archaeological scientists extract lipids using a mixture of chloroform and methanol ($CHCl_3$/MeOH), or dichloromethane (CH_2Cl_2) (Mills and White, 1987). This technique can be applied to encrusted and absorbed residues; however, applications of absorbed residues require the ceramic to be crushed into a powder before extraction to facilitate the extraction process. This technique has been coupled with a variety of chromatographic methods including Thin Layer Chromatography (TLC), GC, GC-MS, and HPLC; GC-MS and GC-C-IRMS are by far the most common used today, and discussed further in subsequent sections of this work (Blanco-Zubiaguirre et al., 2018; Charters et al., 1993; Grillo et al., 2020; Roffet-Salque et al., 2017).

The viability of this type of analysis has been examined and tested for several years. There are still some questions that remain unanswered, however. Most recently, Reber et al. (2018) assessed the fate of lipids dispersed into the clay matrix after the pottery firing process. This is a particularly important experimental examination, particularly in the Americas where shell tempered pottery is common in some regions during different time periods. Reber et al. (2018) found that the clay firing process removes natural occurring alkyl lipids, as well as those which may be introduced during the firing process. Interestingly, they also found that diterpenoid lipids were instead introduced to the ceramic body as condensate from the pinewood used as fuel during open-air firings. It would be interesting in the future to test this archaeologically by examining the residues from assemblages of (assumed) ceramic wasters from a pottery production centre that may have used pine wood.

Dudd, Regert, and Evershed (1998) examined the microbial lipid contributions to lipid profiles of archaeological ceramics through experimental laboratory ageing of fats and oils. It was found that microbial interaction was found to be minimal – a conclusion also drawn by Heron and Evershed (1993). This hypothesis has led researchers to commonly remove the outer 1mm of the ceramic matrix as a precaution to remove any environmental lipids, or contamination from excavators in the field or museum (i.e. fingerprint oils from handling). Little empirical

research has been done on microbial effects on archaeological material such as testing the removed ceramic powder or testing surrounding soil. However, a recent study on ceramic porosity and the preservation of lipids has found that this attribute significantly affects lipid preservation, but the burial environment is still a contributing factor (Drieu et al., 2019). Furthermore, lipid profiles are not homogenous throughout individual vessels. Evershed (2008b) examined different modern cooking pots and determined the highest percentage of lipid material to be held towards the rim of the vessel – particularly in instances of food boiling which causes hydrophobic lipids to rise to the brim of the vessel. Besides this experimental study, little experimental research has been done on vessels used for other types of food procurement or additional uses – i.e. storing, roasting, soaking, fermenting, cosmetics, paints etc. This would be particularly interesting for vessels used for beer making, as it has been hypothesised that porous ceramic provides a place for yeast to inhabit, making the fermentation process simpler, but little work has been done to test this. Lastly, it is unclear which environmental conditions provide the best preservation of lipid residues from archaeological vessels. It has been observed that samples from hot and dry environments and some from acidic soils do not retain high amounts of lipid material (Evershed, 2008b). This early observation has yet to be empirically tested, but several additional factors outside of the local environmental conditions are likely to be at play as lipid preservation is not always consistent across sites. These include the mineral composition of the ceramic and the surrounding soil of the burial environment (if applicable), the use history of the pot, and the conditions in which the vessel was deposited/entered the archaeological record.

In an early application, Robinson (1987) examined a waterproofing agent preserved on wood on the Mary Rose (Tudor) and Etruscan shipwrecks. These materials of tar and pitch were compared with modern samples and found to be derived from pine woods based on constituent tricyclic diterpenoids. Results were obtained through several analytical techniques including elemental analysis, IR, NMR spectroscopy, and GC-MS. In 1993, Charters et al. (1993) identified an adhesive used to repair a roman jar using a liquid chloroform/methanol (2:1 v/v) solvent extraction technique and a series of analytical techniques including TLC, GC, and GC-MS. The lipid composition of the extracted organic material was compared with the composition of artificially aged birch bark tar. Both samples had a series of pentacyclic triterpenoid compounds, known as a series of biomarkers of resin from the *Pinus* genus of plants (Mills and White, 1987). These early examinations found GC-MS to be highly compatible with low concentrations of organic material, as well as with the quantification of

individual biomarkers. This is the primary analytical tool still used today (Bonaduce et al., 2016; Young et al., 2012).

Less common in absorbed residue analysis is the application of alkali saponification techniques for the extraction and derivatization of ancient lipid material. This process is most often applied to encrusted residues, or amorphous lipid samples such as those from works of art (i.e. Bonaduce and Colombini, 2004). Since this procedure is not as commonly applied in the extraction of absorbed residues, it will not be considered fully in this dissertation.

1.2.3.1.2 Acidified Methanol Extraction

Traditional liquid extraction techniques often result in low lipid yields from archaeological ceramics because lipid material strongly binds to the inorganic ceramic body. To address this issue, in 2013-2014, two groups from the University of Bristol and the University of York simultaneously developed an acidified methanol extraction procedure intending to increase organic extractions from archaeological ceramics (Correa-Ascencio and Evershed, 2014; Craig et al., 2013). This method was further defined and tested by Papakosta et al. (2015). By utilizing a sulfuric acid and methanol (H_2SO_4, MeOH 2% v/v) solution, this procedure provides an alternative to traditional chloroform-methanol procedures with increased lipid recovery, lower cost, and high throughput. Its success can be attributed to the ability of the acid solution to break strong interactions between the lipids and the ceramic matrix, releasing free and bound lipids from the surface as well as deeper layers of the ceramic (Correa-Ascencio and Evershed, 2014, p. 1334). These interactions are formed due to the acidic or basic nature of the edges of the crystalline lattice structure of aluminosilicates which make up the ceramic fabric (Correa-Ascencio and Evershed, 2014; Parker and Rae, 1998). The strength and amount of these interactions are determined by the chemical nature of the absorbed residue, e.g. hydroxyl and carboxyl molecules with one or more polar functional group which increases the abundance and strength of interactions with the ceramic fabric. Subsequently, these molecules are more difficult to extract with organic solvents than less- or non-polar molecules (Correa-Ascencio and Evershed, 2014).

The acidified methanol extraction protocol laid out by Correa-Ascencio and Evershed (2014) and Craig et al. (2013) has the unique ability to simultaneously allow for the extraction and

methylation of lipid residues, shortening time and materials required for further chromatographic analysis. As a result, more research using this method and larger sample sizes have been undertaken (e.g. Gibbs et al., 2017). This increase in method application in the mid-2010s can be visualised in figure 1.16. This allows for the ability to learn more about past populations through archaeological ceramics, particularly regarding larger scope analyses encompassing larger spatial or temporal scales. Recent work about food production and pastoral trajectories in the Green Sahara period (Dunne et al., 2012) and dairying in the early Neolithic in Europe (Cramp et al., 2014) are examples of this expanded research.

While recent developments in extraction and analytical techniques have been quite promising, limitations are still prevalent. Primarily, the acid and base catalysed hydrolysis reaction results in the loss of compositional information when triacylglycerols or wax esters are hydrolysed (Roffet-Salque et al., 2017). This means that larger lipid molecules are broken down into constituent free fatty acids. Additionally, all liquid solvent extraction procedures currently require the ceramic sherd to be ground into a powder for analysis. This process is highly destructive and makes the analysis largely unattractive for finite cultural materials – particularly from museum contexts. Lastly, while cheaper than traditional liquid extraction, this technique is still expensive and time-consuming making it unfavourable for large sample sizes, and tight funding budgets.

These limitations are well known, and sometimes procedures to mitigate their effects are undertaken. Unfortunately, these mitigation effects often add extra time and analysis costs. Standard method applications include a basic solvent extraction and trimethylsilylation (BSTFA) of extracts for GC-MS analysis before acidified methanol extractions coupled with GC-C-IRMS is undertaken (Dunne et al., 2019a; Papakosta et al., 2015). This is mainly to account for surviving TAGs or waxes and pitches, often from plants or bees. Thus, some researchers chose to add an additional step where samples are reextracted with either KOH or BF_3 to target short-chain carboxylic compounds found in high quantities in fruit products, which are generally insoluble in organic compounds (Dunne et al., 2019a; Rageot et al., 2019).

1.2.3.2 Supercritical Fluid Extraction

Commonly applied in other fields but new in archaeology is the use of supercritical fluid for the extraction of lipids from archaeological pottery over liquid solvents. Supercritical fluid extraction (SFE) has currently only been published in one archaeological application involving pottery (Devièse et al., 2018). Because the application of this extraction method is still novel in the field, a detailed description of the basic principles and current state of research is given here. Some of these principles also include the use of supercritical fluid for chromatographic separations (SFC). The majority of the discussion here will only include SFE as this technique was further developed and used in this thesis, however, I will mention SFC in certain sections where applicable and necessary.

1.2.3.2.1 History & Principals

The supercritical point of any pure substance rests at the highest temperature and pressure at which a substance exists in a vapour liquid equilibrium as can be seen in figure 1.21 (Nahar and Sarker, 2012). Above this point, a substance exists as a homogenous supercritical fluid with the density of a liquid but the viscosity of gas (Figure 1.21).

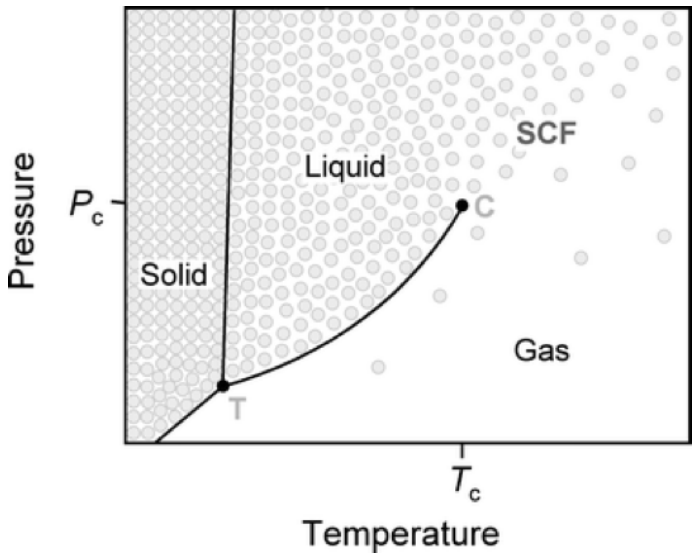


Figure 1.21. Phase diagram showing the critical point (C) at the intersection of the critical temperature (T_C) and critical pressure (P_C). Above this stage, the substance exists in a vapour-liquid equilibrium of various densities depending on the temperature and pressure. SCF stands for supercritical fluid (Nahar and Sarker, 2012, p. 42).

1.2.3.2.2 Parameters

Product solubility can also be optimised by adjusting several parameters: type of solvent (and co-solvent if applicable), temperature, pressure, time of extraction, mode of extraction, flow rate, composition (ratio solvent/co-solvent), etc. The nature of the solvent and co-solvent should be chosen in consideration of the solute to be extracted or eluted. The solvent composition is the most important parameter for supercritical fluid extractions after temperature and pressure (Acevedo-Correa et al., 2018). About 90% of SFE applications employ CO_2 as the primary solvent due to its relatively low critical point at 31.3°C, 7.39 MPa (Pourmortazavi and Hajimirsadeghi, 2007). CO_2 is also preferred as it is non-toxic and environmentally friendly with ozone-depleting emissions, and higher solvating properties than other gases (Bernal et al., 2013). Used CO_2 may also be purified and reused for multiple extractions, unlike liquid solvents.

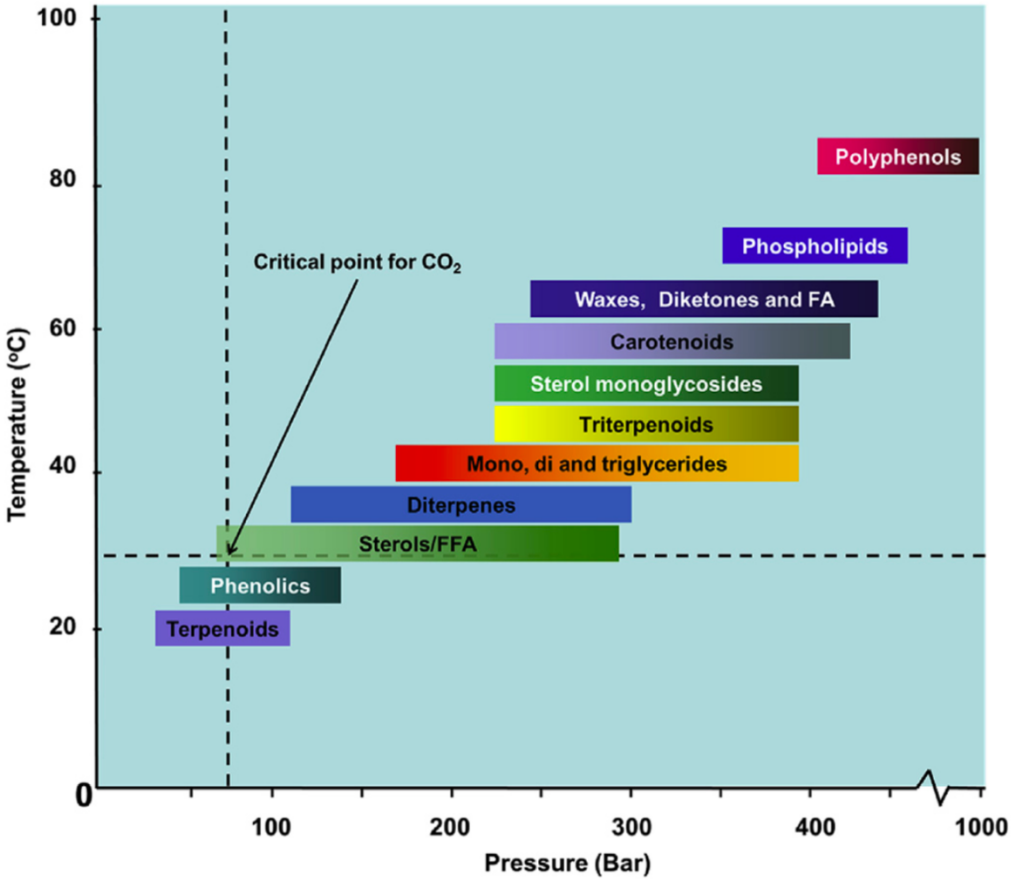


Figure 1.22. Representation of relative pressures and temperatures of CO_2 for the extraction of families of wax and lipid compounds. FFA - free fatty acid, FA - fatty alcohol (Attard et al., 2018b, p. 690)

The density of the mobile phase which consists of fluid in the supercritical state can be highly variable with temperature and pressure – making the diffusivity transitional between a liquid and a gas (Laboureur et al., 2015). The effects of temperature and pressure are the most influential on the solvent properties of a supercritical fluid (Nahar and Sarker, 2012). Adjustments to these key parameters should consider target extracts/elutants and instrument limitations (Figure 1.22). A maximum temperature for organic analytes has been suggested at 60°C using pure CO_2, as higher temperatures may cause degradation (Mićić et al., 2015).

Co-solvents are often added to alter pH or polarity for the extraction of target analytes. Co-solvents (also known as modifiers) are used in their liquid state, and often increase the polarity of non-polar CO_2. For example, hydrocarbon polymers and hydrocarbons of a high molar mass are not readily soluble in carbon dioxide, and often their extraction using supercritical fluid includes a modifier such as methanol (MeOH) (Clifford and Williams, 2000). Their analysis is vital for measuring air quality and pollution effects. The addition of large amounts of co-solvent can significantly alter the critical parameters[9] for the mixture, and analysis is often run in subcritical conditions (Bernal et al., 2013; Laboureur et al., 2015; Pourmortazavi and Hajimirsadeghi, 2007). In studying the extraction efficiency of phenolic compounds, Paula et al. (2014) examined the effects of various combinations of CO_2 and acidified water and ethanol. They discovered that for their specific target analytes, polarity and effects of temperature and pressure were far more important than solvent pH – overall the pure CO_2 was most efficient for extraction of anthocyanins and luteolin with acidified water (0.3% citric acid) having the highest extraction yields overall.

For SFE, the speed at which the solvent flows through the sample in the extraction vessel has a strong influence on the extraction efficiency (Pourmortazavi and Hajimirsadeghi, 2007, p. 12). As it is currently understood, the slower the fluid velocity, the deeper into the sample matrix the solvent will penetrate, and higher flow rates may decrease extraction. This effect has been observed and is presumed to be related to the decrease in solvents linear velocity with the decrease in flow rate, and a decrease in extraction with higher flow rates from an elevated pressure through the extraction cell (Hawthorne, 1993; Özkal et al., 2005).

[9] The critical temperature of CO_2 increases with the increase in polar modifier, causing the analysis to be run in subcritical conditions

For extractions, maximizing the contact between the supercritical solvent and the sample material is fundamental to efficiency. This contact is dependent on multiple parameters, namely, flow rate, but is also heavily affected by the time and mode of extraction (Pourmortazavi and Hajimirsadeghi, 2007). Time simply is the length of the extraction time and mode denoting static with no follow-through or dynamic with follow-through. Cui and Ang (2002) established a procedure for the extraction of St. John's Wort using supercritical carbon dioxide – monitoring temperature, pressure, time and co-solvent. It was found that optimal conditions for extraction included 367 bar and 50°C – and a 10 minute static extraction period followed by a 90 minute dynamic extraction period (Cui and Ang, 2002). The importance of the long dynamic period following the initial static extraction was further highlighted by Pourmortazavi et al. (2004) while testing the influence of mode on extraction efficiency from *Juniperus* communis L. – finding increased extraction times at constant pressures increased the number of heavy compounds with large retention indices in plant oils and waxes. For archaeological pottery – and cultural heritage applications in general – plant waxes and other analytes with higher retention indices[10] are encountered frequently and it is beneficial to run a procedure long enough to extract them (Ávila et al., 2011; Cao and Ito, 2003; Cui and Ang, 2002). Moreover, in cultural heritage, the goal is often to extract as much organic material as possible as these components degrade over time and are often found in small quantities. This results in increased extraction times being more preferential in the field over shorter extractions.

1.2.3.2.3 Commercial and Industrial Applications

In addition to obtaining the properties of both liquid as gas solvents, the use of supercritical fluids for the extraction and isolation of various compounds, and chromatographic separations provide a cheaper, and eco-friendly alternative to liquid solvents use (Nahar and Sarker, 2012). This makes the use of supercritical fluid technologies quite attractive for commercial and industrial applications. Supercritical fluid extraction (SFE) has been prevalent since the 1980s and 1990s, (Donato et al., 2017; Laboureur et al., 2015). This technique continues to gain

[1010] i.e. beeswax, carnauba, candelilla, pine resin and Japan waxes. See Bonaduce and Colombini (2004) and Regert et al. (2005) for more detail.

momentum as a cheaper, quicker, and greener alternative to organic solvent extraction and liquid chromatography for extraction, purification, and recrystallization operations. Some notable applications of lipid extractions are described below, as their basic principles are similar to archaeological organic residue analysis.

The separation and extraction of lipids using supercritical fluids have been highly successful, particularly in the field of food analysis, a field that is parallel to the analysis of ancient food remains. This popularity is due to organic solvents being harmful to humans and the environment, and if their use is avoided then their recovery from the final product is preventable. Lipid analysis often utilized CO_2 with a polar modifier – often methanol, ethanol, isopropanol, and acetonitrile – to increase the solubility of polar analytes and increase the elution strength of the mobile phase (Donato et al., 2017; Laboureur et al., 2015). Acevedo-Correa et al (2018) used supercritical fluid extraction technology to recover oil from grape seeds, a common waste product of wineries rich in vitamin E and unsaturated fatty acids – attractive by-products in culinary, pharmaceutical, cosmetic and medical industries. They found the effects of pressure, temperature, and solvent flow to influence the extraction efficiencies greatly, with a general trend of increasing yields correlating with increasing pressure and decreasing temperature. Similar results were established by Cao and Ito (2003), however, they utilized polar modifiers extensively, finding ethanol and methanol to increase yields. Similar techniques have been adopted for the isolation and characterization of other oils from seeds (Suryawanshi and Mohanty, 2018). Supercritical fluid has also increased in popularity as an alternative to Soxhlet extractions for the isolation and purification of cannabidiol (CBD) from hemp waxes (Attard et al., 2018a). Supercritical fluid has made a large impact on the beer industry, as this technology allows for better quality control of the final product. Lipids affect yeast metabolism during the beer fermentation process positively, however, lipids negatively affect flavour and foam stability (Bravi et al., 2007). Lipid extraction of brewing raw materials including malt and corn girts for fatty acid analysis is a cheaper alternative for quality control that does not require the use of organic solvents (Bravi et al., 2007).

1.2.3.2.4 Cultural Heritage Applications

Applying supercritical fluid to conservation and archaeological sciences has only recently begun. For commercial applications, cost-effectiveness is often a primary factor, whereas

complete extractions are often the primary focus for cultural materials like the ceramics in this study. Using supercritical fluid to extract archaeological material can be inherently difficult, given that target extracts are usually unknown. Commercial applications of supercritical chromatography benefit from knowing the properties of target solutes – thus better-adapting parameters for their efficient extraction. Supercritical CO_2 has been applied to paper from historical records for deacidification – altering paper conservation practices (Français, Perrut, and Brandt, 1997). Rowe et al. (2013) argue supercritical fluid can improve pre-treatment procedures in radiocarbon samples, removing humic acids and exogenous material. Sousa (2007) used CO_2 to clean 18th-century garments from Virgin and Child - A sculpture from Palácio das Necessidades in Lisbon, Portugal. It was found that the addition of water as a co-solvent to supercritical CO_2 assisted the cleaning of the deteriorated silk without physically damaging the garment or losing endogenous material. This method may be further applied to textile conservation in other respects in the future. Näsänen et al. (2013) have applied subcritical fluids to the conservation of actively corroding iron artifacts. While not in the supercritical state, this study provides a better method for preservation based on similar principles – enhancing the desalination solution by altering the solution's viscosity, density, and surface tension improving long term stability. The same properties are altered in supercritical fluid analysis based on the project objective. Supercritical fluid has successfully been applied to waterlogged wood artifacts as a drying agent for conservation, and results have been favourable to traditional freeze-drying techniques (Cretťe et al., 2013).

Devièse et al. (2018) at the Research Laboratory of Archaeology and the History of Art have begun to investigate the use of supercritical fluid for the extraction of organic material from archaeological ceramics. Using a supercritical CO_2 with an ethanol modifier was found to be significantly more efficient than traditional extractions using liquid solvents such as chloroform and methanol ($CHCl_3$/MeOH). This method currently serves as an additional alternative to traditional extraction procedures which is minimally destructive, as ceramic samples which undergo this extraction remain intact, and do not need to be powdered for analysis. This publication is the result of very recent developments and is based on a small number of samples (n= 12). While a promising technique, more methodological developments and comparisons to other procedures are necessary before widespread application. This research will continue this work of testing the applicability of SFE to lipid residues absorbed in pottery as a proof-of-concept and then apply the method to ceramic sherds from

Houtaomuga. Pottery samples from Houtaomuga are unable to be powdered for analysis, making SFE a specifically attractive method to use in this research.

1.2.4 Chemical and Isotopic Analysis of Residues

1.2.4.1 Chromatography

Organic molecules are complex series of carbon and hydrogen chains, and their identification requires analysis that identifies the structure and composition of these chains. Most commonly, the first step of this identification process is separation by chromatography followed by a detection instrument. A primary concern for archaeological sciences is that the sample is of unknown composition and therefore untargeted analyses are required. This is in opposition with most applications in the field of analytical chemistry where the analyte targeted is already known by the operator. Different types of chromatography exist, and they can be defined, for example, by the type of mobile phase: liquid, gas, and supercritical chromatography. Their application to archaeological sciences will be discussed here. As a part of this research, chromatographic methods will be tested for untargeted analyses on archaeological samples.

1.2.4.2 Mass Spectrometry

Mass spectrometry (MS) is the most common detection used in lipid analysis, particularly in archaeological sciences. [11] Other detectors can be coupled with chromatographic instrumentation for lipid determination; however, MS is particularly sensitive and has a high selectivity, making it ideal for quantification and detection of very small molecules. In mass spectrometers, organic compounds are ionised either through electron impact (EI) or reaction processes with a reagent (chemical ionisation – CI). This results in the production and subsequent separation of charged species according to their mass to charge ratio (*m/z*). These charged ions will form predictable fragmentation patterns used for identification. The mass analyser separates gas phase ions according to their mass to charge (*m/z*) ratio when passed through electronic and/or magnetic fields. In recent years, very high sensitivities for qualitative

[11] This work will not cover other detection methods extensively due to the high popularity of MS for lipid analysis in archaeological and heritage sciences.

and quantitative analysis have elevated mass spectrometry resolution between compounds with very similar mass to charge ratios (Losacco et al., 2019).

1.2.4.3 Gas Chromatography and Combustion Isotope Ratio Mass Spectrometry

Gas chromatography (GC) is the most frequently used analytical instrumentation in cultural heritage – most often coupled with MS. GC utilises a gas mobile phase – known as a carrier gas – to transport sample components through packed or capillary columns containing a stationary phase. Most often this includes helium[12] as a mobile phase for its low cost and a stationary phase of bonded silica for lipid separations. This technique uses columns that a narrower and longer than those used in LC and SFC and is capable of separating very complex mixtures of analytes. GC analytes must be volatile – meaning they must have a significant vapour pressure between 30 and 300°C – generally limiting the analysis to analytes under a 500 Da molecular weight. There is a possibility for larger and more complex molecules to be derivatised to increase analyte volatility, however, this process is cumbersome and introduces quantitative errors. For lipids, this means preparing fatty acids into fatty acid methyl esters (FAME) by replacing a hydroxyl group with a methyl group through either acid or base catalysed esterification. This process will fragment larger molecules – such as mono-, di- and triglycerides – into constituent free fatty acids and glycerol of a much lower molecular weight, losing lipid compositional information. In the past, this loss of compositional information has been regarded as unproblematic for residue analysis as it was presumed that large lipids do not survive archaeological time (Evershed, 2008a; 2008b). However, this conclusion was determined when residue analysis was in its infancy, and few ceramic samples from a limited number of burial environments and time periods had been examined using antiquated extraction methods. With increasing detection sensitivity and new lipid extraction methods, it has become apparent that triglycerides do survive under certain conditions (Drieu, 2018). Currently, isotopic analyses to measure bulk $\delta^{13}C$ and $\delta^{15}N$ values, and compound-specific $\delta^{13}C$ values on free fatty acids $C_{16:0}$ and $C_{18:0}$ are performed through GC-C-IRMS to identify a larger variety of products including marine and secondary agricultural products (i.e. milk and cheese) (Copley et al., 2005; Craig et al., 2000; Dunne et al., 2012; Taché and Craig, 2015).

[12] Commonly, GC can also be conducted with a hydrogen mobile phase for coupling with FID. This instrumentation still operates with the same underlying principles.

This analysis requires high abundances of volatile free fatty acids, so the loss of compositional TAG information is often justified by the information that can be learned from a single compound analysis. This compound-specific work is becoming the norm in pottery residue research and is the technique applied most frequently in northeast Asia. To follow best practices and to aid in the comparison of results the technique will also be applied in this research.

1.2.4.4 Liquid Chromatography

While liquid chromatography (LC) is among the most popular chromatography techniques, it is used in cultural heritage applications, but very infrequently for ceramic analysis. The mobile phase in liquid chromatography uses a liquid solvent as the mobile phase to transport the analytes through a column packed with stationary phase material. LC columns and generally shorter and have smaller internal diameters than GC columns. LC does not have the same volatility concerns as GC, instead, any analytes soluble in the mobile phase can be separated by LC., covering a wide polarity range. This gives the operator the ability to choose and alter mobile phases most suitable for the separation of their targeted molecules. LC does not require analyses to be performed at high temperatures, meaning thermal degradation of analytes is avoided. This is a primary concern for the separation of delicate analytes such as fatty acids with double bonds that are susceptible to breakage at high temperatures such as plants (Montañés et al., 2013). Most LC analyses are performed at room temperature (25°C), but column ovens on select instrumentation may allow for temperature alteration. LC also has no real upper molecular weight limit and is suitable for the analysis of large proteins and peptides. Overall, LC is more expansive than GC and uses more organic solvent, raising costs further.

In cultural heritage, LC is also commonly applied to the analysis of dyes and pigments (Claro et al., 2010; Degano, 2018; Degano et al., 2009; Surowiec et al., 2006). Liquid chromatography has very rarely been used to analyse lipids from archaeological ceramics, but three notable exceptions stand out. The first is a complementary use of GC and LC for the characterisation of lipid residue from Basque whalers from the 16th to 17th centuries (Blanco-Zubiaguirre et al., 2018). The motivations behind this article are quite clearly driven by the relatively young age of the samples in question, which will have been subjected to much less degradation than other archaeological samples. The author's goal was to identify triacylglycerols (TAGs) using HPLC-ESI-QToF, and free fatty acids using GC-MS and compare them with modern aquatic oils (including whales) for full lipid profiling and biomarker analysis to identify the contents of the

vessels (Blanco-Zubiaguirre et al., 2018). The second application utilises HPLC for the identification of ergot lipids in Neolithic and protohistoric archaeological vessels from Sardinia and Sicily (Lucejko et al., 2018). This paper outlines an innovative approach to liquid residue analysis which includes the separation and identification of analytes with much higher molecular weights through structural characterisation including DAGs, TAGs, and high molecular weight estolides. Most recently, GC-MS, GC-C-IRMS, and UPLC-HRMS were used in tandem to test the potential usefulness of LC for the analyses of vegetal compounds (Manzano et al., 2019). They found the application promising for the identification of fatty acid amides and polyphenols attributed to various plant and fungus species additionally confirming their hypothesis that the pottery from the funerary site was not made exclusively for burial. *Santa María* style ceramic vessels from El Colorado (Catamarca, Argentina) have also been analysed using HPLC-ESI-Q-TOF to screen for complex organic mixtures including residual underivatized acylglycerides (Lantos et al., 2020).

1.2.5 Radiocarbon Dating and Pottery

Chronology building and pottery have a close relationship. Early pottery from northeast Asia has been radiocarbon dated and frequently used for chronology building, thus warranting its discussion here (Kunikita et al., 2017; Kuzmin, 2006; Kuzmin, 2017). Being ubiquitous in the archaeological record, highly breakable, often discarded, and chronologically sensitive, pottery has been used in establishing relative chronological frameworks since the beginnings of archaeology as a discipline. This can be done through typology building, or through chronometric techniques whether absolute or relative. With the advent of AMS dating, the drawbacks of dating materials associated with human activities rather than materials that themselves are evidence of human activities have become apparent, particularly regarding pottery (Bonsall et al., 2002).

Ceramic styles are often the basis of relative chronology building through typology and seriation. A typology is “a special kind of systematic classification that divided a group of phenomena into discrete types according to their common characteristics” (Hein, 2016, p. 7). While subjective to the researcher, typologies offer a vital source of chronological information. One major downfall for this technique is the difficulty to cross-date associated sites as is often unknown or unclear when ceramic forms change or go out of style, and when styles reach peak popularity, as this process can be inconsistent throughout a region (Rice, 1987). Absolute

dating of ceramics would be vital in the restructuring of typologies in many regions and provide further validation of pottery as a chronological index. Little research on the relationship between typologies and pottery use through direct analysis has been undertaken.

Naturally, ceramic can be dated directly through thermoluminescence (Aitken et al., 1964). This technique has sources of uncertainty and logistical concerns. Luminescence dating gives poorer precision, leaving many laboratories to constrict archaeological applications to dates beyond the limit of radiocarbon dating (around 55,000 BP) (Bonsall et al., 2002). Thermoluminescence as a means to absolutely date ancient pottery is advantageous to relative dating of associated materials (i.e. charcoal and bone) as relative dating is limited by assuming artefact association (Bonsall et al., 2002). Likewise, radiocarbon is cheaper and more straightforward than luminescence measurements. Luminescence dating requires the determination of the environmental dose rate through measurements of the store's radiation dose experienced by the grains in the pottery sample since firing at each sample location (Rhodes, 2011, p. 462). Financial restrictions and time limits of archaeological work often limit the application of this technique to pottery. Mainly, all pottery samples fall within the limit of radiocarbon dating, and this technique is far preferred for cost and data comparability reasons. The likelihood of the application of thermoluminescence dating to a large number of pottery samples in any time period and region is slim.

Gabasio et al. (1986) explored the origins of carbon in archaeological pottery to understand radiocarbon reliability with this material type. With the recent development of AMS, dating small amounts of carbon from ceramics became potentially feasible, and this was the first systematic study of pottery as a radiocarbon sample material. Until Gabasio et al. (1986) only isolated incidents of sites with little or no other datable material were explored. Many of these dates were deemed unreliable and often dated slightly later than expected. Gabasio et al. (1996; 712) identified five potential sources of carbon in ceramics, which have been elaborated upon by Bonsall et al. (2002):

1) *Carbon presented in clay* – this carbon is assumed to derive from naturally occurring organic matter in clay matrixes. Clay deposits have the potential to be of a significant geological age, while those exposed to the surface have the potential for contamination through organic material contemporaneous to the creation of the pot.
2) *Carbon from temper* – Regarding straw, chaff, and other plant-based organic tempers, this carbon source has potential for absolute dating. In many scenarios, this material

will burn off during the firing process, but in some exceptional cases (usually from low open-pit firings) this material may preserve. Temper may be a significant source of carbon contaminants when it includes marine or freshwater shells due to marine reservoir effects and hard water effects, respectively (Bronk Ramsey, 2008).

3) *Carbon from fuel in the kiln* – fuel used in pottery firings has been suggested to be cause for a source of old wood in radiocarbon dating. This is due to the assumption that incomplete combustion of wood fuel due to reduced oxygen conditions in a kiln would give rise to smoke production, which may cause carbon to be absorbed by the pot (Bonsall et al., 2002). This likely only causes a negative effect when wood that is not recently felled is used as fuel, otherwise, this will be contemporaneous to the production of the pot.
4) *Carbon derived from the use of the pottery* – this includes adhering food residues and soot, or smoke absorbed during the firing process, or during cooking. However, Bonsall et al. (2002) point out that adhering food residues are susceptible to geochemical contamination, and the food source of the residue is unknown. If this is derived from fish or shellfish, then radiocarbon dates are likely to be much older than the actual date of the use of the sherd (Bonsall et al., 2002).
5) *Secondary carbon contamination* – regarding carbon contaminants from the surrounding depositional environment, including humic substances, lipids, rootlets, etc.

Heron, Evershed, and Goad (1991) found the effects from the migration of contaminants in the soil to ceramic residues to be minimal, therefore secondary carbon contamination is likely not a major factor in carbon contribution. Since the 1990s, little research had been done on carbon contamination in archaeological pottery until Reber et al. (2018). Studying lipid residues from low fired experimental pottery, they discovered that diterpenoid lipids were being introduced to the clay as condensate from pine (Pinus *spp.*) wood used as fuel (Reber et al., 2018, p. 1). Naturally, the introduction of this organic component would induce additional carbon which is not endogenous to the residue or organic temper being dated. More importantly, this research determined pottery firing removes all organic material naturally occurring in clay (i.e. alkyl lipids) possibly resetting the amount of carbon (Reber et al., 2018).

Radiocarbon dating has been successful at dating several contexts and includes carbonized deposits from within/adhering to the pottery, and organic temper in the matrix.[13] Lipid residues absorbed into the pottery matrix are particularly attractive for radiocarbon dating, as they have fast metabolic turnover rates ensuring ^{14}C ages close to the death date of the organism (Berstan et al., 2015, p. 703). One notable application is the use of this technique on ceramic samples from the sites Houtaomuga and Shaungta in Northeast China by the University of Tokyo (Kunikita et al., 2017). This research has shed light on the very old ages of these samples, however, more dating across the region would be beneficial. As can be seen in figure 1.9, large discrepancies remain between radiocarbon ages from samples coming from the same vessel, however, these discrepancies have not been explained or further explored.

Recent work has applied preparative GC techniques to pottery residues for direct radiocarbon dating of pottery. Compound specific preparative GC was used to isolate palmitic ($C_{16:0}$) and stearic ($C_{18:0}$) fatty acids after they have been extracted from the ceramic matrix using an acidified methanol protocol, and these compounds were then dated using AMS (Casanova et al., 2020). At present, this technique has some limitations, including the need for high amounts of lipid preservation. Moreover, the palmitic and stearic acids could be deriving from multiple uses of the pot, leading to unspecific dates. Despite these concerns, this type of technique is particularly useful for direct dating of pottery use and this research expansion of this type of research in the future would be highly beneficial to understanding the archaeological record.

1.2.6 Organic Residue and Starch Grain Analysis in China

Analysis of organic remains using other methods has occurred in China in the last 30 years. This research has primarily consisted of applying analytical chemistry techniques to identify paint binders, waxes, and lacquers (i.e. Le Hô et al., 2012; Ling et al., 2007; Wei et al., 2015). These studies are often focused on the later periods than the work presented in this dissertation, however, I have chosen to present an overview of this work here to put my thesis results into context with the current state of research. For a detailed discussion of organic analysis in China please see Yang (2021).

[13] This is only possible in analysis of low fired pottery, or poorly open fired pottery, as much of the organic temper will burn away during the firing process.

Most research on food preparation and consumption in Chinese archaeology has focused on micro and macro botanical remains from pottery and grinding stones. Two research groups have used starch grain analysis of Yangshao period (5,000 – 3,000 BC) ceramic amphorae from the sites of Banpo, Jiangzhai, and Mijiaya in Shaanxi province to identify various cereals used for brewing (Liu et al., 2020; Wang et al., 2017). Grinding stones ranging from the late Palaeolithic to early Neolithic have also been subjected to starch grain analysis, providing an understanding for highly varied ancient subsistence economies in North China (Liu et al., 2014, 2013). Liu et. al (2013) have shown that humans began cultivating various plants, including *Triticeae* and *Paniceae* grasses, and various tubers in the Shizitan Locality in North China as early as 23,000 years ago. This means that *Paniceae* grasses, which now produces domesticated millet, were exploited about 12,000 years before their domestication. The earliest domestic millet in northeast China (ca. 8,000–7,500 cal. BP) has been identified in grinding stones using these methods, and its importance along with various other plants have been analysed at the site of Xinglonggou in Inner-Mongolia (Liu et al., 2015). Liu et al. (2015) also identified the earliest evidence of jobs tears in northern China, making it the second domesticate to have become a part of human diets in the region. Additionally, in North China, starch analysis has been undertaken on internal deposits from pottery dating to 11,000 BP at the site of Zhuannian (转年) site, 100 km north of Beijing (Yang et al., 2014). Yang et al. (2014) identified millets, acorns, and grass evidence, implying a primarily plant-focused vessel use. This study however looks at four samples from this site, and larger sample size would draw more confident conclusions. Lately, similar analysis in southern China has identified evidence of rice, jobs tears, acorn, and bean from cooking vessels at the Kuahuqiao site in Zhejiang province (Yang and Jiang, 2010). While highly informative, macro and micro–botanical analysis does not always wholly represent vessel use. This type of analysis is limited to only identifying plant-derived resources and does not account for animal products or resinous products which may have been present in such vessels. This previous research on micro and macro botanical remains in China are specifically relevant to this thesis because it sheds light on plant in hunter-gatherer diets before extensive agricultural uses. Similarly, Houtaomuga has grinding stones which are used by hunter-gatherers, who we know exploited a variety of plant resources (Tang et al., 2020). Residue analysis can help us further understand this plant's use and how it was prepared. While the extent is not yet fully known, hunter-gatherers developed relationships with their environments and the plants that grow in them far before we see evidence for formal agriculture in the archaeological record and more research on how and when this exploitation

occurred is necessary to understand the long-term trajectory of such plants. Further microbotanical analysis on the Song-Nen plain would be highly beneficial for understanding past subsistence patterns.

The majority of organic analysis research on archaeological remains in China was conducted on material from Northwest China. Some portions of this region are exceedingly dry and organic remains are subject to favourable preservation conditions. In Qinghai, starch, proteins, and lipids have been analysed from intact noodles found in the bottom of a ceramic vessel at the site of Lajia (Lü et al., 2014). Lü et al. (2014), identified that the noodles were made out of millet. Similar exceptional preservation is found in Xinjiang, where proteomics and starch grain analysis has been utilized to investigate and identify adhesives, preserved noodles and cakes, along with identifying kefir dairy and millet from the early Bronze Age (Gong et al., 2011; Rao et al., 2015; Yang et al., 2014). Additionally, dairy products have been identified on grass woven baskets from Gumugou Cemetery in Xinjiang, further supporting notions about secondary product use and herding practices in this area during the Bronze Age (Xie et al., 2016). Unfortunately for archaeologists, preservation of organic remains is not as spectacular in northeast China as it is in parts of northwest China and similar remains do not survive in the research area of focus in this thesis. This thesis will shed light on organic preservation of microscopic remains in the area however and the degree of preservation for lipids at Houtaomuga will be discussed. This may hopefully aid future organic residue work in the area in sample selection and methodology development.

1.2.7 Conclusion

Residue analysis primarily concerning lipids has expanded and is now commonly applied to answer archaeological questions. Following methods used in previous research and further developing those in their infancy, this research aims to apply residue analysis non-destructively to pottery samples. This will be done using supercritical fluid extraction to remove the lipid residues from the ceramic body. Before applying this method to archaeological samples from northeast China, the methods for extraction and separation will be tested for applicability. This method will then be applied to archaeological pottery samples whose former contents have already been identified using commonly applied techniques and the results will be compared. This testing will be described in detail in the following chapters. When a protocol for

extraction, separation, and characterization of lipid residues has been established, it will be applied to samples from northeast China. For comparison with data in adjacent regions in Asia, compound specific GC-C-IRMS will be undertaken to further characterize pottery contents, and results will be compared.

Chapter 2 Methods and Materials

2.1 Analytical Strategy

I did not get permission for destructive analysis on archaeological samples from Houtaomuga. Therefore, this dissertation uses non-destructive supercritical fluid instead of traditional liquid extraction methods to extract lipid residue which has been absorbed into the ceramic sherd. Supercritical fluid extraction (SFE) of lipids absorbed in archaeological pottery is still in its infancy and remains to be examined using a wide range of lipid classes or on various archaeological assemblages. In this thesis, I examined the applicability of SFE to various lipid classes commonly found in archaeological contexts and on archaeological ceramics of various ages and types (step 1 of the analytical strategy chart in figure 2.1). This method optimization also included testing of currently established techniques to determine the best approach for my specific research questions. This lipid extraction optimisation was then followed by optimisation and testing of methods for molecular analysis (step 2 of the analytical strategy chat in figure 2.1). The SFE method developed by Devièse et al. (2018) was optimised using modern materials and then applied to archaeological material containing various lipids including free fatty acids, diterpenoids, triterpenoids, waxes, etc. to establish the extraction efficiency of the supercritical fluid procedure. These archaeological samples were chosen as their organic contents were known through previous characterisation using established methods. Both steps 1 and 2 (figure 2.1) use both modern reference materials and lipid reference standards as well as archaeological materials. Unlike lipid extraction and chromatography in food chemistry for example where many components are known, the complex mixtures of lipids from archaeological contexts are unknown. Additionally, archaeological samples come from various environments and are often buried underground for long periods, making them subject to contamination and degradation at varying degrees. Accounting for this requires rigorous method validation and testing on various lipid classes.

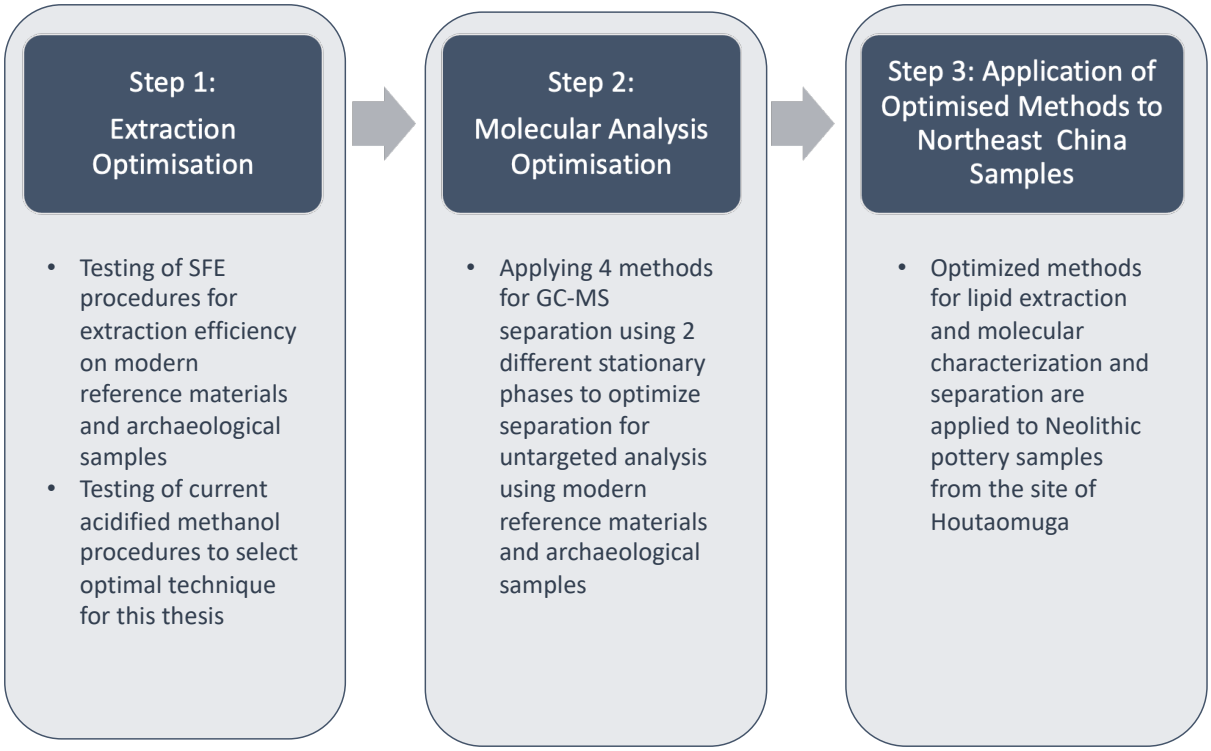


Figure 2.1. The analytical strategy used in this thesis

Supercritical fluid extracts from these tests and modern reference materials were then characterised using two gas chromatography-mass spectrometry (GC-MS) methods using two different stationary phases and an on-column injection and results interpreted (figure 2.1 step 2; figure 2.2). This begins with fully intact sherds undergoing the optimised supercritical fluid extraction (SFE) procedure which was first tested on a series of reference material and archaeological samples to confirm the efficacy and assess applicability to such material due to the novelty of the method. An aliquot of these samples was then derivatized and analysed using gas chromatography-mass spectrometry (GC-MS) on both a cool-on-column injection, which is based on methods from Dunne et al. (2012) and optimised for my research in method development using two different stationary phases, and a split/splitless injection as is standard practice in the field (Colombini et al., 2003; Ribechini et al., 2009)(figure 2.2). The goal of step 2 (figure 2.1) was to evaluate the proper stationary phase and chromatographic conditions for rapid, untargeted analysis of various lipid classes, and build a sufficient workflow for analysing samples from northeast China. The optimised methods determined in steps 1 and 2 (figure 2.1) will then be applied to archaeological pottery from the site of Houtaomuga in step 3 to understand initial pottery use in the Neolithic in this region and how that use changes or stays the same over time. This workflow (figure 2.1) will simultaneously assess the widespread applicability of non-destructive SFE to archaeological pottery and shed light on early pottery

use in northeast China, hopefully teaching us more about the long processes of domestication and the interaction between hunter-gatherer-fishers and their environment.

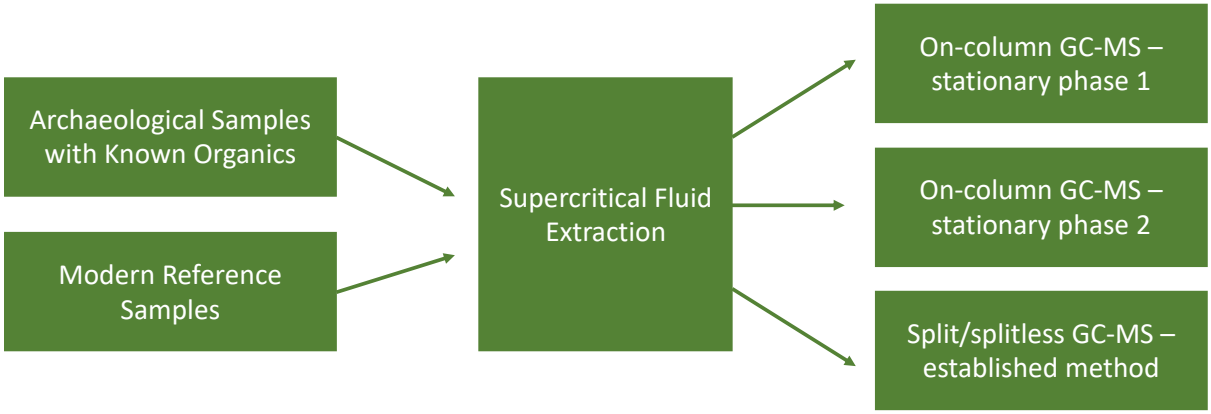


Figure 2.2. Workflow for method optimisation and assessment

This method optimisation (step 1 and 2, figure 2.1) using reference material and archaeological material was specifically necessary before the application of the method to samples from northeast China, as these samples are exceedingly old, rare, and their contents unknown. While a majority of archaeological organic analysis is untargeted, often previous research or ceramic type can give archaeological chemists a clue about the organics preserved, i.e., visible resins or vessel shapes can suggest former use and thus potential former contents. However, for pottery from northeast China, ash deposits or soot on the outside of vessels, possibly coming from use over an open flame, is the only indication of use. This means the procedure and workflow applied to this assemblage must be able to identify a wide range of compounds. Samples with high enough concentrations of palmitic and stearic acid were then subject to GC-C-IRMS analysis to further characterise these extracts (figure 2.1 step 3).

2.2 Methods

2.2.1 Sample Preparation

Before analysis, archaeological samples were weighed and photographed. Subsequently, the outer surface (1mm) of intact ceramic samples was removed using a diamond-tipped Dremel tool before absorbed residue extraction to remove contaminants from burial environment or

excavation handling (Heron and Evershed, 1993). This ceramic powder was retained for any potential future analysis. Details of all laboratory blanks used in analysis are detailed in appendix IX.

2.2.2 Extraction Methods

2.2.2.1 Liquid Extraction

500µl of chloroform/methanol (2:1 v/v) was added to the sample in a clean glass vial to desorb and/or dissolve the lipid residue based on methods published by Heron et al. (1991). Samples were then sonicated at 25°C for 30 minutes. The solution was transferred to a 2mL clean glass vial and evaporated under a gentle stream of nitrogen. This process was repeated an additional two times for three total liquid solvent extraction washes. 50µl of hexane was then re-added to the dry vial to dissolve the extracts, along with 20µl N, O- Bis(trimethylsilyl)trifluoroacetamide (BSTFA) containing 1% trimethylchlorosilane, used for trimethylsilylation derivatization and 5µl tridecanoic acid as internal standard. The sample was then incubated for 30 minutes at 70°C before the addition of 10µl of internal standard 2 – hexadecane – and transferred to a final vial containing a glass micro insert for GC-MS analysis.

2.2.2.2 Acidified Methanol Extraction

Acidified methanol extractions were performed on recovered solvent from supercritical fluid experiments after they were blown down under a gentle stream of nitrogen to keep results consistent with current research. This method is adapted from Craig et al. (2013), Correa-Ascencio and Evershed (2014), and Papakosta et al. (2015). 1.5 mL glass vials (I) containing SFE fractions, 10µl tridecanoic acid ($C_{13:0}$) as internal standard (1.87×10^{-6} mol/µl), and 5mL H_2SO_4-MeOH (2% v/v). Samples were whirlmixed and incubated at 70°C for 4 hours. The pH was tested at the beginning of each extraction, as well as after 30 minutes of incubation, and post-incubation. The solution was then centrifuged at 2500 r.p.m for 10 minutes and the clear supernatant was transferred to a clean glass vial (II) along with 2mL of MilliQ H_2O. 3mL of hexane was then added to vial I to recover any remaining material and then centrifuged for an additional 10 minutes at 2500 r.p.m. The hexane phase was then extracted and added to vial II. The solution was whirlmixed and the hexane phase removed and transferred to a third (III) glass vial. This process was repeated an additional two times, with 3mL being added to vial II

and whirlmixed to extract any remaining material, and the hexane phase being transferred to vial III. This hexane extract was evaporated to dryness under a stream of nitrogen. 100µl of hexane was then re-added to the dry vial (III) to dissolve the extracts, along with 40µl N, O-Bis(trimethylsilyl)trifluoroacetamide (BSTFA) containing 1% trimethylchlorosilane, used for trimethylsilylation derivatization and 10µl tridecanoic acid as internal standard. Vial III was then incubated for 30 minutes at 70°C before the addition of 20µl of internal standard 2 – hexadecane – and transferred to a final vial with a glass micro insert for GC-MS analysis.

2.2.2.3 Supercritical Fluid Extraction

Supercritical fluid extraction was performed using a Semi-Prep SFC-SP-2068 instrument from Jasco©. The system had been modified to allow for both supercritical fluid chromatography and supercritical fluid extraction to be performed, allowing for the auto-sampler to be bypassed and having the solvent flow through an extraction vessel in the oven rather than a chromatographic column. Each sample was collected in one fraction. During method development and proof of concept testing, the collection vials were weighed before each extraction, and again after each extraction after blowing down excess solvent under a stream of nitrogen. The idea in mind for this experimental procedure is an attempt to measure the amount of material being extracted and collected to compare different procedures.

For method development, 10µL of olive oil was added to modern ceramic pellets. The modern ceramic was subjected to supercritical fluid extractions using different parameters. These include both dynamic mode as defined in Devièse et al. (2018) and a static-dynamic extraction. The dynamic mode parameters included a co-solvent (EtOH) to solvent (CO_2) ratio of 15%, a consistent flow rate of 2 mL/min and a consistent temperature of 50°C for a total extraction time of 90 minutes. The parameters for the static-dynamic extraction were chosen in an attempt to further optimise the extraction. Primarily, a 10-minute static period (1 mL/min flow rate for a ramp in pressure) was added to an 80-minute extraction (2mL/min) for a total extraction time of 90 minutes. This time frame was selected because it has been demonstrated that a static period longer than 10-minutes does not increase the extraction efficiency (Cui and Ang, 2002). The full extraction time was not altered, as a primary concern for archaeological sciences is a

complete organic extraction rather than a quick extraction[1]. The high efficiency of a long dynamic extraction as employed here has been demonstrated notably by Pourmortazavi, Baghaee, and Mirhosseini (2004) on the composition of essential oil from *Juniperus communis* L. – finding the increasing of extraction time at a constant pressure improves the extraction of heavy compounds with larger retention indices in plant oils. The pressure of the static-dynamic extraction is set to build for 10 minutes from to 30 MPa to increase contact between the sample and the solvent/co-solvent. The pressure of the run then remains at 30 MPa from minute 10 to minute 85, after which it then ramps down linearly to 15MPa for the end of the extraction. The temperature of the run is kept at a constant 50°C – and temperature and pressure follow similar properties outlined by Deviese et al. (2018) and Attard et al. (2018) for the extraction of hemp waxes. The solvent composition was altered from 16% ethanol to 20% methanol due to the vast similarities in both solvents, and to further compare results with other methanol liquid extractions. The co-solvent proportion was increased to 20% as Devièse et al (2018) found there to be little difference between 15-20% co-solvent, but a review of the literature demonstrates the benefit of increasing the co-solvent for the extraction of largely polar lipids using the very a-polar CO_2 (Johner et al., 2018). The rate of co-solvent was also set to decrease from 20%-0% at a linear rate from minute 75 to 90, to help extract any potential a-polar material. In addition to altering parameters of extraction, a series of experiments were undertaken to freeze dry a subset of ceramic samples for 24 hours and compare results to samples that did not undergo this pretreatment. This is due to the possibility of excess water allowing for CO_2 to partition into two phases.

For archaeological material, parameters included a co-solvent (MeOH) to solvent (CO_2) ratio of 15%, a consistent flow rate of 2 mL/min and a consistent temperature of 50°C and a pressure of 30 MPa for a total extraction time of 90 minutes. At 75 minutes, the co-solvent ratio decreases linearly from 15% to 0% at 90 minutes allowing the majority of the co-solvent to travel through the system. Samples were subject to this dynamic extraction with the lower co-solvent ratio after experimental results showed little to no effect on extraction efficiency (results can be found in section 3.1.2). Future work may include testing to reduce the extraction time, however, archaeological objectives continue to prioritize full lipid extraction over speed, and this will remain consistent throughout this study. Liquid SF extracts were collected, and

[1] I choose to mention this, as it is a high priority for the majority of the literature on supercritical fluid extractions, as these publications are written with an industrial focus rather than a heritage one.

the solvent was evaporated using a Genevac© centrifugal evaporator. Lipid extracts were rehydrated in 1mL $CHCl_3$/MeOH (2:1 v/v), agitated, and moved to a clean 2mL glass vial. This process makes solvent quantities consistent for consistent lipid concentration comparison. For GC-MS analysis, 200 µL aliquots of extract were evaporated to dryness using N_2 gas. For derivatisiation, extracts were rehydrated in 100µl hexane with 20µl N, O-Bis(trimethylsilyl)trifluoroacetamide (BSTFA) containing 1% trimethylchlorosilane, used for trimethylsilylation derivatization and 10µl tridecanoic acid as internal standard. The samples were then incubated for 30 minutes at 70°C before the addition of 20µl of internal standard 2 – hexadecane – and transferred to a final vial containing a glass micro insert for GC-MS analysis.

2.2.3 Analysis

2.2.3.1 GC-MS

The instrument used for analysis was an Agilent© 8890 gas chromatograph equipped with both a cool-on-column and split/splitless injector. The mass spectrometer used was an Agilent© 5977B quadrupole, operated in electron ionization mode (70 eV). For the on-column injection, either a Restek© Rtxi-65TG column (30m x 0.25mm ID x 0.1 film thickness, crossbond diphenyl dimethyl polysiloxane stationary phase) or an Agilent© DB-5ms column (30m x 0.25mm ID x 0.25 film thickness, nonpolar phenyl arylene polymer, virtually equivalent to (5%-phenyl)-methylpolysiloxane) were used. The injection volume was set at 1µL with an inlet pressure of 19.55 psi and a flow rate of 1.2mL/min. The mass spectrometer operated with a scan range of m/z 50-950. The source temperature was set at 350°C and the quad at 180°C. The initial GC oven temperature was 35°C and held for 2 minutes followed by a ramp in pressure at a rate of 10°C/min to 350°C and held for 30 minutes. For method testing, these conditions were altered to 20°C/min to 350°C, 10°C/min to 325°C, and 20°C/min to 325°C. All further conditions for cool-on-column method testing remained consistent.

For split/splitless injections, an Agilent© HP-5 column (30m x 0.25mm ID x 0.25 film thickness, 5%-phenyl-methylpolysiloxane stationary phase), was utilised with an autosampler. The mass spectrometer used was an Agilent© 5975 quadrupole, operated in electron ionization mode (70 eV) with a scan range of m/z 50-650. The initial GC oven temperature was set at

80°C and held for 2 minutes followed by a ramp in pressure at a rate of 10°C/min to 200°C and held for 4 minutes and increased again at 6 °C/min until reaching 280°C and held for 20 minutes. The oven temperature then continues to increase from 280°C to 300°C at a rate of 6°C/min and held for the remaining 35 minutes. The injection volume was set at 1µL and run in splitless mode for archaeological samples, with an inlet pressure of 11.681 psi and a flow rate of 1.2mL/min. The same mass spectrometer operated with a scan range of m/z 50-650. The source temperature was set at 230°C and the quad at 150°C. Compound concentrations were quantified using methods described in appendix IX.

2.2.3.2 GC-C-IRMS

All GC-C-IRMS was conducted at the Archaeological Research Laboratory at the University of Stockholm, Sweden All samples were dissolved in 50 mL toluene. The GC-C-IRMS analyses were conducted using a Trace GC equipped with tandem capillary columns, a DB-1ms (30 m × 0.25 mm × 0.25 µm) followed by a DB-5ms (30 m × 0.25 mm × 0.25 µm) and coupled with a Thermo© Delta V mass spectrometer. Samples were injected through a programmable temperature vaporization (PTV) injector operated in solvent split mode. The injection volume was 2 µl, the injection pressure was 70 kPa, and the initial temperature was 40 °C. The splitless time was set to 1.0 minutes, the solvent vent temperature was 100 °C, and the vent flow was 100 mL per minute. Evaporation pressure was set to 140 kPa, evaporation temperature was 40 °C, evaporation rate was 10 °C per second, and evaporation time was 0.16 minutes. Transfer pressure was 210 kPa, transfer temperature was 300 °C, and the transfer rate was 12 °C per second. The cleaning temperature was 320 °C, the cleaning rate was 14.5 °C per second, cleaning time was 20 minutes, with a cleaning flow of 100 mL per minute. The GC oven was temperature programmed with an initial isothermal of 2 minutes at 120 °C, followed by an increase of the temperature with 20 °C per minute to 200 °C, followed by a temperature ramp of 5 °C per minute to 315 °C, and held at this temperature for 7 minutes. The target fatty acid methyl esters were converted to carbon dioxide (CO_2) in the IsoLink II reactor system. Pulses of reference CO_2 gas were injected through a ConFlo IV unit. The CO_2 pulses were calibrated against a certified fatty acid methyl ester standard of known $\delta^{13}C$, and this was done individually for both C16:0 and C18:0. In-house standards of known $\delta^{13}C$ were analysed between samples to monitor instrument precision and accuracy. Carbon isotopic values were expressed in per mil (‰) relative to the Pee Dee Belemnite (PDB) standard.

2.3 Materials

2.3.1 Solvents, Reagents, and Standards

Solvents used for this analysis were of an analytical grade, including tridecanoic acid ($C_{13:0}$), methanol, sulfuric acid, hexane, and hexadecane. These were all purchased through Sigma Aldrich (USA), except for hexane, which was purchased from VWR international. BSTFA containing 1% trimethylchlorosilane, used for trimethylsilylation derivatization was purchased in 1 mL ampoules.

2.3.2 Reference Materials for Method Validation

2.3.2.1 Modern Experimental Lipids

Alkane standards (C_8-C_{20}) free fatty acids (saturated and unsaturated, $C_{8:0}$-$C_{24:0}$), and triglycerides ($TG_{8:0/8:0/8:0}$, $TG_{10:0/10:0/10:0}$, $TG_{12:0/12:0/12:0}$, $TG_{14:0/14:0/14:0}$, $TG_{16:0/16:0/16:0}$) were all purchased from Millipore-Sigma©. Beeswax was purchased from L. Cornelissen & Son.

2.3.2.2 Modern Clays Pellets

Ceramic pellets for experimental analysis were prepared using Kaolinite clay. Clay was purchased from Fischer instruments and was 'general-purpose grade'. Ceramic pellets were fired at 825°C with a plateau at 493°C for dihydroxylation to prevent cracking. Olive oil used to impregnate pellets for experimental extraction was chosen for this study as it is commonly chosen in similar published literature testing certain procedures (Evershed, 2008). This is likely since many studies have been carried out investigating the adulteration of olive oil with much cheaper vegetable oils in Europe and the Americas; supplying an abundance of research for data comparison making pure olive oil easily identified (e.g. Christopoulou et al., 2004; Fasciotti and Pereira Netto, 2010). Additionally, olive oil consists of various saturated and unsaturated fatty acids, covering a wider range of lipid types than animal fats for example. It is further preferred for this type of analysis because it is easily accessible and relatively cheap compared to other pure oils.10μL olive oil was added to each modern ceramic pellet and artificially aged at a temperature of 600°C for 6 hours before being subject to supercritical fluid extraction techniques with varying parameters. After extraction, an aliquot of the SFE extract

was transferred to 2mL glass vials and blown down under a gentle stream of nitrogen to dryness. 100µl of hexane is then re-added to the dry vial to dissolve the extracts, along with 40µl N, O- Bis(trimethylsilyl)trifluoroacetamide (BSTFA) containing 1% trimethylchlorosilane, used for trimethylsilylation derivatization and 10µl tridecanoic acid as internal standard. Vial III is then incubated for 30 minutes at 70°C before the addition of 20µl of internal standard 2 – hexadecane – and transferred to a final vial with a glass micro insert for GC-MS analysis.

2.3.3 Archaeological Materials of Known Organic Composition Used for Method Validation

As mentioned in section 2.1, archaeological samples are often more complex mixtures of organics and have undergone significant degradation. Additionally, these samples are often buried in the ground for long periods and consist of heterogeneous mixtures and contaminants whose effects must be properly mitigated. Therefore, any methods tested for archaeological samples should be examined using archaeological material which also holds such difficulties in tandem with modern reference materials. To achieve this, archaeological material which had been previously examined using other methods of analysis was selected for method validation. This technique of using archaeological material for testing in this way is novel and has not been applied previously. The goal of this innovative technique is to better replicate samples for testing, as opposed to utilising modern materials which have been artificially aged.

These samples include 110 pottery sherds from the Bobarita Road midden site in Betty's Bay, Western Cape South Africa. The site in question is currently unpublished. Previous residue analysis has determined these sherds contain a mixture of monocarboxylic acids, fatty acyl wax esters and n-alkanes. The archaeological test samples also include 8 pottery samples from the Roman Villa site at Gerace, Sicily (Wilson et al., 2021). These samples are Roman amphorae with visible residue adhering to the inner wall of the sherd and date to the second half of the 5th century AD. Previous analysis has shown this resin comes from trees in the *Pinaceae* family and mainly contain abietic acids along with other polycyclic aromatic hydrocarbons and free fatty acids.

2.3.4 Houtaomuga Pottery Samples

A total of 153 pottery samples were selected from the site of Houtaomuga for non-destructive absorbed lipid residue analysis. Information regarding specific samples and information on each sample can be found in appendix I. The number of samples for each occupational phase, their respective radiocarbon dates, pottery firing temperature and temper material are detailed in table 2.1, as each of these characteristics could influence results. The archaeological context of these samples has been described in detail in Chapter 2. Samples were selected across all four phases as available. Phases 2 and 3 did not contain as many pottery samples which could be selected resulting in a lower number of samples from these phases than Phase 1 and Phase 4. Samples that were unwashed and the correct size to fit in the supercritical extraction vessels were preferentially selected over larger and cleaner samples. All samples selected are body sherds ranging from 418mg to 3219mg. Pottery samples coming from the same feature were selected based on colour and decoration in an attempt to avoid selecting samples that come from the same vessel. Samples which visible dark sooting or evidence of use were selected over samples that appeared free of residue or unused.

Table 2.1. Samples from northeast China that were analysed in this dissertation. The estimated firing temperatures listed are the results of an experimental study about Houtaomuga pottery production by Liu et al. (2016).

Site	Phase	Date	Number of Samples	Estimated Firing Temp	Temper
Houtaomuga	Phase 1	13,000 to 11,000 BP	65	<600ºC	Sand/fibre
Houtaomuga	Phase 2	8,000–7,000 BP	21	<730ºC	Shell
Houtaomuga	Phase 3	6,800–5,500 BP	20	<770ºC	Shell
Houtaomuga	Phase 4	5,500 to 4,900 BP	47	850ºC	Shell

2.3.4.1 Houtaomuga Phase 1 Pottery

The earliest known pottery in northeast China comes from the first period of Houtaomuga I and dates to 12,900-11,100 cal BP (Kunikita et al., 2017; Wang and Sebillaud, 2019). Dates obtained from carbonized crust adhering to the surface of vessels at Houtaomuga are 1,000-1,500 years older than dates obtained from wood charcoal (Kunikita et al., 2017). These dates are similar to pottery dates from Amur River region sites in modern-day Russia (Kuzmin, 2006). Phase 1 pottery is either grey-brown or yellow-brown with uneven surface colour and blackened cores (Figure 2.3). This pottery type is thick (0.8 to 1.5 cm), contains organic grass

temper, and is made by coiling along with a thin slip of clay added to the inner and outer surface of the vessel (Liu et al., 2016). The majority of vessel bottoms are flat or slightly concave (Wang and Sebillaud, 2019). As indicated by experimental testing, pottery during this phase was likely fired in the open air at temperatures below 600ºC, resulting in an extremely brittle ceramic body (Liu et al., 2016). Analysis of clay composition indicates that the raw material is a high iron fusible clay, which is available locally but still possibly suggests this material was selectively chosen over other clay types in the region (Liu et al., 2016). This could imply that the clay is being selected with future pottery use in mind, or because of another benefit, this clay type gives during the production process. Further research is needed to better understand the motivation behind this selectivity and here residue analysis can provide further insights.

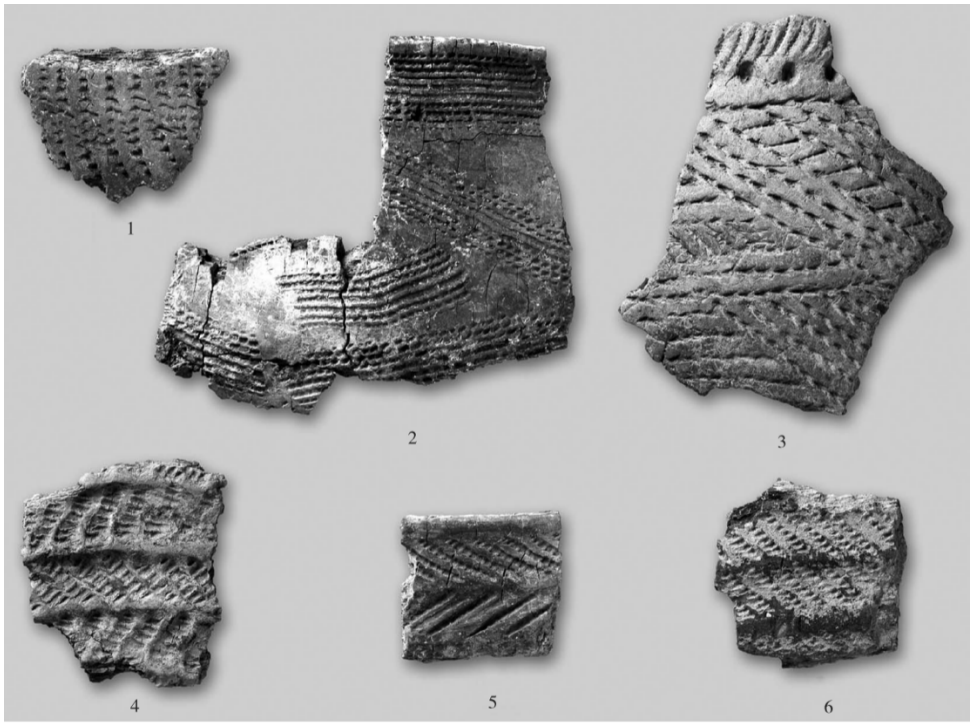


Figure 2.3. Pottery from Houtaomuga Phase 1 (Wang et al., 2016, p. 323)

Numerous questions have arisen regarding the origin of pottery vessel technology during this period, specifically if this Phase 1 pottery is the product of invention or adoption. Wang & Sebillaud (2019) believe these vessels to be the product of the adoption of ceramic technology rather than independent invention because the evidence is absent for a long period of exploration of pottery production techniques found at Houtaomuga. Instead, they argue that these vessels are the result of an extended technological progression involving adaptive changes to local environmental conditions which require further explanation. While understanding the function of pottery vessels may shed light on the choices made in their

adoption/innovation, this dissertation does not aim to determine where and when pottery technology is introduced in this area, and this topic will not be discussed in detail. Forming a hypothesis on such a large question requires further empirical evidence, i.e., pottery provenience analysis, from a much larger geographic range and is beyond the scope of this work. The research presented in this thesis will likely contribute to such discussions in the future, however, I am unable to draw such conclusions using pottery residue data alone and additional lines of evidence are needed.

The majority of unwashed samples came from Phase 1 resulting in more samples from this phase being chosen. 65 samples from Phase 1 were selected for this study from 6 features (Table 1.2). More features from Phase 1 at the site had been excavated fully at the time of sampling, resulting in more sherds from this phase being available. Phase 1 is also the earliest occupational phase at Houtaomuga, and this period of initial vessel use is of particular interest to archaeologists. Moreover, the samples are contemporaneous with other pottery in northeast Asia which has undergone similar residue analysis. With these considerations in mind, samples from Phase 1 were preferentially selected for analysis and this phase is better represented that the later phases. The yellow-brown colour of the sherds described by other researchers is visible on all Phase 1 samples as well as the fibre tempering. Some examples of this colour and tempering evidence can be seen in figure 1.9. Sample B (Figure 2.4) shows vertical voids indicative of fibre temper which has been burned away during the firing process. No samples from this phase show evidence for remaining fibres within the vessel walls. Samples A and C (Figure 2.4) are decorated with comb marking. These samples come from different features, so they are not a part of the same vessel. These samples are a good example of differences in decoration that were taken into consideration during sampling, and samples with varying decoration were assumed to be from different vessels as many of these had an all-over surface treatment of comb marking in the past.

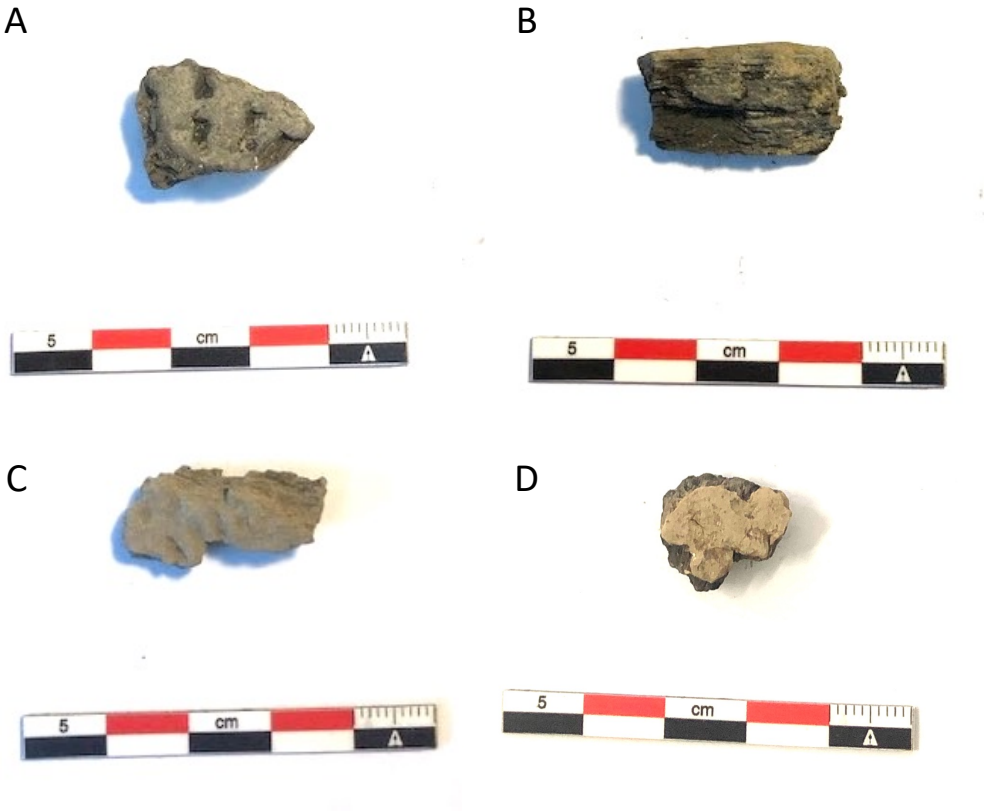


Figure 2.4. Four pottery samples from Phase 1 selected for this study. Figure A is 12 DHA III G18 (2)_S26; Figure B is 12 DHA III G18 (2)_S21; Figure C is 12DHAIII H248 (1)_S1; Figure D is 12 DHA III G21 (1)_S2.

2.3.4.2 Houtaomuga Phase 2 Pottery

Phase 2 pottery (Figure 2.5) dates to 8,000–7,000 BP (Zhang et al., 2019). Pottery from Houtaomuga Phase 2 is predominantly grey-brown with large amounts of intentionally added crushed shell inclusions (Liu et al., 2016). Due to the large quantities of inclusions and their arrangement within the clay matrix, it is likely the shell temper was crushed into the clay upon mixing and this is a very early example of this technique (Liu et al., 2016). This pottery is less brittle than pottery from Phase 1, but likely still fired at lower temperatures (<730ºC) without a kiln and vessel walls range from 0.4 to 0.8 cm thick – slightly thinner than Phase 1 (Liu et al., 2016).

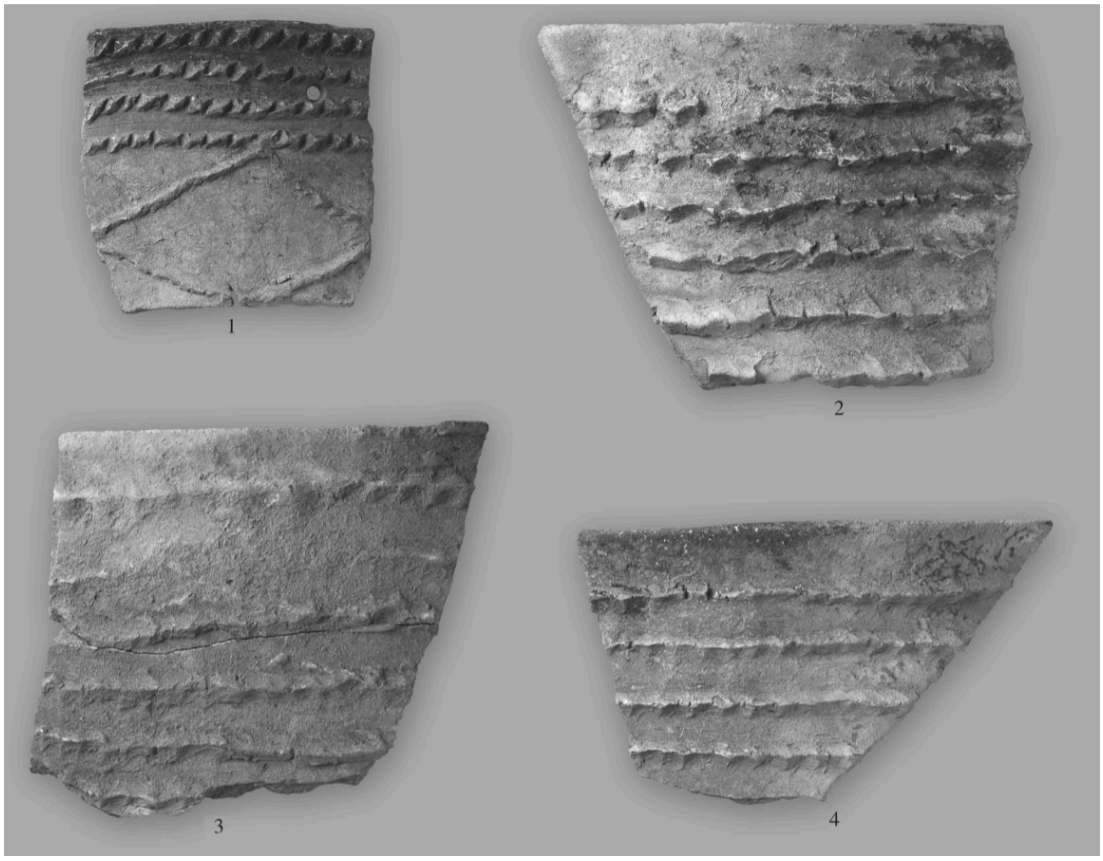


Figure 2.5. Phase 2 pottery from Houtaomuga (Wang et al., 2016, p. 325)

Less pottery samples from Phase 2 were available for sampling. A total of 21 samples from Phase 2 were selected for this study and some examples of these samples can be seen in figure 2.6. Samples from Phase 2 come from three different features (Table 1.2). The difference in colour between Phase 1 and Phase 2 can be seen here, as these samples in figure 2,6 are greyer in colour. Visible shell tempering can be seen in sample B from figure 2.6, and the extensive shell tempering described by Liu et al. (2016) can be identified clearly. Sample A shows evidence for burring either during the production process or during vessel use over a fire. This type of use-wear is prevalent in samples selected from this phase.

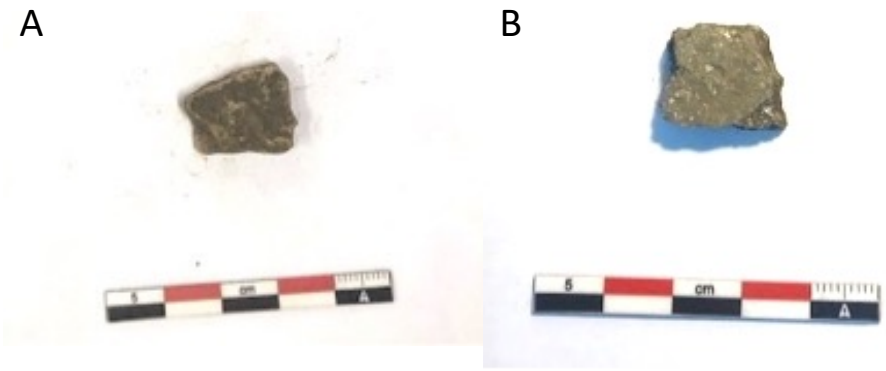


Figure 2.6. Two pottery samples from Phase 2 selected for this study. Figure A is 11 DHA III G002 (1)_S4; Figure B is 11 DHA III G13_S1

2.3.4.3 Houtaomuga Phase 3 Pottery

Phase 3 pottery (Figure 2.7) has a different brown colour than Phase 2 and dates to 6,800–5,500 BP (Zhang et al., 2019). Pottery production during the third Phase at Houtaomuga exhibits a direct continuation of the production techniques of Phase 2, including shell tempering and coiled bodies, however, the execution is more refined as displayed by uniform and directional arrangement of shell temper inclusions (Liu et al., 2016). The thickness is similar to Phase 2, but less brittle and fired at slightly higher temperatures (maximum 770ºC). Vessels in this phase are mostly decorated with patterns including nail marks and comb stamping focusing on the mouth or upper abdomen of the vessel. Two types of decoration are often included on the same vessel, and shapes are mostly cylindrical but there are a few more curved-belly jars and small-mouthed deep-bellied jars than in previous phases (Liu et al., 2016).

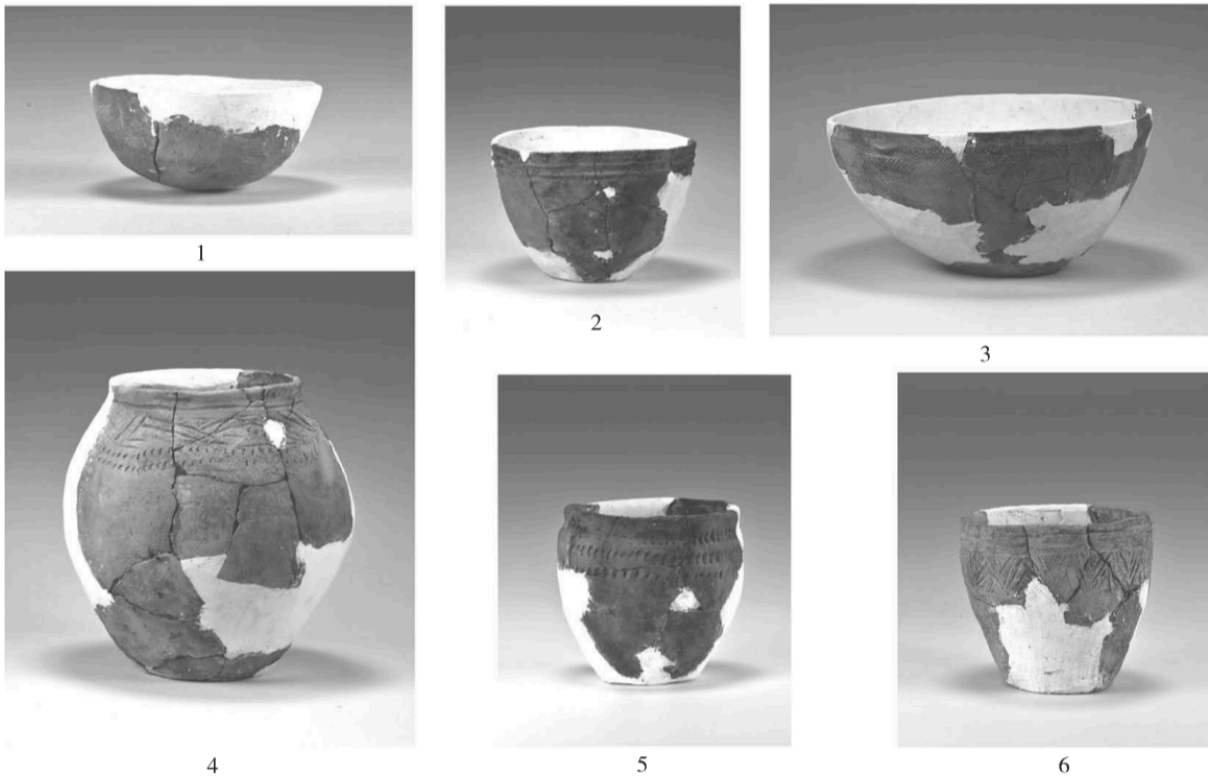


Figure 2.7. Phase 3 pottery from Houtaomuga (Wang et al., 2016, p. 326).

Less pottery samples from Phase 3 were available for sampling than the other phases and all Phase 3 samples come from one feature (table 1.2). A total of 20 samples from Phase 3 were selected for this study and some examples of these samples can be seen in figure 2.8. The difference in colour between Phase 2 and Phase 3 can be seen here, as these samples in figure 2.8 are darker in colour than figure 2.7. Visible shell tempering can be seen in sample A (figure 2.8). Decoration which includes comb marking can be seen on sample B (figure 2.8).

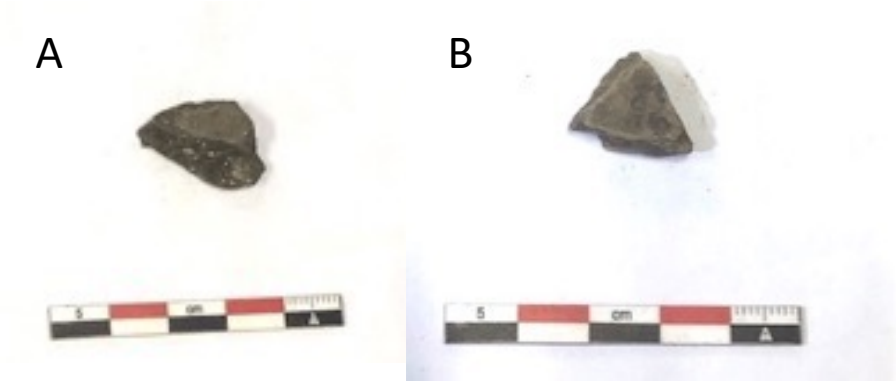


Figure 2.8. Two pottery samples from Phase 3 selected for this study. Figure A is 11 DHA III G001 (1)_S4; Figure B is 11 DHA III G001 (1)_S11

2.3.4.4 Houtaomuga Phase 4 Pottery

Phase 4 pottery (Figure 2.9) dates to 5,500 to 4,900 BP (Zhang et al., 2019). While still being coil built and open-air fired, Phase 4 pottery at Houtaomuga is thinner and fired at higher temperatures (850°C) than in previous phases (Liu et al., 2016). Sherds have significantly less shell temper than Phase 2 and 3, and this could possibly be intentional to change vessel performance in some way, however, more research is needed (Liu et al., 2016). Residue analysis could shed light on this change in vessel performance if the vessel use in this period differs from previous periods. Vessel surfaces are mostly plain but with small amounts of a pitted pattern (grid-like grass rope pattern, seen around rims in figure 2.12 vessels 2 and 3) or embossed curved zig-zag patterns and shapes include small-mouth, deep abdomen, bulging shoulders double ear pot, cylindrical pot, and bowls (Liu et al., 2016). Shell arrangements within the clay matrix are similar to Phase 2 and Phase 3, showing Phase 4 craftsmanship is likely inherited from previous phases (Liu et al., 2016).



Figure 2.9. Phase 4 pottery from Houtaomuga (Wang et al., 2016, p. 327).

A total of 47 samples from Phase 4 were selected for this study and some examples of these samples can be seen in figure 2.10. Samples from Phase 4 come from 11 different features, mainly house floors (Table 1.2). Samples from Phase 4 are harder than other phases, which coincides with their higher firing temperatures. Visible shell tempering can be seen in samples A and D from figure 2.10, and the extensive shell tempering described by Liu et al. (2016) can be identified clearly. Image A and B are the same sherd, with image A showing the interior of

the vessel and B showing the exterior. This darker exterior was assumed to be a sign for use and potential lipid deposits. This is an example of sherds that were chosen over ones that seemed unused. This type of use-wear is prevalent in samples selected from this phase.

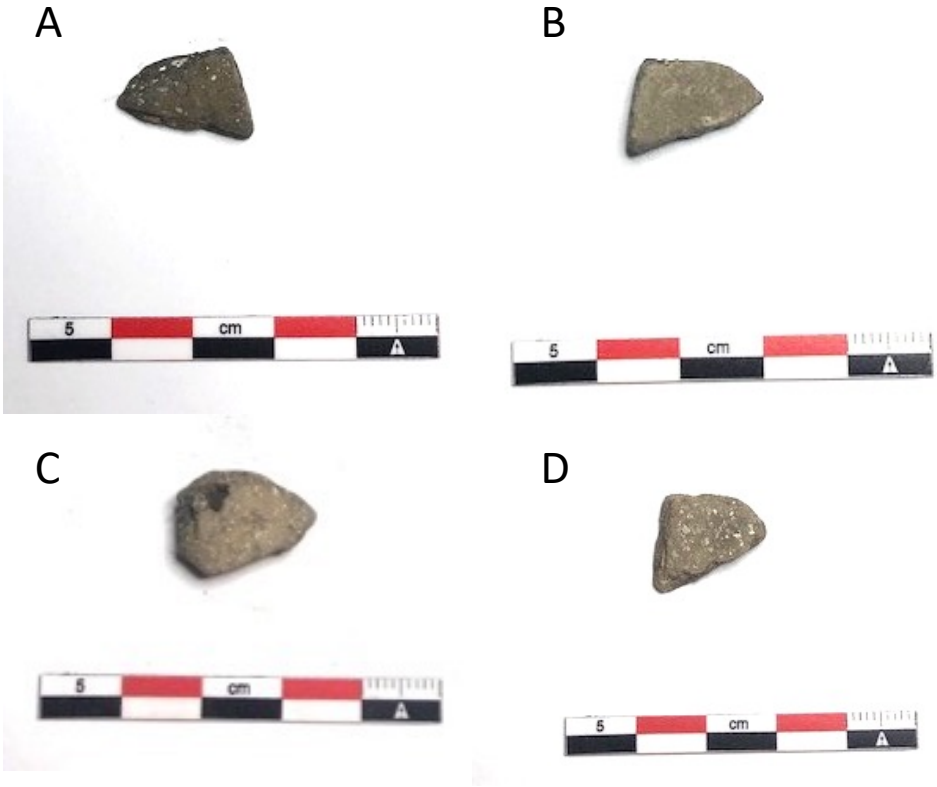


Figure 2.10. Four pottery samples from Phase 4 selected for this study. Figure A and B is 13 DHA I F008 (1)_S3; Figure C is14 DHA I F016 (1)_S2; Figure D is 15 DHA I F021 (2)_S1

Chapter 3 Results of Analytical Work

3.1 Method Development Results and Discussion

3.1.1 Acidified Methanol Extraction Procedure

One of the first steps of this research was determining an acidified methanol extraction procedure to adopt as there are currently two separate protocols. While they are overall very similar, they differ in their reaction time. This indicates the amount of time the powdered archaeological ceramic reacts with the acidified methanol at 70°C. The first procedure, developed by the University of Bristol, requires a 1-hour reaction time (Correa-Ascencio and Evershed, 2014). The second comes from the University of York and utilises a 4-hour reaction (Craig et al., 2013). Since this discrepancy is quite large and may have a considerable effect on lipid extraction, both methods were tested in triplicate with modern ceramic impregnated with olive oil. The results of this test can be seen in figure 3.1. The 4-hour reaction time significantly increased lipid yields. Specifically, yields of unsaturated fatty acids increased significantly. This is important because unsaturated fatty acids are the primary constituents of plant oils. Since the primary goal of archaeological research is to extract the majority of the organic material, rather than decrease the cost or time of the procedure, the 4-hour reaction method as outlined by Craig et al. (2013) will be undertaken for this research.

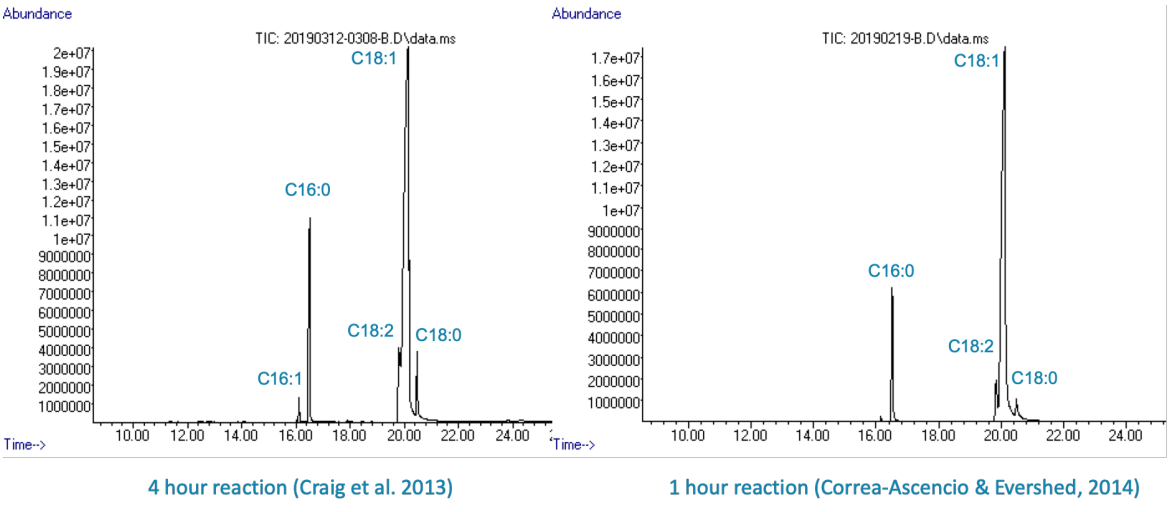


Figure 3.1. Comparison of extraction efficiencies between two prevailing acidified methanol extractions on modern olive oil. These procedures differ in their reaction times. As shown here, the 4-hour reaction (left) shows higher relative abundances of all lipid components of modern olive oil. Notably, the biggest discrepancy lies in the unsaturated fatty acids ($C_{16:1}$, $C_{18:1}$, $C_{18:2}$) which are vital components of plant oils.

3.1.2 Supercritical Fluid Extraction Optimisation

Before applying SFE techniques to the proposed material from northeast China, I applied the extraction method to experimental material and archaeological material from northwest China (Keute et al., 2021). Additional archaeological pottery from Bettys Bay South Africa as described in section 2.3.3 was also tested and results from this analysis in the form of a publication are forthcoming. The objective of these investigations was to extract and characterise surviving residues absorbed into pottery sherds. These results also shed light on the applicability of supercritical fluid extraction to various archaeological pottery types of varying ages, and the method's overall benefits and limitations. This testing is important in developing a methodology for this study on material from northeast China because a similar study has not been undertaken in the area. Unlike for some other regions, in the case of northeast China, we are unsure of what organic biomarkers might be in absorbed pottery residues and this procedure needs to prove feasibility for a series of compound classes. These findings are discussed here as a means to create a project workflow and ensure the effectiveness of the chosen method to answer my proposed hypotheses.

Modern ceramic pellets were impregnated with olive oil, artificially aged, and subsequently subjected to two SFE procedures as outlined in section 2.2.2.3. These procedures are either the previously defined dynamic extraction as outlined by Devièse et al. (2018) or the static-dynamic extraction which included an initial pressure ramp and is followed by a decrease in co-solvent. Extracts were then derivatized for characterisation and quantification by GC-MS. A table displaying this quantification for target compounds for both extraction modes can be seen in table 3.1. The results of two of these tests are seen in figures 3.2 and 3.3. Results between the two procedures are highly similar, and both produce expected quantities of each individual free fatty acid. A selection of static-dynamic extractions yielded slightly higher amounts of oleic acid; however, this result was not consistent when replicated. In light of experimental results, the method was altered for archaeological material and included a co-solvent (MeOH) to solvent (CO_2) ratio of 15%, a consistent flow rate of 2 mL/min and a consistent temperature of 50°C and pressure of 30 MPa for a total extraction time of 90 minutes. At 75 minutes, the co-solvent ratio decreases linearly from 15% to 0% at 90 minutes allowing the majority of the co-solvent to travel through the instrument.

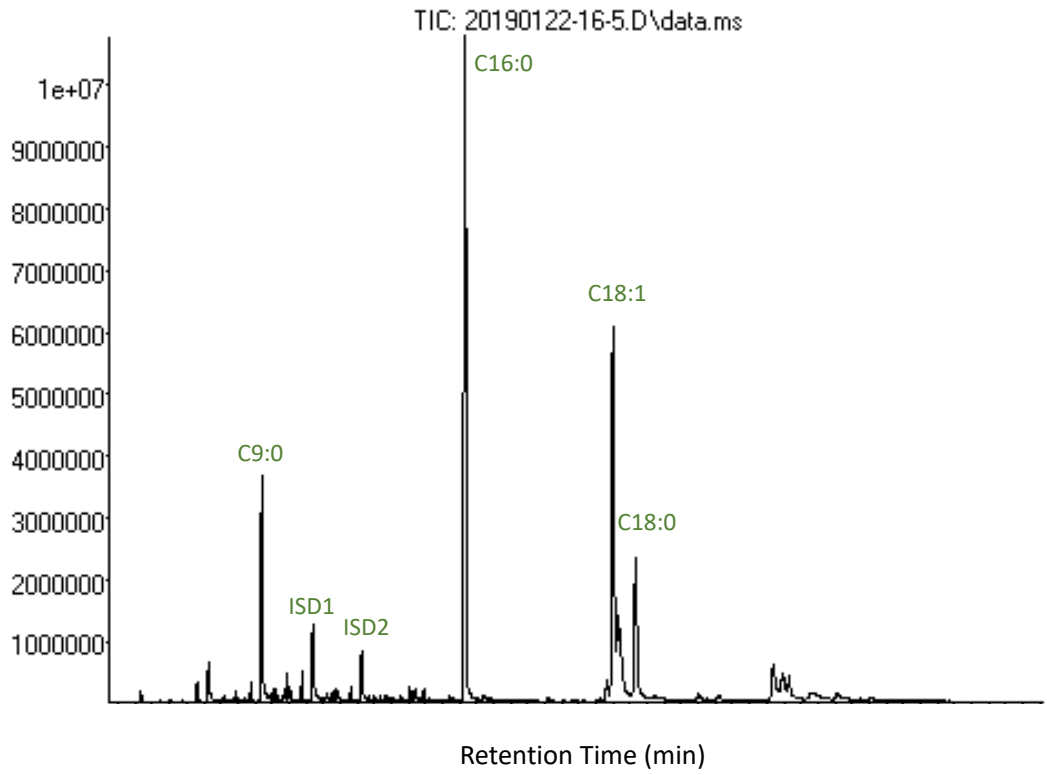


Figure 3.2. Chromatogram of dynamic extraction of ceramic pellet impregnated with olive oil and artificially aged.

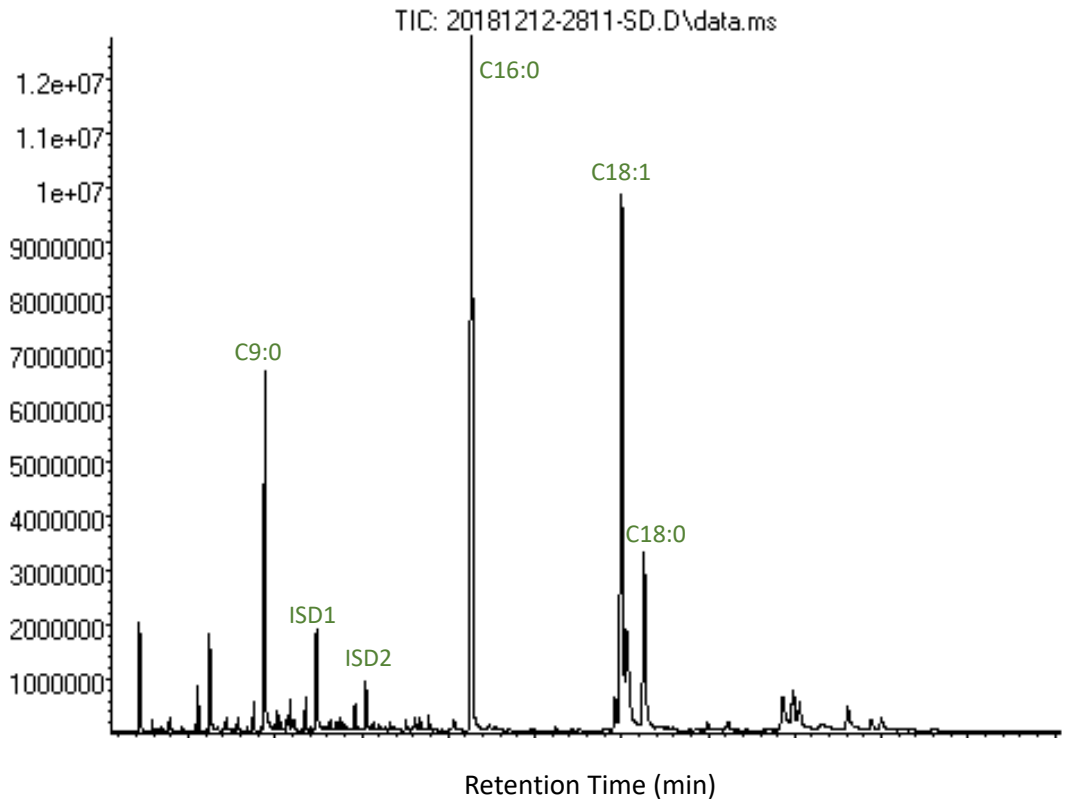


Figure 3.3. Chromatogram of static-dynamic extraction of ceramic pellet impregnated with olive oil and artificially aged.

Table 3.1. Quantification of target compounds extracted using supercritical fluid static-dynamic mode and dynamic mode. Lipid quantitation was achieved using methods detailed in appendix IX.

Sample	C16:1 µg/g	C16:0 µg/g	C18:2 µg/g	C18:1 µg/g	C18:0 µg/g
29-SFE-SD	54.7	6937.3	150.8	6920.2	390.6
29-SFE-D	49.4	8576.1	166	7750.7	1311.5
25-SFE-SD	44.8	520.0	297.2	2278.8	178.3
25-SFE-D	150.7	1185.2	183.6	457.4	332.2

Before using the optimised procedure on samples from northeast China, the procedure was used to extract lipids from archaeological pottery samples with known lipid components. One of these tests was published in a recent study by Keute et al. (2021) on pottery from The Museum of Far Eastern Antiquities (MFEA) in Stockholm. This research explored liquid and supercritical fluid extraction techniques for molecular and isotopic characterisation of lipids to understand pottery use and subsistence practices in northwest China (Keute et al., 2021). This was a pilot study conducted to establish the feasibility and prolificacy of conducting residue analysis on this collection, testing if after nearly a hundred years of storage the sherds in this collection contain sufficient residues and are not compromised too much by the intrusion of modern elements to infer on their original content and usage. This included investigating the

efficiency and applicability of three different extraction methods on this collection and evaluating their feasibility for a large-scale study. This type of prescreening is vital, as different methods of residue analysis can be well suited to different material types and target analytes.

Table 3.2. Molecules identified in 9 selected samples of pottery from northwest China which underwent different extraction techniques (Keute et al., 2021, p. 18).

	Glycerol	Caproic acid	Enanthic acid	Caprylic acid	Pelargonic acid	Capric acid	Undecylic acid	Lauric acid	Myristic acid	Pentadecanoic acid	Palmitic acid	Margaric acid	Stearic acid	Oleic acid	α-Linolenic acid	Phthalates	Benzoic Acid	Dehydroabietic acid (DHA)	7-oxo-DHA	Cholesterol	Abietic Acid	Monostearin
DASHI-K3248-02 (SFE-GCMS)	x			x	x	x	x	x	x	x	x	x		x	x	x	x	x				
SIWA-2715 (SFE-GCMS)	x			x	x	x		x	x	x	x	x			x	x	x	x	x			
XIAXIHE-K2165-3 (SFE-GCMS)	x			x	x	x	x	x	x	x	x	x	x	x	x	x					x	x
DASHI-K3248-02 (CH3-GCMS)															x							
SIWA-2715 (CH3-GCMS)		x	x	x	x					x					x							
XIAXIHE-K2165-3 (CH3-GCMS)		x	x	x	x	x			x		x				x							
DASHI-K3248-02 (H2SO4-MeOH)										x	x	x	x		x							
SIWA-2715 (H2SO4-MeOH)										x		x			x							
XIAXIHE-K2165-3 (H2SO4-MeOH)									x	x	x	x	x									

Liquid extracts and supercritical fluid extracts in this study were seemingly comparable in terms of lipid concentration and profile (table 3.2). However, these results do not represent a direct and formal comparison between liquid and supercritical extracts because lipid compounds are dispersed homogeneously within pottery vessels depending on way the vessel was used (Drieu et al., 2022). Future work comparing these extraction procedures would require extensive testing of modern experimental materials. Extracts for GC-C-IRMS analysis which underwent an acidified methanol extraction showed higher concentrations of palmitic and stearic acids. This result is expected as the procedure breaks strong interactions between the lipids and the ceramic matrix, releasing free and bound lipids from the surface as well as deeper layers of the ceramic (Correa-Ascencio and Evershed, 2014; Parker and Rae, 1998). Supercritical fluid extraction results include compounds that are not present in chloroform-methanol or acidified methanol extracts – namely, milliacin, cholesterol, glycerol monostearate, and diterpenoids such as dehydroabietic acid and 7-oxo-dehydroabietic acid. In acidified methanol extracts, glycerol monostearate would be fragmented into respective

glycerol and stearic acid methyl esters making this result unsurprising, however highlighting the benefit of this technique when working with such compounds.

The molecule miliacin was identified in the residues of four samples from northwest china and this compound can be used as a biomarker for millet (Figure 3.4 and 3.5). During the Neolithic, millet was the dominant crop produced in northwest China (Lee, 2016). By 7,000 BP, settlements with domesticated broomcorn millet emerge in the western Loess Plateau at Dadiwan (Bettinger et al., 2007). This compound is important to identify in test samples before testing material from northeast China because this area was a centre for millet domestication and the full extent of the use of this food at Houtaomuga is unknown.

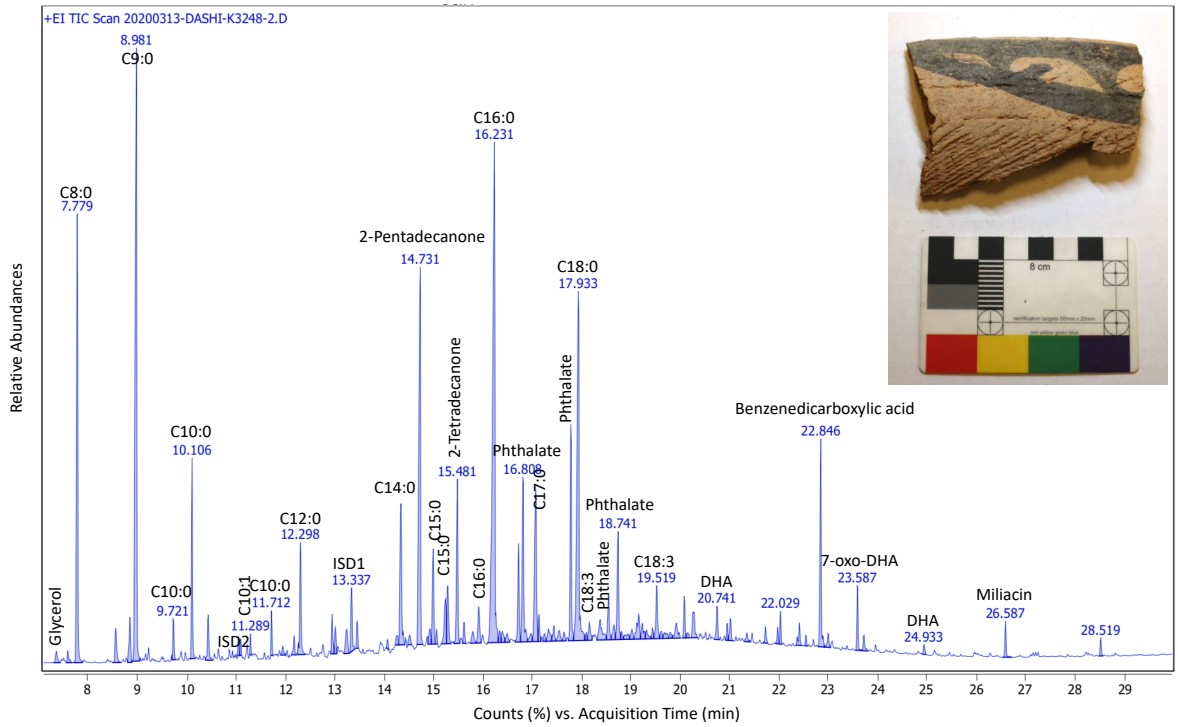


Figure 3.4. Chromatogram of lipid profile extracted from pottery sherd using supercritical fluid. This samples shows miliacin, a biomarker for millet being extracted using the novel technique(Keute et al., 2021, p. 10).

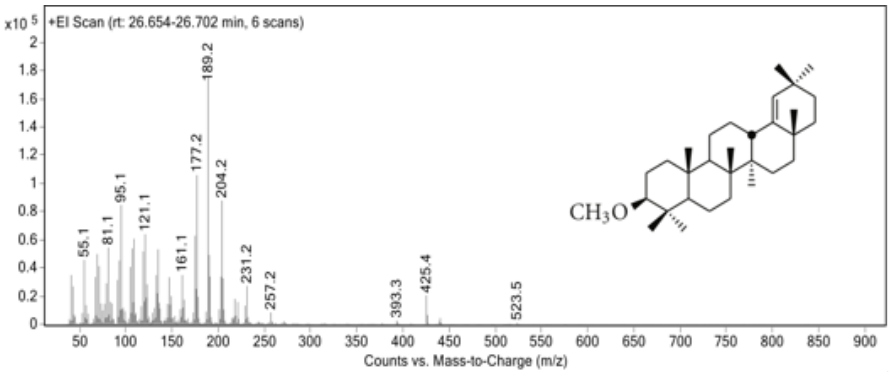


Figure 3.5. Mass spectra and structure of miliacin, a biomarker for millet from sample in figure 3.4 (Keute et al., 2021, p. 11).

The compound miliacin was not identified in the liquid extracts of this pilot study, however, it was identified in supercritical extracts. This may occur for various reasons. Firstly, lipid profiles are not homogenous in pottery samples, and it may have only been present in the sherd portions which underwent SFE. Next, this may indicate the SFE procedure is yielding higher lipid concentrations or is better suited for certain lipid classes. This differentiation would be vital to better understand for archaeological chemistry. To certify the latter, more experimental research is needed which is outside the scope of this work. For chemical analysis on pottery from northeast China, the ability of the SFE procedure to extract this compound is of primary importance. Domesticated millet appears rapidly and extensively in northeast China during the early Holocene and in a broad region over North China by 8,000–7,500 cal. BP, meaning the domestication process was initiated far earlier (Lee, 2016). It is feasible that pottery from northeast China selected in the study may contain this biomarker, making its efficient extraction and characterisation in the chosen methodology crucial.

Diterpenoids identified in pottery from northwest China are common in archaeological assemblages as they derive from trees in the *Pinaceae* family, often used in the form of waterproofing agents, glues, and in funerary practices (Bonaduce and Colombini, 2004; M P Colombini et al., 2003; Devièse et al., 2010; Ribechini et al., 2008). The compounds identified in this study can derive from the thermal dehydrogenation of abietic acid naturally present in the wood, and when this occurs in an oxygenated environment, produces 7-oxo-dehydroabietic acid, and this is likely where they derive from in this context (Colombini and Modugno, 2009). Diterpenoids and additional compounds including polycyclic aromatic hydrocarbons deriving from a resin of the *Pinaceae* family absorbed into the ceramic matrix have also been successfully extracted using supercritical fluid. Absorbed and encrusted residues from Roman amphorae dating to the second half of the 5th century AD from the site of Gerace, Sicily were examined using both the traditional liquid extraction method and the new procedure using supercritical fluids (figure 3.18-3.20). Extracts obtained on these amphorae using both extraction methods have been quantified and compared using GC-MS. This study demonstrates the supercritical fluid extraction technique is successful in extracting these compounds which are highly common in archaeological contexts in high concentrations. Future archaeological investigations seeking to extract similar compounds may benefit from employing this technique, particularly when necessitating non-destructive pathways of analysis.

The SFE procedure was also tested on pottery from South Africa to extract a wide range of saturated and unsaturated free fatty acids, wax esters, APAA's, and long-chain alcohols from a large archaeological assemblage. In this study, SFE was applied to a large set of potsherds (n=110) from the Bobartia Road Midden Site, Betty's Bay, Western Cape, South Africa. These sherds were selected, as they contain unsaturated fatty acids indicative of fish fats, and the ability of the method workflow to preserve and identify these compounds is important to apply this workflow in northeast China, as preliminary research suggests fish was a commonly utilized resource (Wang and Sebillaud, 2019). In addition to evidence of higher lipid yields than chloroform-methanol extractions, SFE was selected to preserve the sherds and return them after residue analyses as required by permitting. Through SFE and consecutive characterisation and quantification by gas chromatography-mass spectrometry (GC-MS), this research allowed us to better understand well preserved organic residues and pottery use at this site. This archaeological application differs from the analysis of museum material from northwest China, as the South Africa material is recently excavated, a far coarser pottery type, and contains a distinctive suite of organic compounds.

In summary, these results show the SFE procedure developed is capable of extracting relevant absorbed lipid material and biomarkers from archaeological pottery of various porosities and ages at high concentrations. These compounds include APAA's, waxes, resins, diterpenoids, triterpenoids, mono- and dicarboxylic acids and sterols. While organic contents of samples from northeast China are unknown, current literature suggests the presence of lipids deriving from fish or millet may be present. If correct, the procedure developed will be able to extract these absorbed residues, furthering our understanding of pottery function during the early Holocene.

3.1.3 Gas Chromatography-Mass Spectrometry Optimisation

Initially, samples were analysed using GC-MS with a split/splitless injection following Colombini et al. (2003) with modifications. While this method is effective at separating a series of lipid materials, it is limited in the MS scan range of m/z 50-650. Combined with utilising lower temperatures, this method is, therefore, unable to characterise analytes at higher molecular weights and/or boiling points. Similar procedures are routine in many archaeological applications (Dunne et al., 2018; Ribechini et al., 2008; Salque et al., 2013; Smyth and Evershed, 2016). When archaeological residue analysis was in its infancy, it was presumed that

large lipid molecules such as triglycerides did not survive antiquity – thus focusing on constituent degradation products and free fatty acids (Evershed, 2008). Recently, new methods have improved extraction procedures, and we know such components can survive under certain conditions (Drieu, 2018). Shorter and higher temperature methods have become more preferred in recent years to efficiently screen for palmitic and stearic acids before compounds specific isotopic characterisation, and additional larger compounds (Dunne et al., 2012).

Presumably, to target larger analytes, archaeological chemists have developed methods of liquid chromatographic separations of lipids (Lucejko et al., 2018; Manzano et al., 2019; Saliu et al., 2011). These techniques are highly promising and show high-quality separations. One difficulty however of the overall adoption of such a technique is that LC is considerably more expensive than GC and produces substantive solvent waste which requires proper disposal. There are current methods for GC-MS analysis that screen for triglycerides and other large lipids (Dunne et al., 2019). As mentioned previously, these analyses use a limited MS scan range, and retention times are lengthy.

To mitigate some of these factors, a series of tests on the GC-MS instrument involving different columns and chromatographic conditions were undertaken to improve untargeted analysis. The main goal of this analysis was to include high molecular weight analytes such as triglycerides into lipid screening. The columns tested were the Restek© Rtxi-65TG column (30m x 0.25mm ID x 0.1 film thickness, crossbond diphenyl dimethyl polysiloxane stationary phase) and the Agilent© DB-5ms column (30m x 0.25mm ID x 0.25 film thickness, nonpolar phenyl arylene polymer, virtually equivalent to (5%-phenyl)-methylpolysiloxane), both on on-column injections with a deactivated silica precolumn. Four GC-MS methods were tested, and their conditions can be seen in figure 3.6 and table 3.3. Each method was tested using both stationary phases using an on-column injection for all reference and archaeological materials for comparison. Each method began at 35°C and was held for 2 minutes followed by a ramp in pressure at various rates to different maximum temperatures.

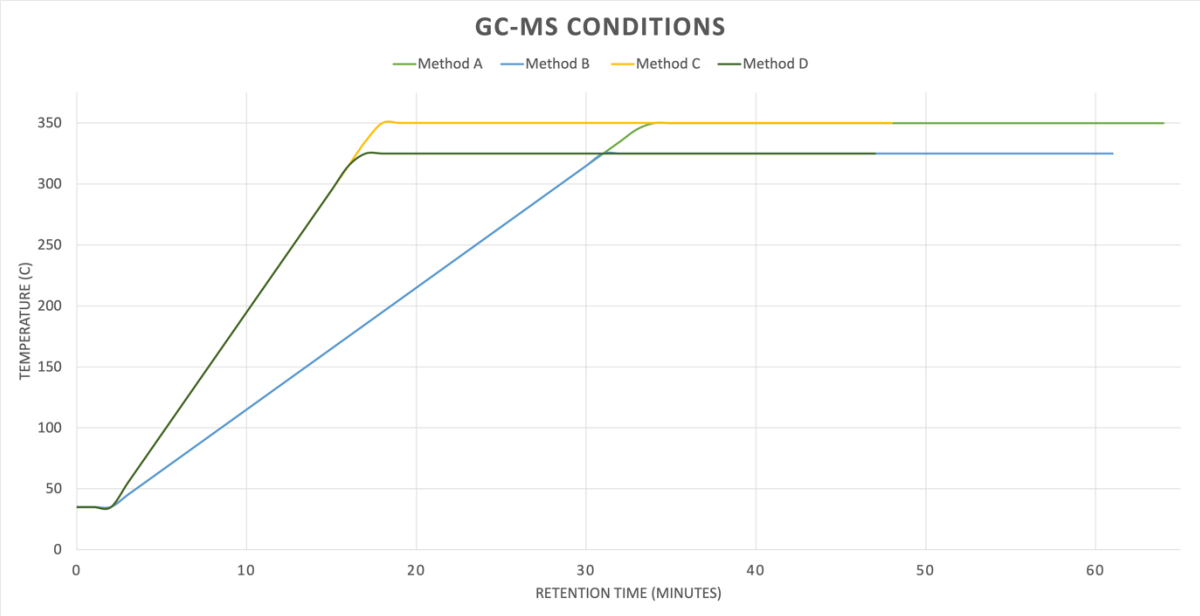


Figure 3.6. illustration of GC-MS method conditions tested in this thesis

Table 3.3. GC-MS method conditions tested in this thesis

	Method A	Method B	Method C	Method D
Initial Temperature	35°C	35°C	35°C	35°C
Ramp	10°C/min	10°C/min	20°C/min	20°C/min
Final temperature	350°C	325°C	350°C	325°C
Hold time	30 Minutes	30 Minutes	30 Minutes	30 Minutes
Total run time	64 Minutes	61 Minutes	48 Minutes	47 Minutes

The conditions outlined above were applied to the separation of various reference materials to test the efficiency of the columns and chromatographic conditions. These materials were chosen as they are the most common groups identified in the current literature. The first of these tests was an alkane standard solution (C_8-C_{20}, Millipore-Sigma©). In all applications, the Restek© Rtxi-65TG column showed superior thermal stability and column bleed was negligible (figure 3.7). Applications at lower temperatures still showed significant bleed of the Agilent© DB-5ms column (Figures 3.8 and 3.9). Peak resolution of alkane standards increased when switching from the the Rtxi-65TG column to the DB-5ms column, particularly for higher chain alkanes, however peak asymmetry improved. Furthermore, lower temperatures combined with the DB-5ms column also increased elution times significantly. This is highlighted in figure 3.8 showing the Agilent© DB-5ms column and GC-MS method D, as the late eluting (RT 44.037) C_{20} peak exhibits significant tailing (figure 3.8) compared to the same alkane when eluting on the Rtxi-65TG column (RT 32.199) with GC-MS method A (figure 3.7). This compound is also showing tailing and eluting later (RT 34.403) with increased temperatures (figure 3.9). For

quicker alkane separation with symmetrical peak shape, the Rtxi-65TG is preferred. For smaller compounds, the DB-5ms column is preferred for improved resolution.

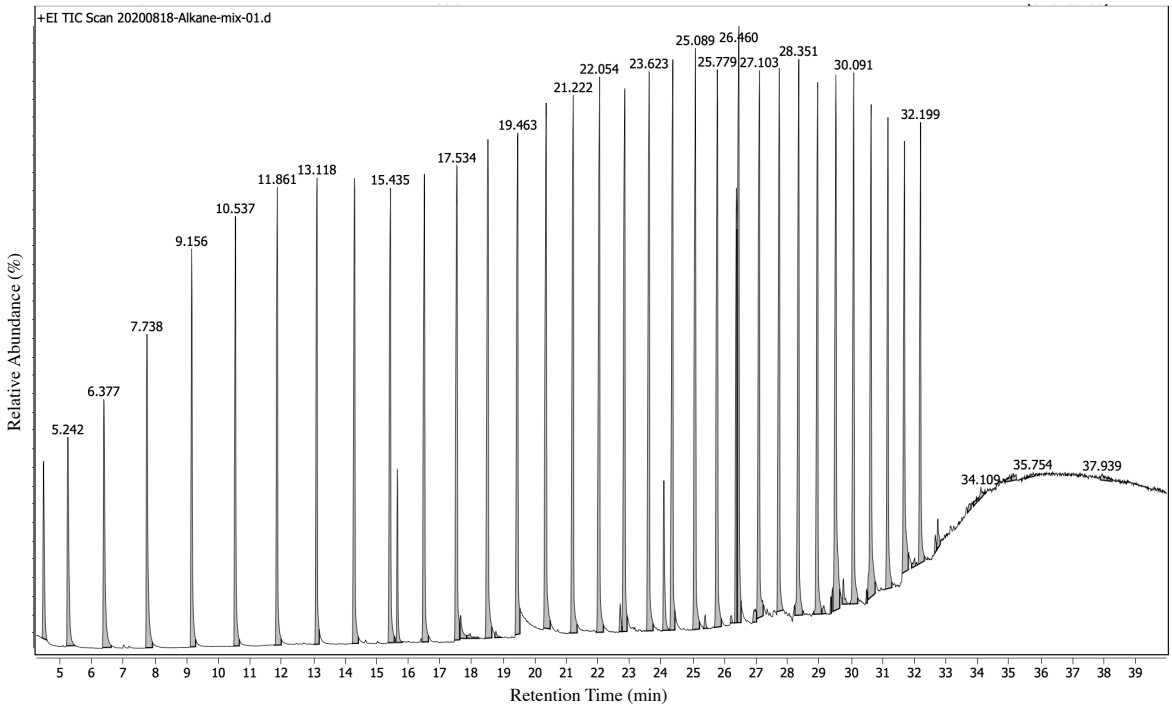


Figure 3.7. Chromatogram of alkane mixture (C_8-C_{20}) on Restek© Rtxi-65TG column and GC-MS method A (10°C/min ramp to a maximum temperature of 350°C)

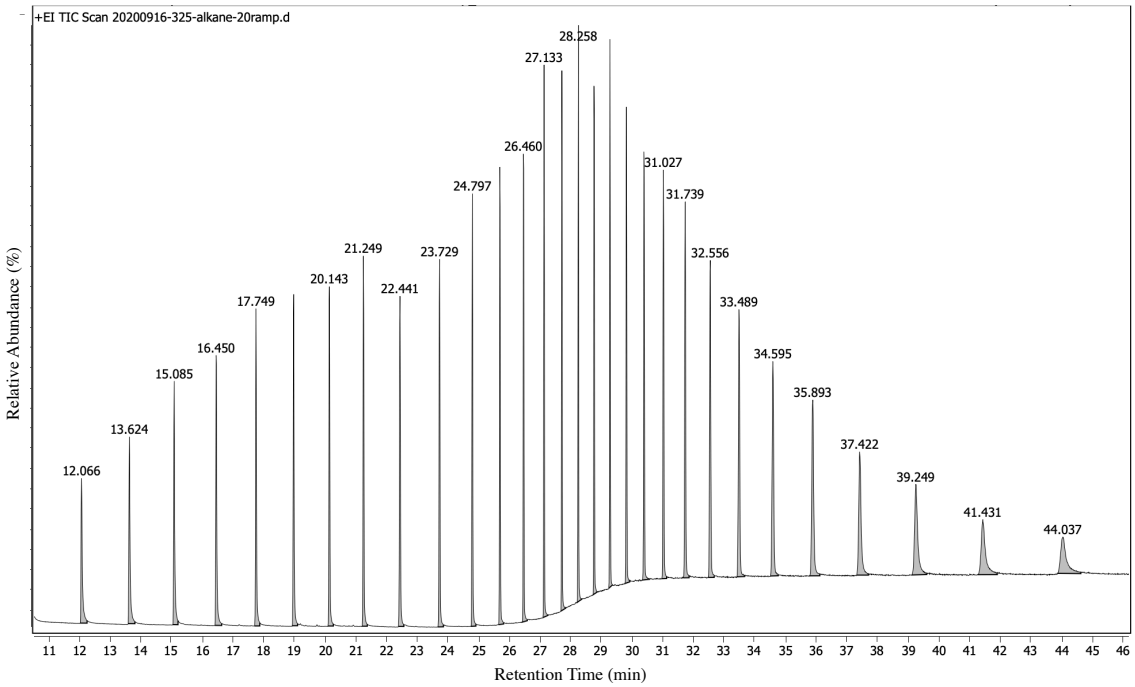


Figure 3.8. Chromatogram of alkane mixture (C_8-C_{20}) on Agilent© DB-5ms column and GC-MS method D (20°C/min ramp to a maximum temperature of 325°C)

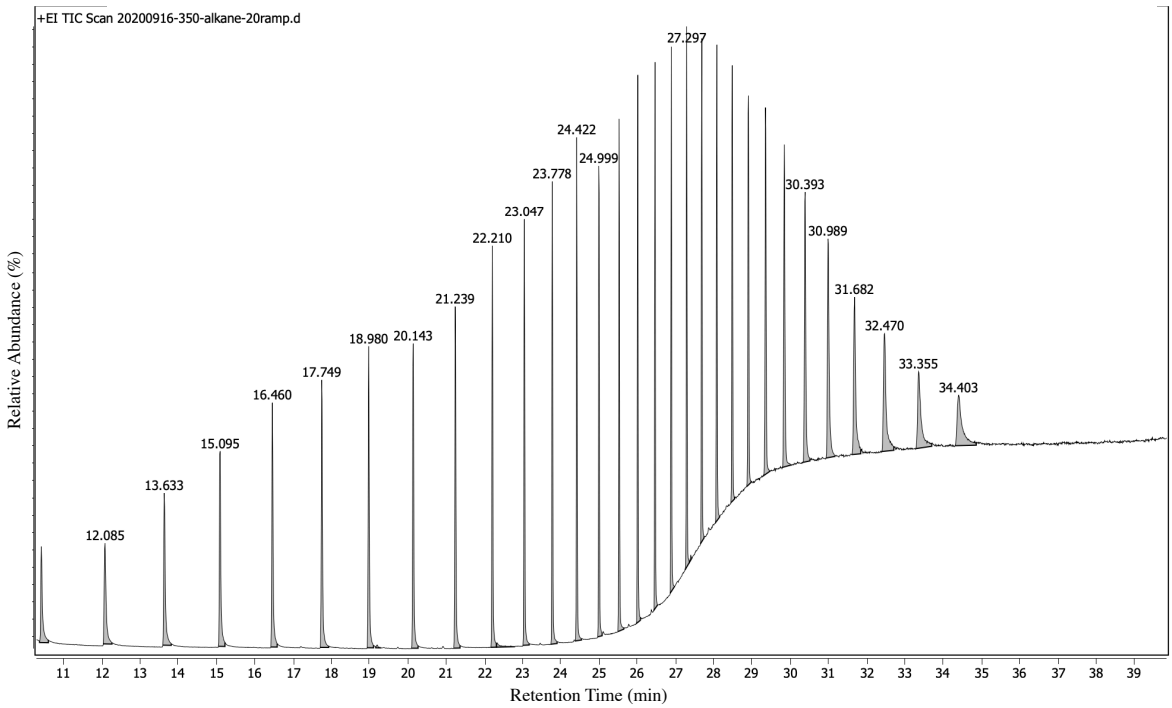


Figure 3.9. Chromatogram of alkane mixture (C_8-C_{20}) on Agilent© DB-5ms column and GC-MS method C (20°C/min ramp to a maximum temperature of 350°C)

The next reference material examined was beeswax. For over a millennia, beeswax has been used in technological, cosmetic, and medicinal settings, and is often found in archaeological ceramics (Rageot et al., 2016; Roffet-Salque et al., 2015). Beeswax consists of a complex suite of lipids involving n-alkanes, n-alkanoic acids and fatty acyl wax esters (Roffet-Salque et al., 2015). Similar to the alkane tests, the Rtxi-65TG column exhibited high thermal stability throughout the examination and the column bleed was negligible. The opposite is true for the DB-5ms column which exhibited consistently increased background noise with increasing temperatures. This is improved slightly in applications running at 325°C versus 350°C but the concern is still prevalent. For the DB-5ms column, the preferred application would be at GC-MS method D, however, this method still exhibits a high background and long-chain alkanes, and wax esters have poor resolution, significant peak asymmetry, and elute later than other methods (figure 3.10). The preferred method for the Rtxi-65TG column would be the GC-MS method C as the elution time is shorter and peak shapes precise. One concern with this application however compound separation is not as complete and resolution is poor (less than 1.5 R, Appendix VIII) as methods employing the 10°C/min method (figure 3.11 and 3.12). If the desired goal of the operator is complete peak separation for preparative work, for example, the 10°C/min method would be preferred.

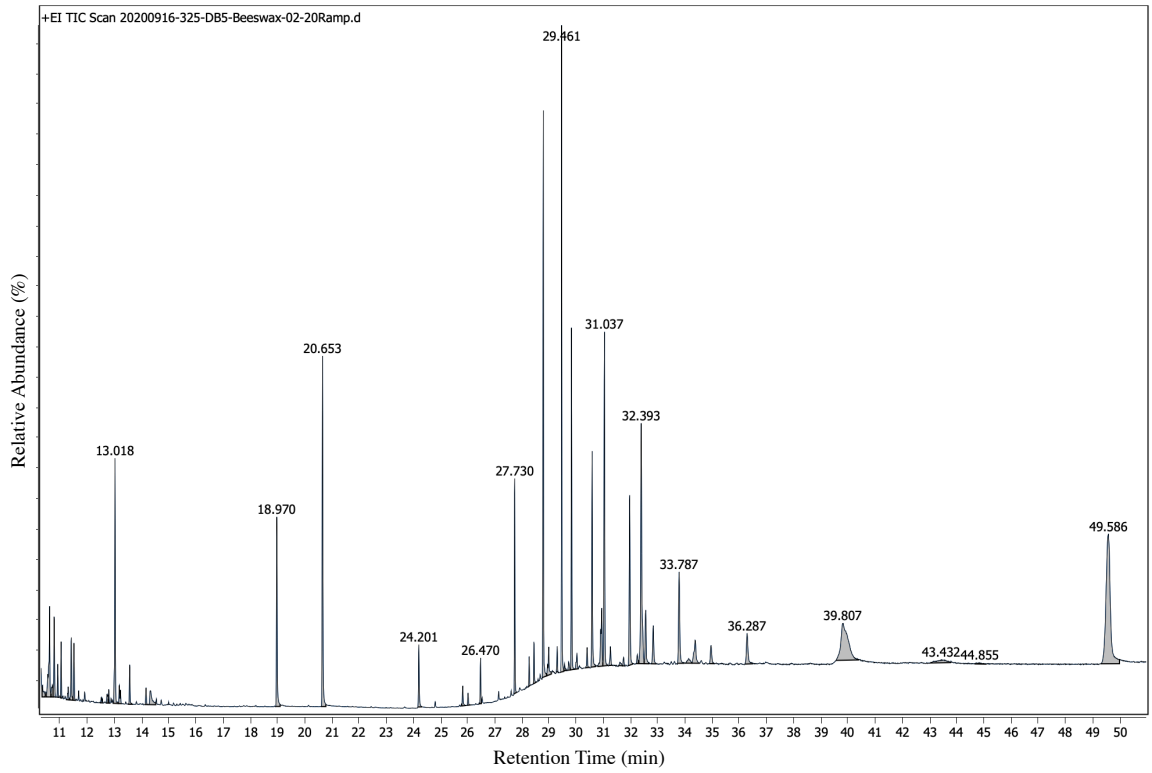


Figure 3.10. Chromatogram of beeswax on Agilent© DB-5ms column and GC-MS method D (20°C/min ramp to a maximum temperature of 325°C)

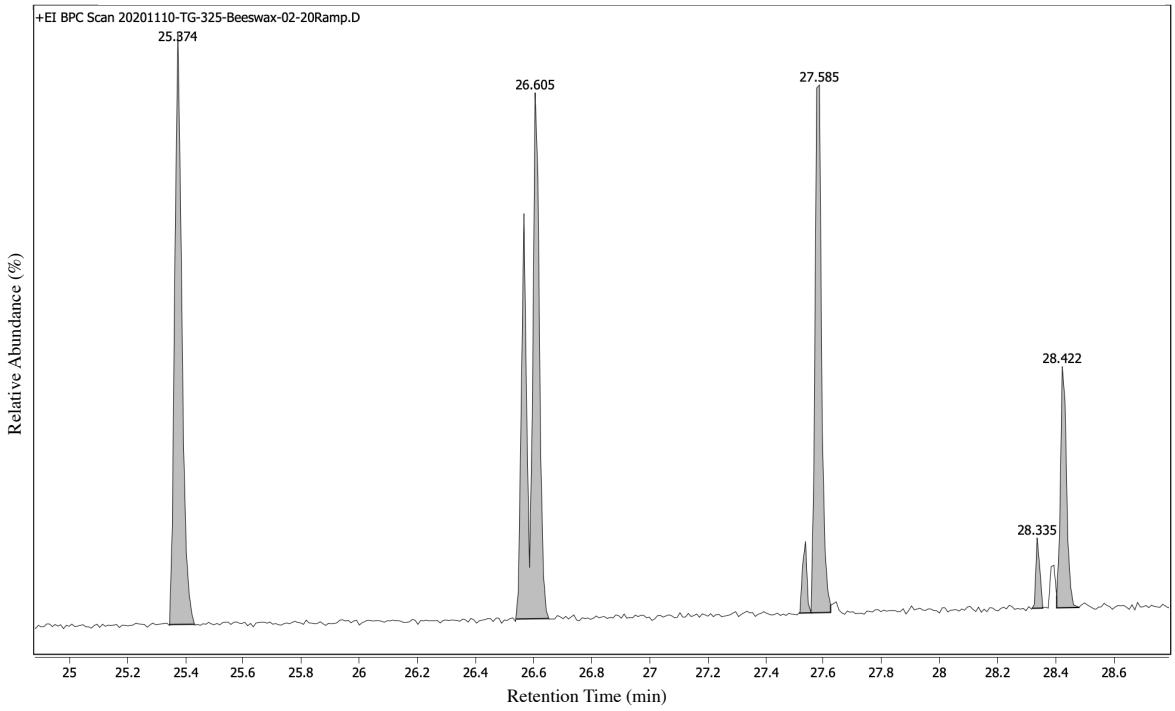


Figure 3.11. Chromatogram of beeswax on Restek© Rtxi-65TG column and GC-MS method D (20°C/min ramp to a maximum temperature of 325°C) detailing poor resolution of n-alkane and alcohol at 26.605 minutes

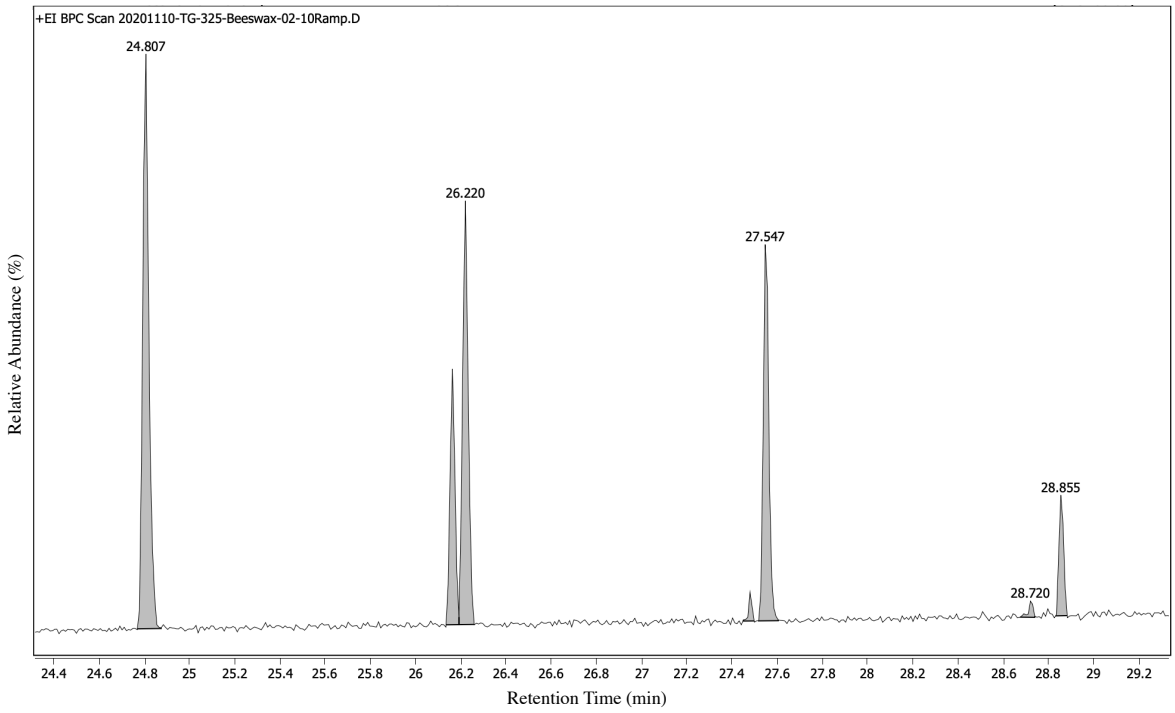


Figure 3.12. Chromatogram of beeswax on Restek© Rtxi-65TG column and GC-MS method B (10°C/min ramp to a maximum temperature of 325°C) detailing improved separation of n-alkane and alcohol at 26.220 minutes

Lastly, a mixture of alkane standards (C_8-C_{20}, Millipore-Sigma©), free fatty acids (saturated and unsaturated, $C_{8:0}$-$C_{24:0}$, Millipore-Sigma©), and triglycerides ($TG_{8:0/8:0/8:0}$, $TG_{10:0/10:0/10:0}$, $TG_{12:0/12:0/12:0}$, $TG_{14:0/14:0/14:0}$, $TG_{16:0/16:0/16:0}$, Millipore-Sigma©) were examined using the chromatographic conditions described on both columns. As with the previous two

examinations, column bleed on the Rtxi-65TG was significantly lower than the DB-5ms. Separation on the DB-5ms column was slower compared to the Rtxi-65TG in all methods tested. The largest triglyceride tripalmitin ($TG_{16:0/16:0/16:0}$) was either missing or poorly/late eluted in each application making this column a poor choice for such large compounds. Free fatty acid and alkane separation was sufficient (figure 3.13). Separation of the various lipid classes on the Rtxi-65TG was sufficient in all applications. Most notably, the resolution and peak shape of the various triglycerides was improved with increased ramp temperature. In figure 3.14 and figure 3.15, tripalmitin elution time is decreased from 43 to 38 minutes and peak resolution is greatly improved. For this reason, the Rtxi-65TG with GC-MS method C is preferential for this broad mixture.

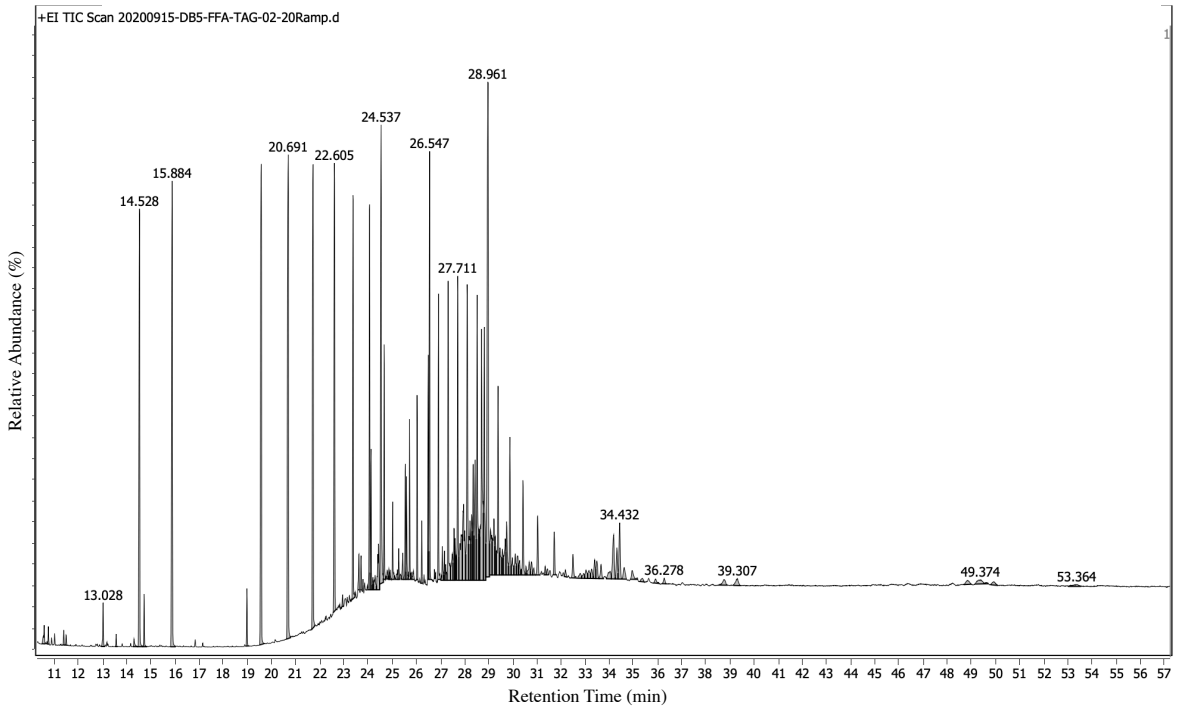


Figure 3.13. Chromatogram of free fatty acid, alkane, and TAG mixture on Agilent© DB-5ms column with GC-MS method C (20°C/min ramp to a maximum temperature of 350°C)

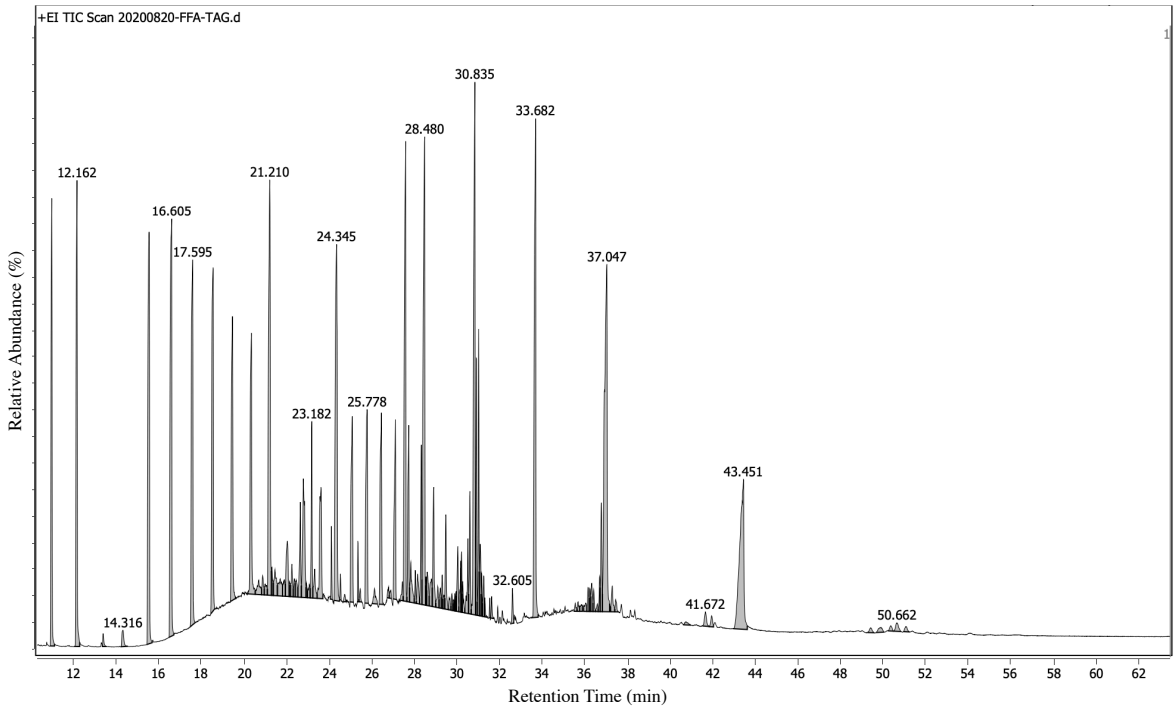


Figure 3.14. Chromatogram of free fatty acid, alkane, and TAG mixture on Restek© Rtxi-65TG column and GC-MS method A (10°C/min ramp to a maximum temperature of 350°C)

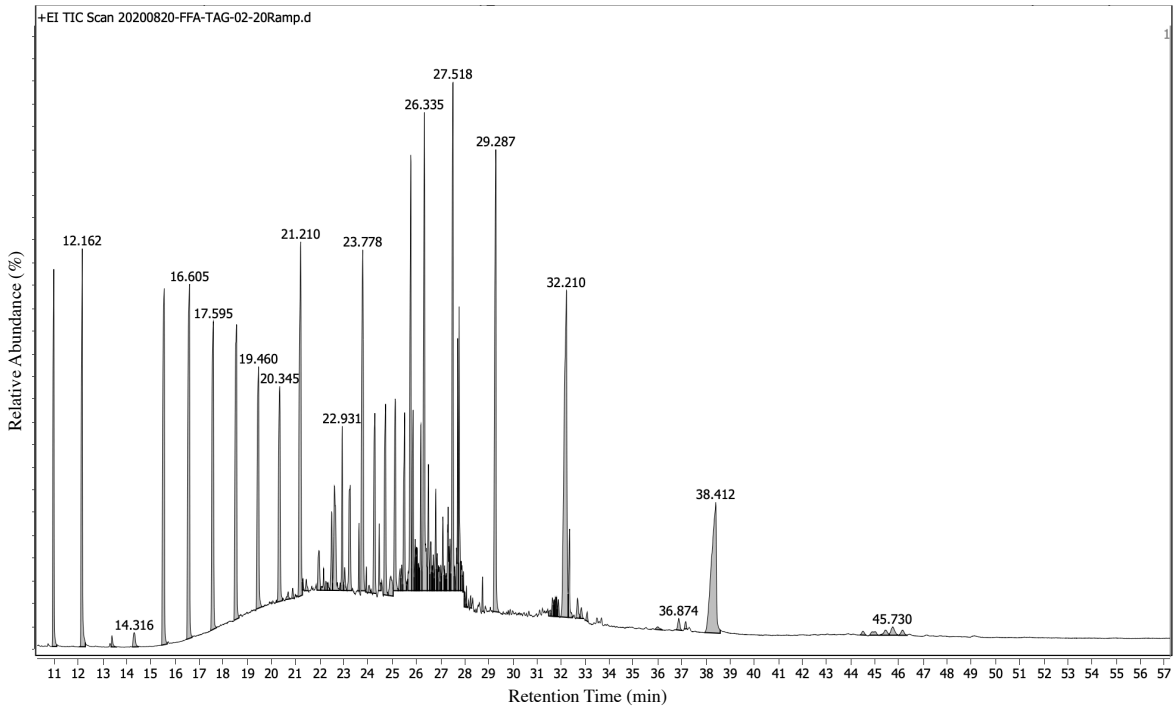


Figure 3.15. Chromatogram of free fatty acid, alkane, and TAG mixture on Restek© Rtxi-65TG column GC-MS method C (20°C/min ramp to a maximum temperature of 350°C)

The same chromatographic conditions (figure 3.6; table 3.3) were applied to modern reference materials were used on archaeological materials to evaluate the application on aged materials. The first of these samples are absorbed lipid residue from archaeological ceramics from Bettys Bay, South Africa and contain a mixture of monocarboxylic acids, fatty acyl wax esters and n-

alkanes. These samples were chosen to evaluate the separation of this complex mixture which has undergone an authentic degradation process that can be difficult to reproduce in a lab. I have included samples from two separate contexts which have both undergone supercritical fluid extraction (LFS-7 & CFS2-4). Total lipid extracts from these samples underwent the same conditions as highlighted above.

For both samples, the Rtxi-65TG showed significant thermal stability. Increased column bleed in the DB-5ms can be observed in figure 3.16. Resolution on quicker eluting and lower abundance compounds decreases with the Rtxi-65TG, and this elution is much sharper with the DB-5ms. The resolution on both columns is comparable to later eluting compounds. Overall, elution times on the Rtxi-65TG are faster and retention times are shortened for nearly all compounds without sacrificing peak symmetry. Sample LFS-7 contains long-chain fatty acyl wax esters which include Free fatty acids and long-chain alcohols and n-alkanes. In figure 3.17, some of the largest of these compounds can be seen eluting at 28.393 and 29.528 minutes. Comparatively, these compounds are eluting at 29.566 and 30.633 on the same method with the DB-5ms column (figure 3.18) and separation between these larger compounds is quicker. This trend is observed across the board with these columns, where elution times are quicker with increased peak shape on the Rtxi-65TG. For the DB-5ms column, the preferred method would be GC-MS method C (20°C/min to 325°C) for both LFS-7 and CFS2-4. The preferred condition for the Rtxi-65TG is GC-MS method A (Figure 3.17; 10°C/min reaching 350°C).

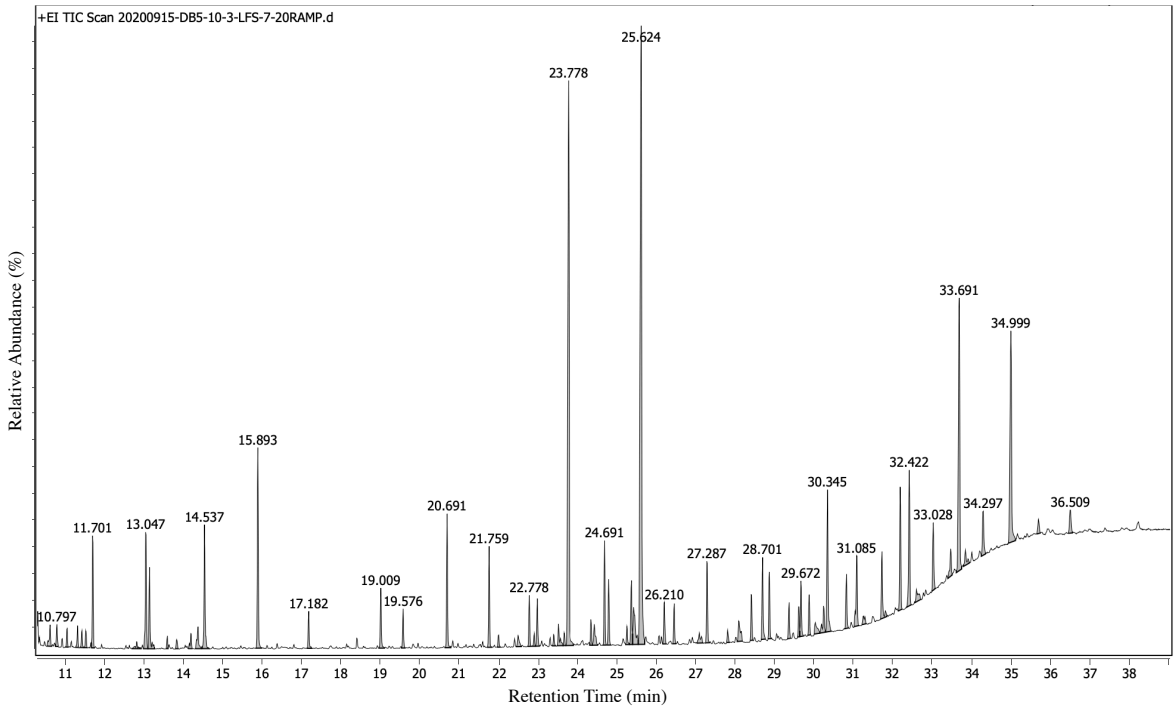


Figure 3.16. Chromatogram of sample LFS-7 from Betty's Bay, South Africa on Agilent© DB-5ms column GC-MS method C (20°C/min ramp to a maximum temperature of 350°C). Peak assignments can be seen in appendix X.

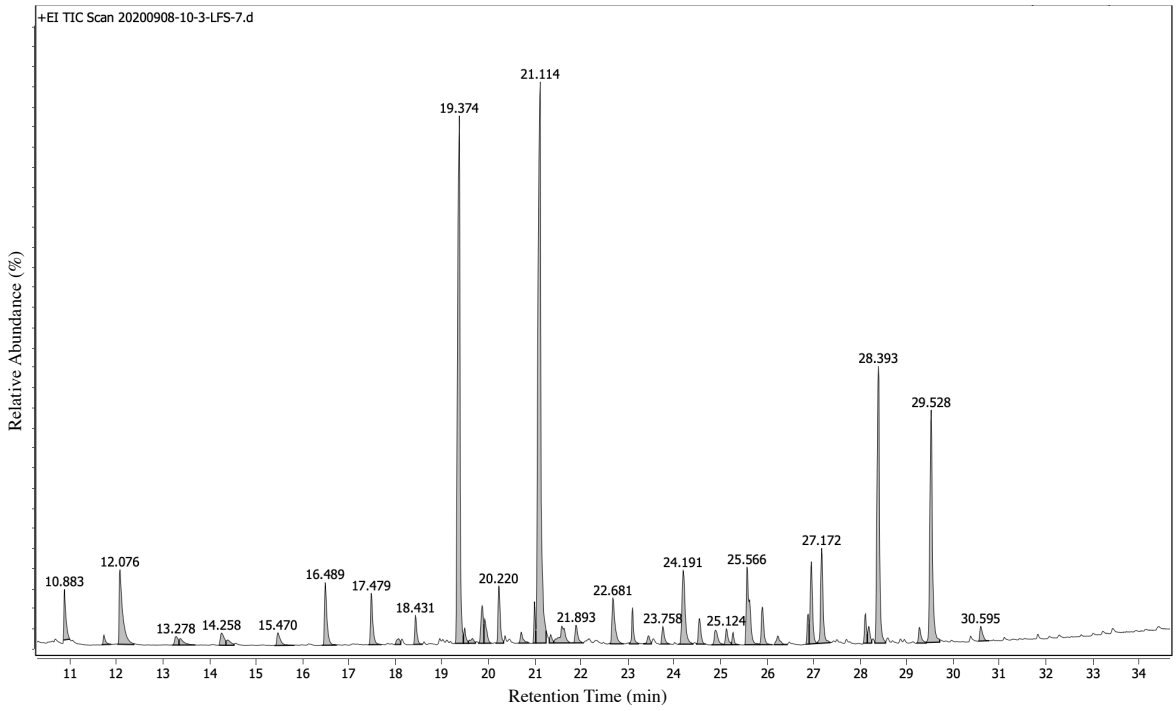


Figure 3.17. Chromatogram of sample LFS-7 from Betty's Bay, South Africa on Restek© Rtxi-65TG column and GC-MS method A (10°C/min ramp to a maximum temperature of 350°C). Peak assignments can be seen in appendix X.

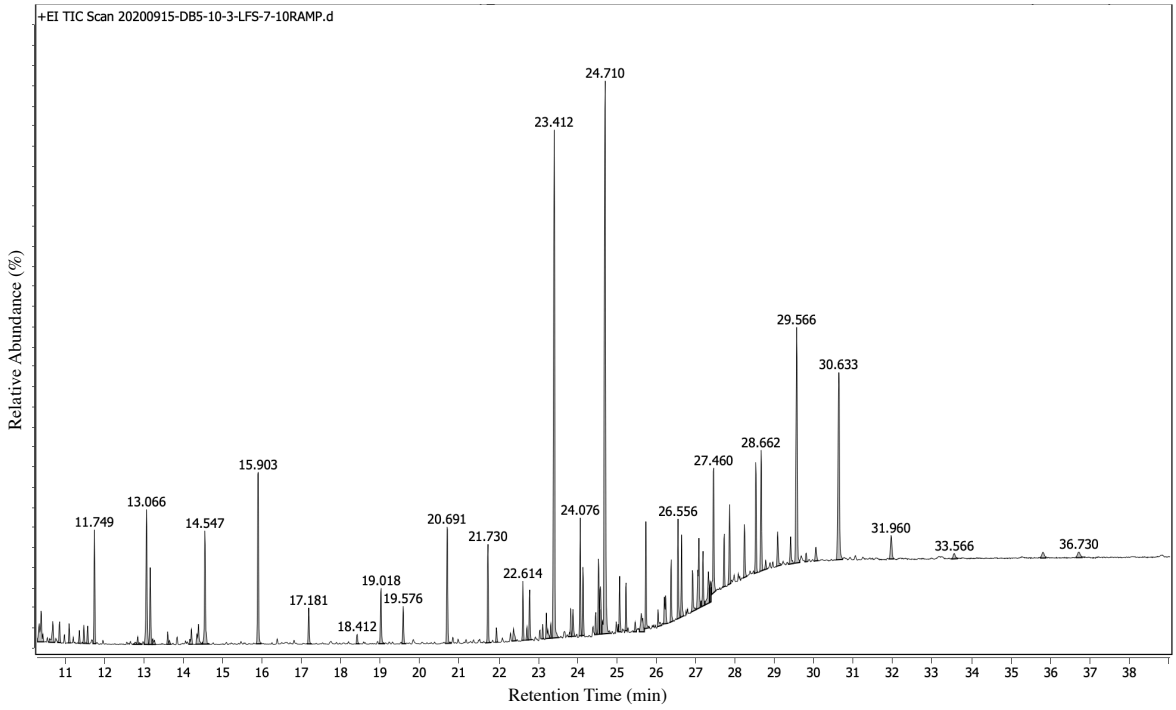


Figure 3.18. Chromatogram of sample LFS-7 from Betty's Bay, South Africa on Agilent© DB-5ms column and GC-MS method A (10°C/min ramp to a maximum temperature of 350°C). Peak assignments can be seen in appendix X.

The second archaeological sample is a resinous material that has been removed from a roman amphora dating to the second half of the 5th century AD from the site of Gerace, Sicily. Previous analysis has shown this resin comes from trees in the *Pinaceae* family and mainly

contain abietic acids along with other polycyclic aromatic hydrocarbons and free fatty acids. This resin type is often identified archaeologically, and the method in question must be able to confidently characterise this.

Separation with the DB-5ms column is quicker on all applications. Lower temperatures maxing out at 325°C (GC-MS methods B and D) show reduced column bleed and are preferred for analysis, however increasing temperature decreases analysis time (figure 3.19 and 3.20). The Rtxi-65TG shows adequate resolution and improved peak shape with decreased elution times for larger compounds (figure 3.21). For this particular type of residue, the Rtxi-65TG column with GC-MS method B (figure 3.22) has an ideal chromatographic profile with all no compounds co-eluting, little background signal, and overall quick elution times.

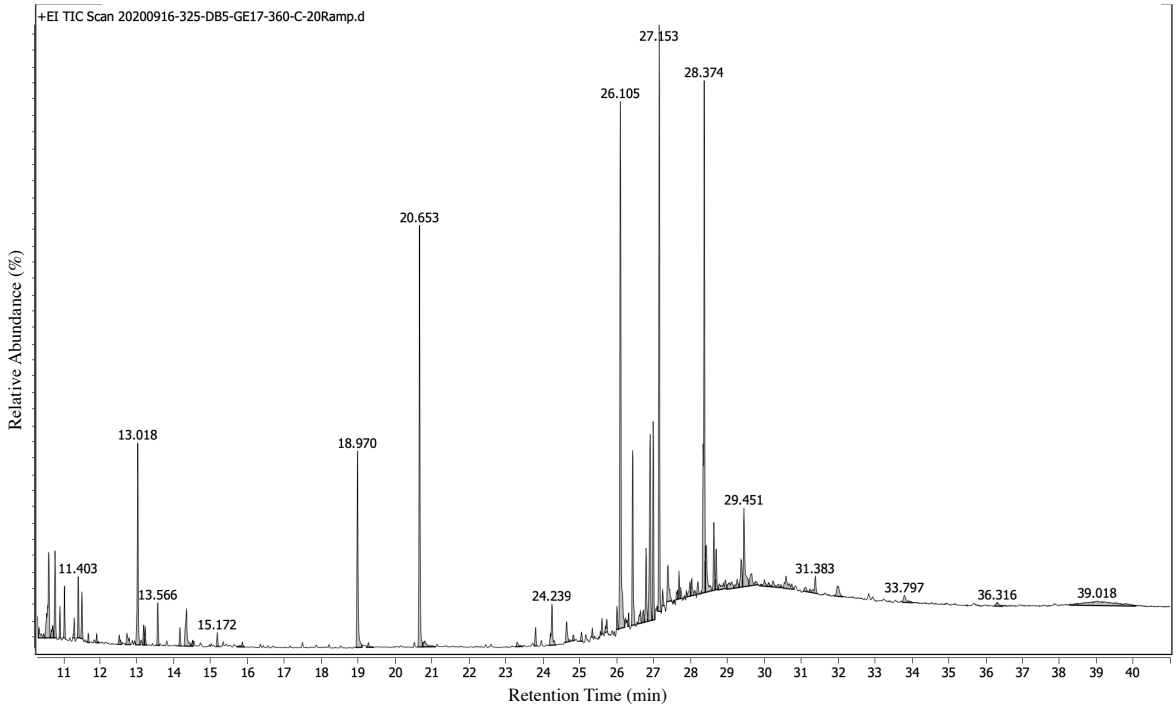


Figure 3.19. Chromatogram of sample GE17-360 from Gerace, Sicily on Agilent© DB-5ms column and GC-MS method D (20°C/min ramp to a maximum temperature of 325°C). Peak assignments can be seen in appendix X.

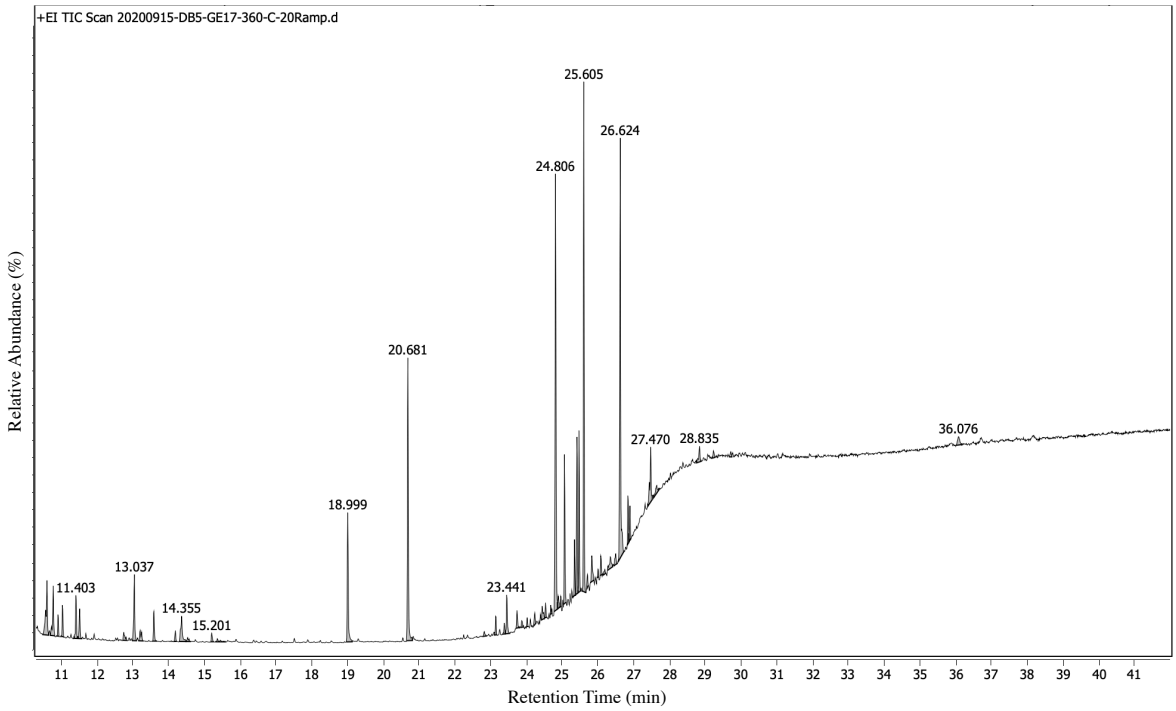


Figure 3.20. Chromatogram of sample GE17-360 from Gerace, Sicily on Agilent© DB-5ms column and GC-MS method C (20°C/min ramp to a maximum temperature of 350°C). Peak assignments can be seen in appendix X.

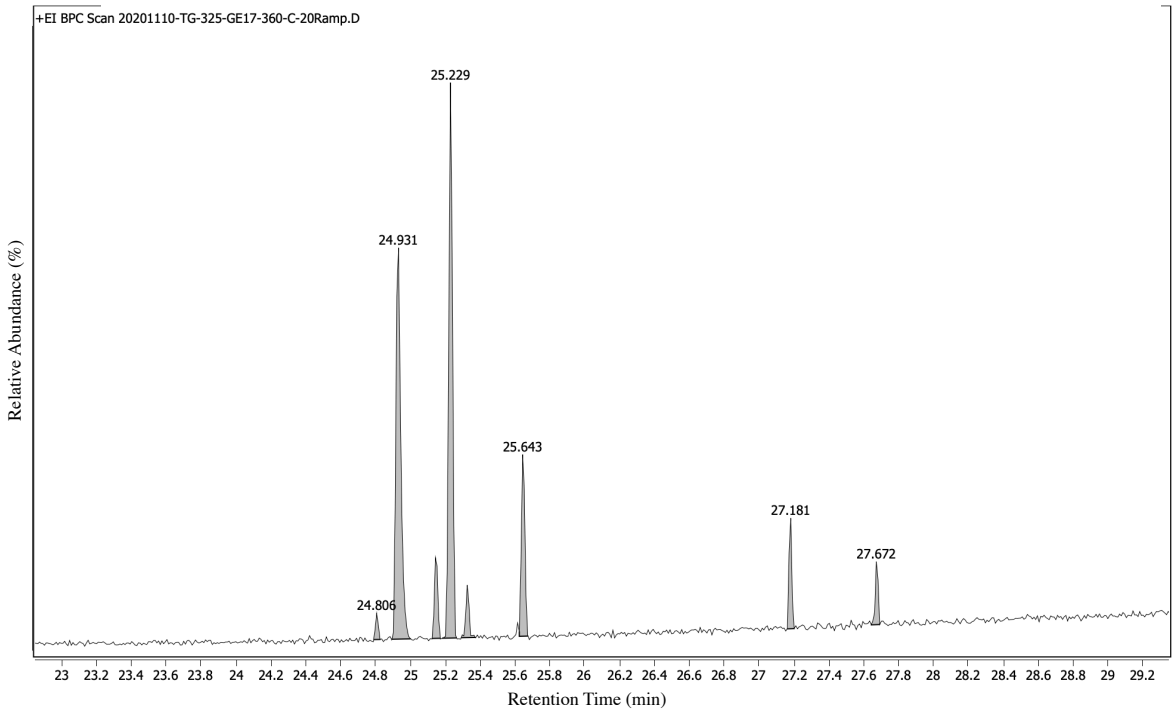


Figure 3.21. Chromatogram of sample GE17-360 from Gerace, Sicily showing diterpenoids on Restek© Rtxi-65TG column and GC-MS method D (20°C/min ramp to a maximum temperature of 325°C). Peak assignments can be seen in appendix X.

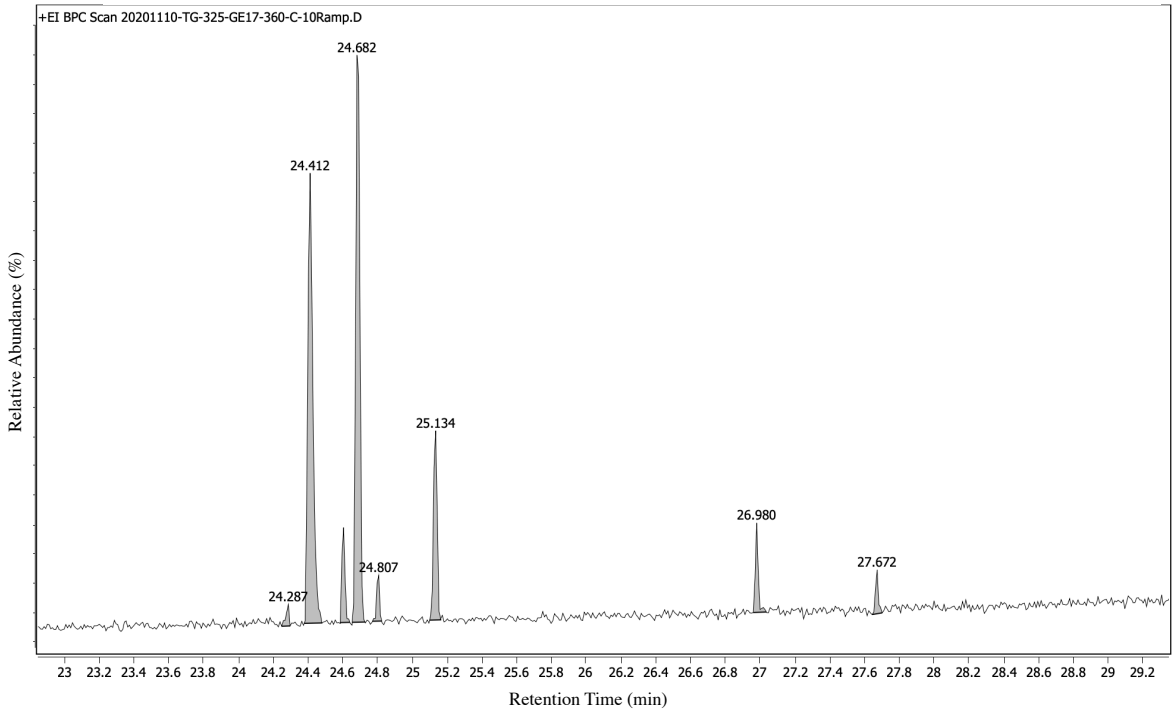


Figure 3.22. Chromatogram of sample GE17-360 from Gerace, Sicily showing diterpenoids on Restek© Rtxi-65TG column and GC-MS method B (10°C/min ramp to a maximum temperature of 325°C). Peak assignments can be seen in appendix X.

Overall results allow me to conclude that the best GC-MS method utilises the cool-on-column inlet at higher temperatures that include the Rtxi-65TG with GC-MS method A (10°C/min ramp to a maximum temperature of 350°C). This method was chosen to be most efficient based on quick elution, peak shape and resolution, and signal to noise ratio as described above. The method run time can be decreased with an increase in temperature, however, the concern of co-elution of compounds outweighs these benefits. Additionally, this method was found to be efficient for the majority of compounds often included in archaeological residues.

3.1.4 Testing GC-MS methods on samples from Houtaomuga

Samples from northeast China analysed for this thesis were subjected to two gas chromatographic methods. The first is a novel cool-on-column method A (10°C/min ramp to a maximum temperature of 350°C) as outlined above employing a crossbond diphenyl dimethyl polysiloxane stationary phase (Restek© Rtxi-65TG, 30m x 0.25mm ID x 0.1 film) and the second being a prototypical method using a split splitless injection and a 5%-phenyl-methylpolysiloxane stationary phase (Agilent© HP-5 column, 30m x 0.25mm ID x 0.25 film thickness) at lower temperatures. The goal of this duplicate analysis is to evaluate the efficiency and applicability of the novel method to archaeological assemblages by comparison.

Results show the overall separation of a variety of lipid classes in both procedures. An example of this application on the same sample from Houtaomuga (G18-2-S19) can be seen in figure 3.23 with the HP5-ms column and figure 3.23 with the Rtxi-65TG column. Overall retention times are significantly decreased with the Rtxi-65TG column, and this result is expected considering experimental results. Some compounds (i.e., $C_{12:0}$ and $C_{14:0}$) which are present in low abundances are not present in the Rtxi-65TG chromatogram but are present in the HP5-ms chromatogram (figure 3.23 and 3.24). A selection of samples from northeast China contains a significantly low abundance of free fatty acids including palmitic and stearic acids – the most sought-after components of isotopic characterisation by GC-C-IRMS. These low abundance compounds show poor elution on the Rtxi-65TG column. Future work will include a series of limit of detection (LOD) tests to determine if there are limitations to this column's ability to work with highly degraded small compounds. This would be a particular concern for future archaeological applications, particularly for assemblages with low lipid yields. This may still be useful for other art and heritage applications whose targets tend to have better preservation conditions.

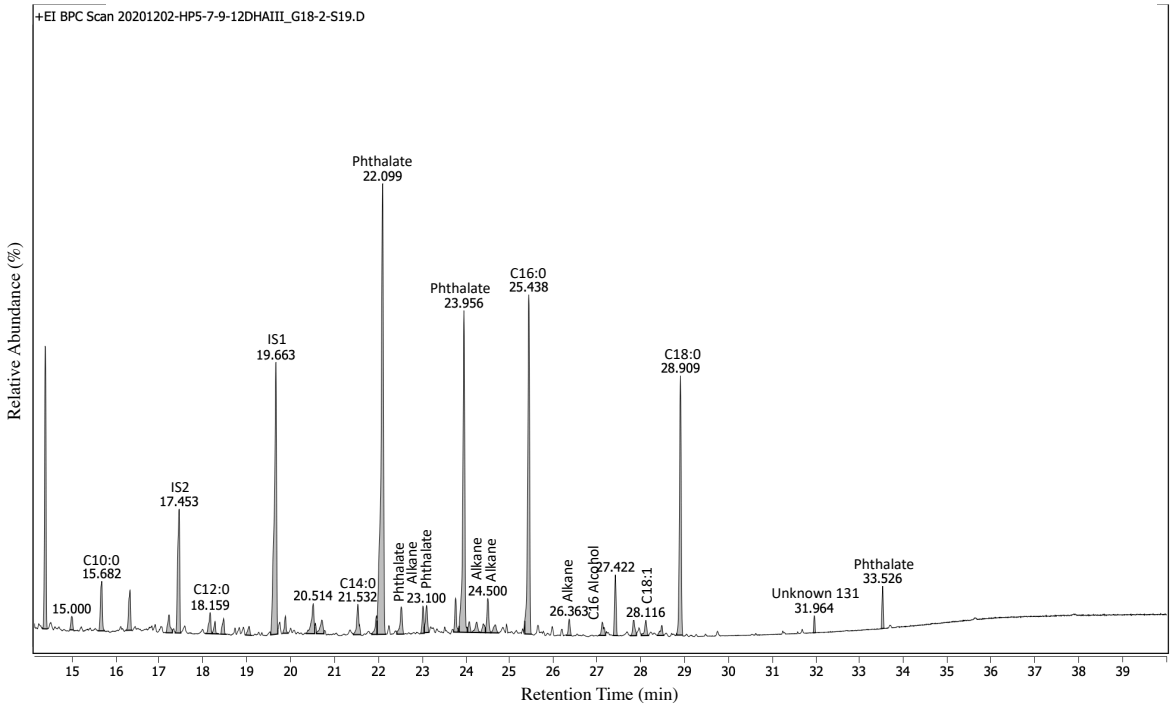


Figure 3.23. Chromatogram of sample G18-2-S19 from Houtaomuga in northeast China on Agilent© HP-5ms column with split/splitless injection with a maximum temperature of 300°C

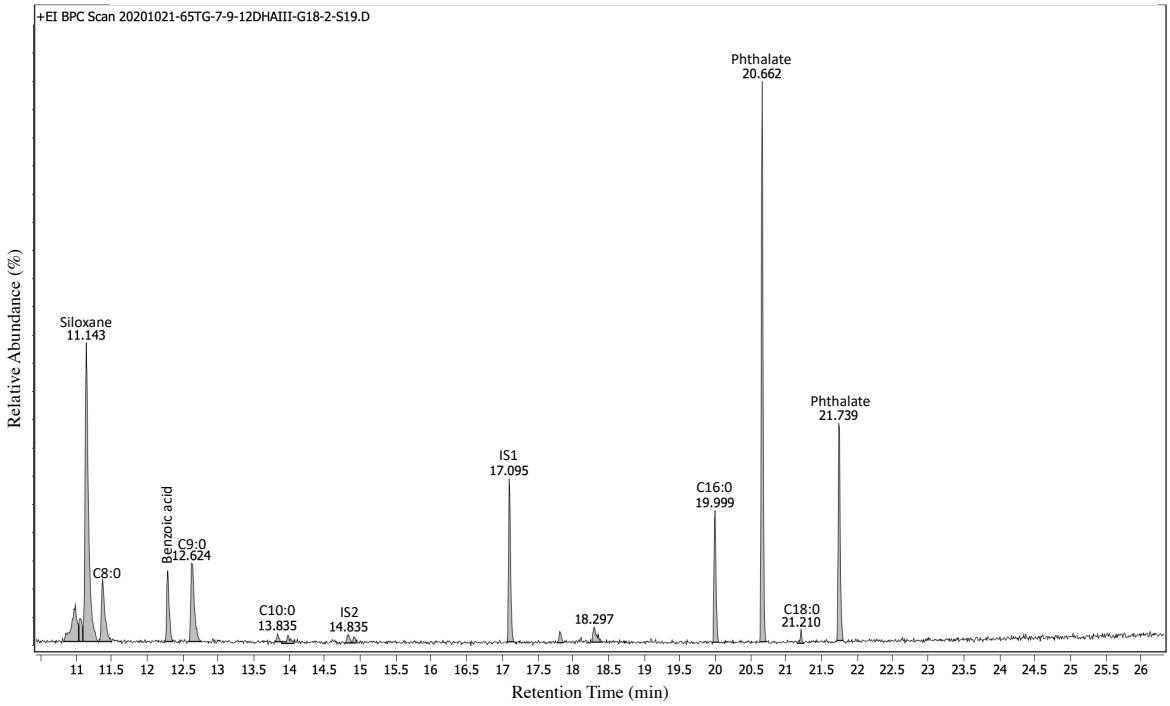


Figure 3.24. Chromatogram of sample G18-2-S19 from Houtaomuga in northeast China on Restek© Rtxi-65TG column with cool-on-column injection at a 10°C/min ramp with a maximum temperature of 350°C

3.1.5 Conclusions

Before SFE on samples from northeast China, the protocol for lipid extraction by SFE was optimised and refined using reference materials followed by extensive testing on archaeological materials. This was required due to the novelty of the protocol and to determine if the protocol was a suitable choice for the proposed research questions in this thesis regarding pottery use during the Neolithic in northeast China. Results from experimental materials as well as those from archaeological contexts showed that this SFE procedure is comparable to currently established methods using liquid solvents and total extraction of a wide variety of compounds commonly found in archaeological pottery.

Methods for molecular analysis were also optimised and tested as a part of this research. Testing of various stationary phases and GC-MS methods on a cool-on-column injector port exhibited various benefits for a variety of compound types often identified in archaeological contexts. For archaeological samples in northeast China, GC-MS method A proved to be the most opportune method for identifying target compounds. An aliquot of extracts from pottery samples from northeast China was subject to this on-column method and an established method on a split/splitless injection port for comparison. The main goal of this analysis was to better identify large compounds which normally have difficulty eluting using established methods.

Samples from northeast China did not contain any such molecules and the comparison between these methods will not be discussed in detail here. Archaeological lipid chromatography is often untargeted, and we do not know the contents of a sample until it is analysed, making this result a possibility.

3.2 Results of Lipid Residue Analysis on Pottery from Houtaomuga

3.2.1 Houtaomuga GC-MS Results

Lipid extracts from ceramic sherds from northeast China that underwent SFE were characterised and quantified by GC-MS (appendix I; table 3.3, 3.6). The majority of samples analysed yielded a series of short and mid-length even chain monocarboxylic acids, notably $C_{12:0}$, $C_{14:0}$, $C_{16:0}$, $C_{18:0}$ (Table 3.4). While these compounds are not indicative of any specific organic source, they likely derive from a series of plant oils based on high abundances of $C_{16:0}$ and the absence of odd-chain monocarboxylic acids (Evershed et al., 1999; Regert, 2011). Samples contain unsaturated monocarboxylic acids ($C_{16:1}$, $C_{18:1}$, $C_{22:1}$) and glycerol. All these molecules are characteristic of degraded fatty substances from human or plant origin. Several samples also contain saturated odd-chain monocarboxylic acids ($C_{15:0}$ and $C_{17:0}$), suggesting the presence of animal fats, however, this is uncommon in the assemblage as a whole. A summary of these results by feature can be seen in table 3.3 and quantities of compounds identified for each sample can be seen in appendix IV.

Most samples contained polycyclic aromatic hydrocarbons (PAH) and other hydrocarbons indicative of burning. These confirm what has been anticipated based on the existing literature (Wang and Sebillaud, 2019), meaning that these pots were heated with fire. Additionally, samples contain dehydroabietic acid, which can be a by-product of wood combustion (Reber et al., 2018). While many of these pots contain ash deposits (sooting) on their exteriors, a common sign of cooking, the possibility of these compounds being introduced during the firing process cannot be ruled out. Similar results were obtained from modern unused low fired experimental pottery by Reber et al. (2018), showing these compounds occur as by-products of the firing process with this wood is used. These compounds could be introduced to pottery through soil contamination, however, the outside 1mm of ceramic sherd was removed prior to extraction and this is unlikely.

Table 3.4. Summary of organic compounds identified using GC-MS in each feature examined from Houtaomuga in this study. Other n-alkanoic acids include but are not limited to compounds such as $C_{15:0}$ and $C_{17:0}$. Diterpenoids often include oxidized abietanes as described in this section. Organics are mostly homogenous between features and similar throughout the assemblage as a whole.

Feature	Phase	Glycerol	Diterpenoids	Hydrocarbons	Palmitic acid	Stearic acid	Other n-alkanoic acids	Long Chain Alcohols	Squalene	Aromatic Compounds	Phthalates	Chemical Sunscreen	DEET
G18-1	1			X	X	X	X			X	X		
G18-2	1	X	X	X	X	X	X	X	X		X	X	
G21-1	1		X	X	X	X	X				X		
G22	1			X	X	X	X				X	X	
H146	1		X	X	X	X	X				X		
H248-1	1		X	X	X	X	X				X	X	X
G002	2		X		X	X	X				X		
G013	2		X	X	X	X	X	X		X	X		
H131	2				X	X	X				X		
G001	3		X	X	X	X	X	X		X	X		
F008-1	4		X	X	X	X	X				X		
F010-1	4		X	X	X	X	X				X		
F013	4				X	X	X				X		
F016-1	4				X	X	X				X		
F016-2	4				X	X	X				X		
F016-3	4				X	X	X				X	X	
F017-D1-2	4				X	X	X				X		
F018-2	4				X	X	X				X	X	X
F021-2	4				X	X	X				X		
F023-2	4				X	X	X				X		
H085	4		X	X	X	X	X				X		

Diterpenoids were identified in extracts from 10 features (table 3.4). These mainly consist of dehydroabietic acid (DHA) and 7-oxo-DHA. These oxidized abietanes are indicative of exudates from *Pinaceae* species (Colombini et al., 2005; Mills and White, 1987). With the absence of additional compounds with abietane structures, the presence of these compounds alone likely derives from pottery use or production (i.e., wood smoke from firing or cooking), and may not be due to the presence of resinous exudates (Reber et al., 2018). It is highly likely these compounds derive from the production process in this context as they are fired at low temperatures and possibly in open-air contexts or from direct cooking over wood fuel from the *Pinaceae* family. This is further supported by the additional presence of polycyclic aromatic

hydrocarbons (PAH) in many of these samples. Such compounds are indicative of wood combustion (Pimentaa et al., 1998).

A small number of samples contain squalene (TMS ester). Squalene is a triterpene that is a major component in plant essential oils and a precursor to cholesterol, often linked to contamination of handling during and following excavation (Malainey, 2010). This compound was not identified in any laboratory blanks (detailed in appendix IX). Samples in this examination also contain a series of n-alkanes and hydrocarbons. In certain contexts and in the presence of additional organic components, these compounds can be indicative of petroleum products, coal, certain waxes, or bituminous material, however, these characteristics are not present (Mills and White, 1987). These compounds are instead interpreted as evidence for contamination from the burial environment or burning, including the use of the pot over an open flame or the firing process. Other compounds identified include aromatic compounds, mainly in the form of benzoic acid and glycerol (TMS derivatives), a structural component in glycerides.

3.2.2 Resinous Substances

The full suite of compounds with abietic structures as described in section 3.2.1 along with polycyclic aromatic hydrocarbons, specifically, retene and pyrene, are indicative of resin deriving from trees in the *Pinaceae* family. These types of resins are common in archaeological contexts across the globe. One sample (G13-S1, Houtaomuga) does exhibit this full suite of biomarkers indicative of a resin from *Pinaceae* tree exudate, and a chromatogram showing these compounds is pictured below (figure 3.25). It is unclear if these biomarkers are evidence for such a resin or glue in this context. No visible resin in the form of a waterproofing surface treatment is present on the samples in question, but this does not negate the possibility of its existence due to degradation. Evidence for resin has not been identified in the area previously, however, it was commonly used in the past and by modern people as a hafting agent or glue and this result is unsurprising (Kozowyk et al., 2020; Regert, 2004). It has also been used as a waterproofing agent and may have initially lined the inside surface of the vessel to serve this purpose (Colombini et al., 2003; La Nasa et al., 2020).

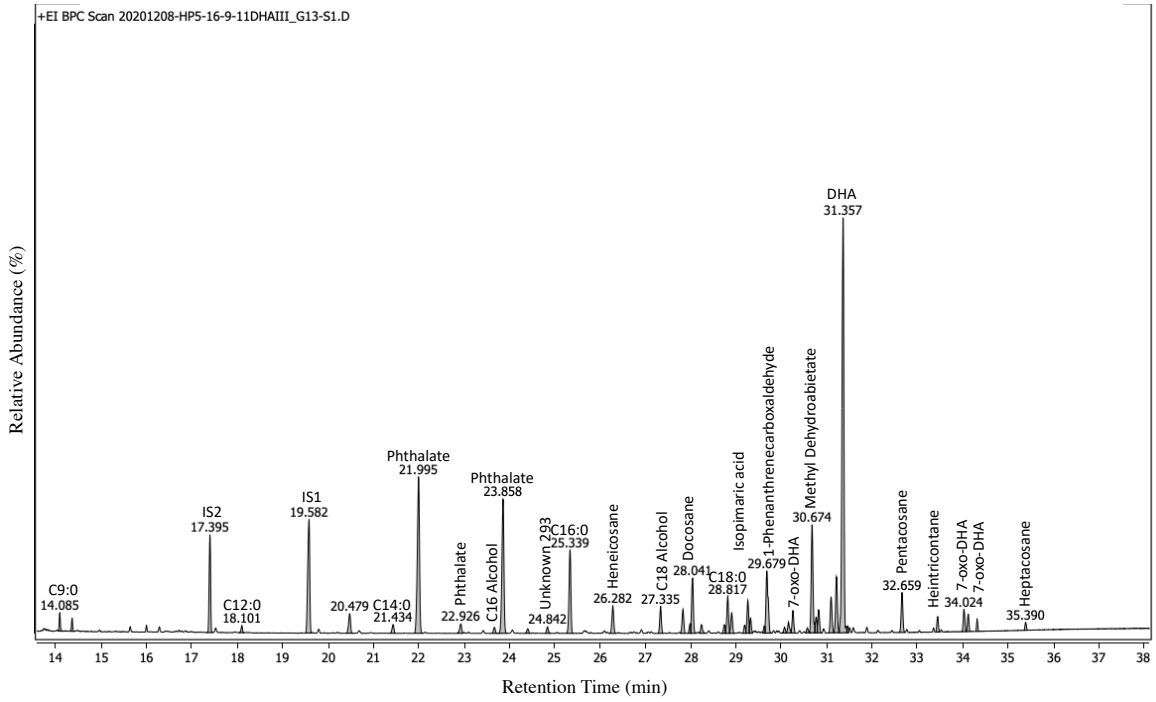


Figure 3.25. Chromatogram of sample G13-S1 from the site of Houtaomuga showing possible resin deriving from trees in the *Pinaceae* family. Such resin is commonly used as a hafting or waterproofing agent by hunter-gatherer populations.

3.2.3 Houtaomuga GC-C-IRMS Results

GC-MS analysis of pottery from Houtaomuga as described above primarily included carboxylic acids of unspecific origin. To further characterize these extracts to better understand the originating source of these compounds, GC-C-IRMS was undertaken (n=94; single compound peak abundance for palmitic and stearic acid >5000). Results of this IRMS analysis of samples with adequate lipid preservation (peak abundance >1000; n=64) exhibit exceedingly depleted values for $\delta^{13}C_{16:00}$ and $\delta^{13}C_{18:00}$ (-33.3 to -25.1; table 3.5, figure 5.1) consistent with food commodities primarily including C_3 plants and freshwater resources (figure 5.1 and 5.2) (Craig et al., 2007). Overall, the distribution of values for $\delta^{13}C_{16:00}$ and $\delta^{13}C_{18:00}$ fall within a narrow range implying pottery at Houtaomuga had specific limited uses and was not used for an exceedingly large range of product processing. The increased depletion of $\delta^{13}C_{16:00}$ values in these samples strongly suggest resource mixing with predominantly C_3 plants (figure 4.2) (Craig et al., 2013, 2004; Lucquin et al., 2018). The $\Delta^{13}C$ ($\delta^{13}C_{18:00}$ - $\delta^{13}C_{16:00}$) proxy was calculated to further distinguish between the origin of wild ruminant, non-ruminant, and freshwater fats (Craig et al., 2007; Cramp and Evershed, 2013; Lucquin et al., 2016). The results cluster between -0.8 to 2.1‰ $\Delta^{13}C$ which falls in the range established for non-ruminant and aquatic fat resources. This result is consistent with faunal assemblages at the site which

shows extensive exploitation of freshwater resources including clams, molluscs, and freshwater fish (Song et al., 2016; Wang et al., 2016). The results of this analysis do not show the exploitation of C_4 plants such as millet and terrestrial animals in pottery. They also do not show increased variability between features or between occupational phases, and the narrow range of isotopic values at Houtaomuga remains consistent over time.

Table 3.5. Results of GC-C-IRMS analysis from all samples from four phases from the site of Houtaomuga

Field Number	Phase	$\delta^{13}C_{16:0}$	$\delta^{13}C_{18:0}$	$\Delta^{13}C$	Standard Deviation $\delta^{13}C_{16:0}$	Standard Deviation $\delta^{13}C_{18:0}$
12 DHA III G18 (1)_S3	Phase 1	-29.2	-28.6	0.6	0.019	0.02
12 DHA III G18 (1)_S4	Phase 1	-29.8	-27.7	2.1	0.0395	0.189
12 DHA III G18 (1)_S5	Phase 1	-30.6	-29.8	0.9	0.0495	0.045
12 DHA III G18 (1)_S6	Phase 1	-31.1	-29.9	1.2	0.0625	0.0405
12 DHA III G18 (1)_S8	Phase 1	-31.2	-29.7	1.5	0.0255	0.0155
12 DHA III G18 (1)_S9	Phase 1	-30.6	-29.5	1.2	0.0735	0.0655
12 DHA III G18 (2)_S3	Phase 1	-30.4	-29.9	0.5	0.022	0.025
12 DHA III G18 (2)_S4	Phase 1	-30.0	-29.2	0.8	0.1835	0.048
12 DHA III G18 (2)_S6	Phase 1	-29.5	-28.8	0.8	0.0125	0.0425
12 DHA III G18 (2)_S7	Phase 1	-30.0	-29.4	0.7	0.0255	0.015
12 DHA III G18 (2)_S9	Phase 1	-29.9	-29.0	0.9	0.0425	0.0225
12 DHA III G18 (2)_S10	Phase 1	-29.8	-29.1	0.7	0.091	0.003
12 DHA III G21 (1)_S1	Phase 1	-31.3	-29.5	1.8	0.096	0.397
12 DHA III G21 (1)_S3	Phase 1	-30.9	-30.0	1.0	0.03	0.0445
12 DHA III G21 (1)_S4	Phase 1	-31.0	-29.8	1.2	0.0665	0.081
12 DHA III G21 (1)_S5	Phase 1	-29.3	-29.2	0.1	0.107	0.0335
12 DHA III G22 (1)_S2	Phase 1	-29.3	-28.6	0.8	0.0635	0.0705
12 DHA III G22 (1)_S3	Phase 1	-30.6	-30.0	0.6	0.3615	0.069
12 DHA III G22 (1)_S5	Phase 1	-30.4	-29.5	0.9	0.067	0.213
12 DHA III G22 (1)_S8	Phase 1	-29.2	-29.0	0.2	0.4475	0.595
12 DHA III G22 (1)_S9	Phase 1	-31.4	-30.3	1.2	0.0645	0.0265
12 DHA III G22 (1)_S10	Phase 1	-30.4	-30.1	0.4	0.1145	0.045
12 DHA III H146 (1)_S2	Phase 1	-30.2	-29.2	1.0	0.106	0.6
12 DHA III H146 (1)_S3	Phase 1	-29.9	-29.1	0.8	0.013	0.3525
12 DHA III H146 (1)_S4	Phase 1	-30.1	-29.8	0.3	0.0135	0.0305
12 DHA III H146 (1)_S5	Phase 1	-30.3	-30.1	0.2	0.13	0.3745
12 DHA III H248 (1)_S1	Phase 1	-29.2	-28.5	0.7	0.037	0.046
12 DHA III H248 (1)_S2	Phase 1	-28.3	-27.4	0.8	0.2775	0.423
12 DHA III H248 (1)_S3	Phase 1	-25.4	-25.1	0.3	0.09	0.06
12 DHA III H248 (1)_S4	Phase 1	-28.3	-27.3	1.0	0.077	0.2325
12 DHA III H248 (1)_S5	Phase 1	-28.1	-27.9	0.2	0.0305	0.0195
11 DHA III G002 (1)_S1	Phase 2	-31.1	-30.3	0.7	0.0065	0.0275
11 DHA III G002 (1)_S2	Phase 2	-30.3	-30.9	-0.6	0.0365	0.0915
11 DHA III G002 (1)_S3	Phase 2	-30.4	-29.4	1.0	0.0395	0.049
11 DHA III G002 (1)_S4	Phase 2	-29.5	-29.0	0.5	0.0255	0.033
11 DHA III G002 (1)_S5	Phase 2	-29.4	-29.6	-0.2	0.0245	0.002
11 DHA III G002 (1)_S6	Phase 2	-29.9	-29.5	0.3	0.0025	0.0255
11 DHA III G002 (1)_S8	Phase 2	-31.3	-31.9	-0.7	0.034	0.033
11 DHA III G002 (1)_S10	Phase 2	-29.5	-29.4	0.1	0.0465	0.0935
11 DHA III G13_S1	Phase 2	-31.0	-30.1	1.0	0.0605	0.011
11 DHA III H131 (1)_S1	Phase 2	-29.9	-29.8	0.1	0.2215	0.0405
11 DHA III H131 (1)_S2	Phase 2	-29.6	-29.6	0.0	0.0115	0.048
11 DHA III H131 (1)_S3	Phase 2	-29.5	-29.9	-0.4	0.131	0.046
11 DHA III H131 (1)_S6	Phase 2	-30.3	-30.5	-0.1	0.1285	0.0895
11 DHA III H131 (1)_S8	Phase 2	-29.1	-29.4	-0.3	0.0645	0.0385
11 DHA III G001 (1)_S1	Phase 3	-29.5	-28.6	1.0	0.024	0.0285
11 DHA III G001 (1)_S2	Phase 3	-29.9	-33.3	-3.5	0.023	2.9675
11 DHA III G001 (1)_S7	Phase 3	-30.0	-30.3	-0.3	0.002	0.0585
11 DHA III G001 (1)_S10	Phase 3	-29.8	-29.9	-0.1	0.1015	0.0015
13 DHA I F008 (1)_S3	Phase 4	-30.4	-31.2	-0.8	0.001	0.643
13 DHA I F008 (1)_S4	Phase 4	-30.4	-30.4	0.1	0.0575	0.1325
14 DHA I F010 (1)_S1	Phase 4	-30.2	-30.3	-0.1	0.0415	0.022
14 DHA I F010 (1)_S2	Phase 4	-29.6	-29.2	0.4	0.1075	0.046
14 DHA I F010 (1)_S3	Phase 4	-30.1	-30.3	-0.2	0.0065	0.031
14 DHA I F013 (1)_S1	Phase 4	-30.1	-30.2	-0.1	0.095	0.1095
14 DHA I F013 (1)_S3	Phase 4	-30.4	-30.2	0.3	0.0415	0.147
14 DHA I F013 (1)_S4	Phase 4	-30.4	-29.5	0.9	0.0785	0.0225
14 DHA I F017 D1 (2)_S1	Phase 4	-29.7	-29.4	0.3	0.071	0.033
14 DHA I F017 D1 (2)_S2	Phase 4	-30.6	-30.2	0.4	0.181	0.038
14 DHA I F017 D1 (2)_S4	Phase 4	-30.5	-30.5	0.0	0.3185	0.018
15 DHA I F021 (2)_S1	Phase 4	-30.3	-30.1	0.2	0.0455	0.0225
11 DHA III H085(1)_S3	Phase 4	-30.8	-30.6	0.1	0.1815	0.1715
11 DHA III H085(1)_S4	Phase 4	-30.5	-30.6	-0.2	0.0475	0.137

3.2.4 Preservation

Samples selected for this thesis from the site of Houtaomuga are exceedingly old and the degree of lipid preservation was not well understood before undertaking this work. Results of surviving lipid concentrations can be seen in table 3.6. This table shows that on average samples from Phase 1, the earliest occupational phase, has the best lipid preservation. This is the opposite of expected due to the significant age of the material, however, most pottery samples from Phase 1 were not washed, while pottery from later phases had been washed and scrubbed with toothbrushes post-excavation. This seems to have had a significant influence on lipid preservation, and future work should avoid typical cleaning before scientific analysis. Azelaic acid, a saturated dicarboxylic acid consisting of a 9-carbon chain, is a common degradation product of long-chain unsaturated monocarboxylic acids (Colombini and Modugno, 2009). This is due to the point of unsaturation commonly occurring at the 9th carbon. This compound is not present in the majority of samples, leaving some open questions about their preservation. Mainly they are likely not heavily degraded, further supporting the hypothesis that post-excavation practices have heavily influenced preservation. The higher lipid preservation rate in Phase 1 might be due to additional factors. Pottery from this occupational phase could also have been more porous than those of later phases, allowing for lipid material to remain in the ceramic body for longer periods. Lipids absorbed in the ceramic body of sherds are better protected from outside elements including the burial environment.

Table 3.6. Total lipid concentration for each Phase analysed in this thesis from the site of Houtaomuga. Lipid concentration for each sample appendix IV. Quantitation methods are described in appendix IX.

Phase	Average lipid concentration (µg/g)
Phase 1	2292.7
Phase 2	1352.2
Phase 3	733.5
Phase 4	375.5

Twenty-seven samples from Houtaomuga did not yield any lipid compounds and have been excluded from calculations in table 3.5. The majority of these samples with no lipid preservation come from Phase 4 (5,000 cal BP). This result could suggest a change in pottery use between each phase which could result in decreased lipid retention within the ceramic, e.g. storing rather than cooking food, or processing foods with lower lipid concentrations (i.e.

plants) on the whole. However, results of IRMS analyses (discussion section 4.3) show that pottery use between phases is similar, making this issue likely the result of preservation rather than use. I believe this result further supports the hypothesis that the pottery that was extensively cleaned lost significant lipid material in the process, the majority of these samples being from Phase 4. Alternatively, this decrease in lipid yield could be caused by the firing temperature of the ceramic which is higher in Phase 4 than Phase 1 and Phase 2, meaning these earlier ceramics are could be more porous and absorb more lipid residue.

With the higher lipid preservation in Phase 1 and 2, samples selected for stable isotope analyses skew more towards these phases, and samples that were successful for IRMS analyses largely come from these phases. Thirty samples selected for IRMS analysis did not exhibit adequate carbon preservation for analysis and these samples are noted in Appendix V. Over 88% (31/35) of samples from Phase 1 and over 76% (13/17) of samples from Phase 2 analysed using IRMS had adequate lipid preservation for analysis. Respectively, 40% (4/10) of samples from Phase 3 and 47% (14/30) of samples from Phase 4 generated isotopic data. These preservation rates are similar to GC-MS results regarding preservation, and either post-excavation techniques or pottery porousness is altering the rate of lipid preservation between Phases 1 and 2 and Phases 3 and 4. Future work at the site, and possibly the region should consider this when selecting samples for IRMS analysis.

3.2.5 Contaminants

Contaminants are present in all samples analysed and are not present in internal lab blanks (as described in appendix IX). Primarily, these include plasticizers in the form of phthalates. These are not unanticipated, considering the samples have been stored in plastic bags after excavation for curation. Additional contaminants are not abundant or in high concentrations but include DEET and chemical sunscreens, likely from excavation handling. All contaminants identified are common in archaeological contexts and are quantified for each sample in appendix IV based on calculations described in appendix IX.

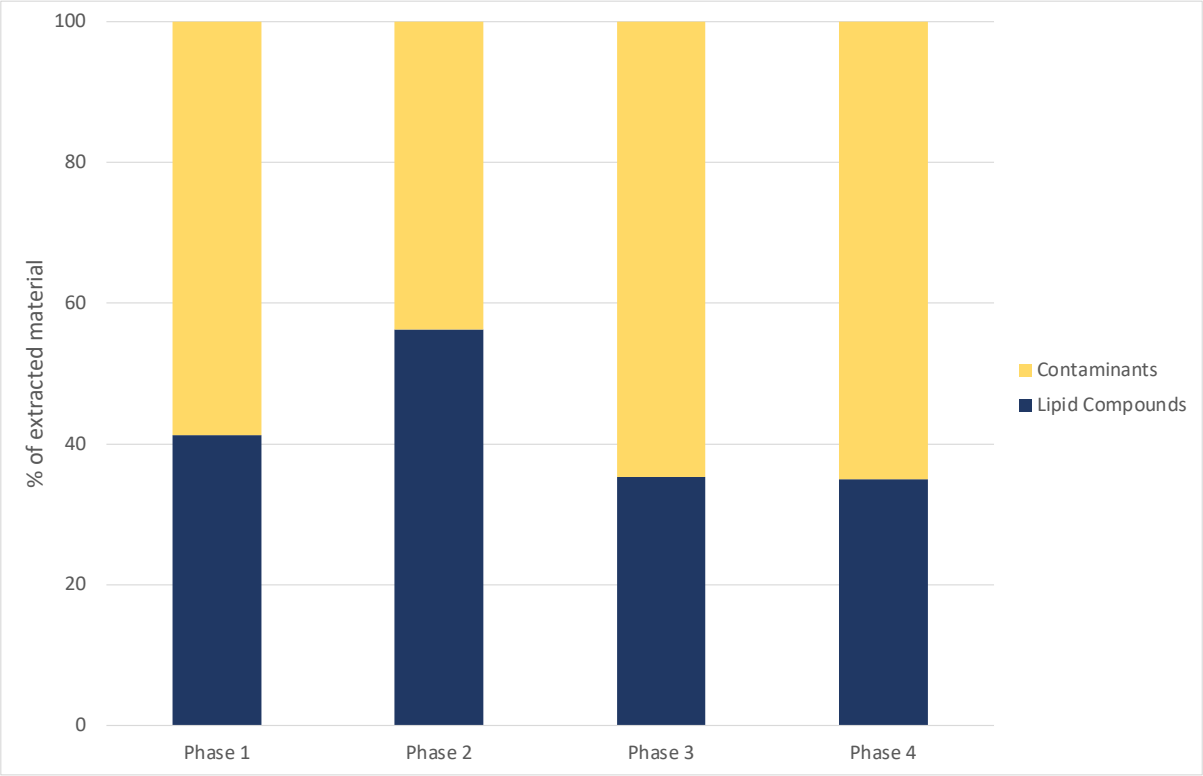


Figure 3.26. The proportion of compounds to contaminants extracted from pottery samples by Phase. This figure is showing that post-excavation practices which were different between each Phase has less effect on introducing contaminants than post-excavation practices which remained consistent across each Phase, primarily the storage of sherds in plastic bags.

The proportion of contaminants to lipid compounds for each phase are shown in figure 3.26. These proportions are not unanticipated, and overall consistent across all phases. As mentioned in section 3.2.4, samples between phases were subject to different post-excavation procedures and samples from phases 1 and 2 were unwashed while samples from phases 3 and 4 were washed and scrubbed. Figure 3.26 suggests post-excavation washing is not the main cause of contaminant introduction as the ratio between contaminants and compounds is similar across all phase groups. Instead, this likely supports the notion that plastic bag storage is the main factor contributing to contaminants in lipid extracts. Future work should avoid this practice.

3.3 Summary

GC-MS results from Houtaomuga did not produce evidence for compounds previously found in pottery residues in northeast Asia, including evidence for fish exploitation in the form of polyunsaturated carboxylic acids and ω-(o-alkylphenyl)alkanoic acids (APAA's) evidence for millet in the form of the biomarker miliacin which could have been present in the later phase

ceramics. These compound classes in question are highly susceptible to degradation, particularly polyunsaturated lipids deriving from fish, and many researchers, therefore, preferer to test for them via GC-C-IRMS. The results of supercritical fluid extraction followed by GC-MS analysis and GC-C-IRMS analysis indicate that this was the main source of fats in the vessels along with C_3 plants. These aquatic products likely contain high amounts of shellfish rather than large carnivorous fish. Samples from all four phases are highly similar in their isotopic characterisation, meaning pottery use was not highly diverse within the site. Over time, pottery use becomes more specific as values in Phase 1 show the largest distribution of values.

Contaminants were present in most GC-MS samples, and these are expected as samples were handled by excavators and stored in plastic bags. Results of GC-MS analysis shows the majority of samples analysed yielded a series of compounds likely of plant origin. Samples also contain contained multiple compounds indicative of burning, likely from the production process or from heating the vessels for cooking. These results likely indicate that the hypothesis that these vessels were used for cooking as opposed to storage is correct. One sample possibly shows evidence of a resin deriving from a tree in the *Pinaceae* family which could have been used as a waterproof seal on the vessel interior or as a glue or hafting agent. Stone tools at Houtaomuga include microblades that were likely hafted into composite tools making this scenario plausible.

Chapter 4 Discussion of Archaeological Results

The site structure, settlement patterns, local environment as well as faunal and palaeobotanical assemblage at Houtaomuga have been outlined in Chapter 2. The implications of the results of pottery residue analysis in context with these findings are presented here along with an additional interpretation of the implications of results discussed in Chapter 3. It is obvious from these lines of evidence that the people who lived at Houtaomuga had very diverse diets and a mixed economy during all phases of occupation (Luan, 2016; Song et al., 2016; Wang et al., 2016). What this section aims to better understand is what part of this diverse diet involved pottery use and how that use changed over time and space within the site. Additional non-food related pottery uses will also be considered.

4.1 Changes over time

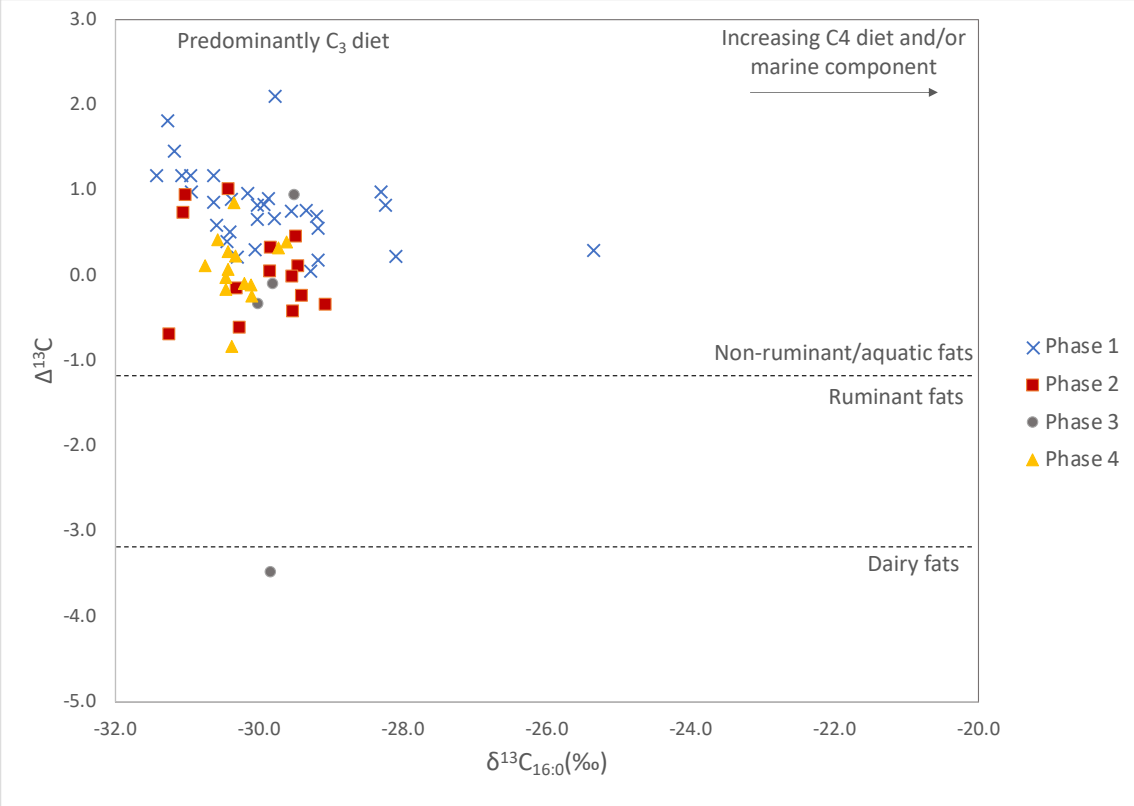


Figure 4.1. Plot of $\Delta^{13}C$ ($\delta^{13}C_{18:00}$ - $\delta^{13}C_{16:00}$) values against $\delta^{13}C_{16:00}$ values from the four Neolithic phases from the site of Houtaomuga allowing for the distinction between various animal fats.

Phase 1 exhibits the widest distribution of $\Delta^{13}C$ values as well as $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ monocarboxylic acids compared to later phases (figure 4.1, 4.2). This may suggest a broader range of resources were exploited in the earlier phase and later phases show signs of narrowing subsistence practices. Phases 3 and 4 includes more samples that display decreasing $\Delta^{13}C$ values which may suggest the addition of more ruminant fats in addition to aquatic resources and C_3 plants. Nevertheless, these values still fall confidently within the aquatic resource range.

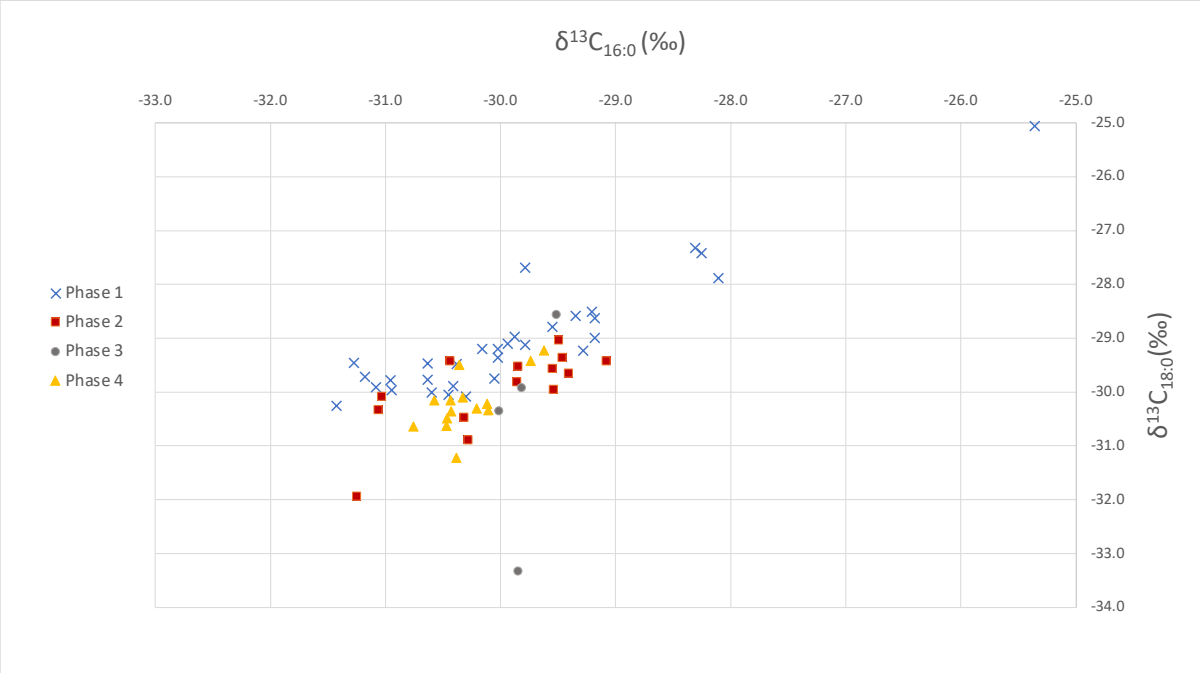


Figure 4.2. Figure 2. $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ monocarboxylic acids extracted from four Neolithic phases of pottery from the site of Houtaomuga

$\delta^{13}C$ values of $C_{18:0}$ are more depleted for samples from phases 2, 3, and 4 than those from Phase 1 (Figure 4.2). This can be interpreted as evidence for increased use of ruminant fats in later periods. Indeed, these samples all cluster primarily within the range for freshwater resources, meaning this was still the predominant resource group processed in the vessels over all phases. Four samples from Phase 1 are less depleted in $\delta^{13}C_{18:0,}$ however, this is still within the range for primarily freshwater resources. Three of these samples are also less depleted in $\delta^{13}C_{16:0,}$ suggesting these samples may be exhibiting additional processing of more C_4 plant

resources or other large carnivorous fish[1] which are present in the faunal assemblage (Song et al., 2016). Moreover, this continues to suggest slightly different pottery usage patterns within Phase 1 which may have been more variable than later phases.

One sample from Phase 3 (11 DHA III G001 (1) S2) exhibited a $\Delta^{13}C$ value of -3.5‰ and $\delta^{13}C_{16:00}$ value of -29.9 and $\delta^{13}C_{18:00}$ of -33.3 (figure 4.1 and 4.2, grey bullet point). Since there is no evidence for dairying practices and domesticated livestock during this period, this value is likely the result of increased ruminant fat processing that is further supported by the faunal assemblage which is terrestrial mammal dominant during Phase 3 (Song et al., 2016). Zhang (2015) argued that the site of Houtaomuga is a short term feasting site occupied in autumn, showing early evidence for human social complexity, largely circulating around intense short term seasonal auroch hunts (Zhang, 2015). This conclusion was based on evidence from Phase 3 and 4 refuse pits which primarily contain aurochs' bones. This isotopic result could support the notion that hunting practices increased during Phase 3 observed by Zhang (2015), yet other samples from Phase 3 exhibit values consistent with freshwater resource exploitation. This suggests that if used as a short-term hunting site, residents at Houtaomuga did not solely use the site for auroch hunts and subsistence practices were varied. Increased sample size from this phase specifically may provide further insight.

The distribution of values of $\delta^{13}C$ of $C_{16:0}$ and $C_{18:0}$ monocarboxylic acids between each phase are shown in Figures 4.3 and 4.4. These figures further highlight the narrowing of the distribution of these values between Phase 1 and Phase 4. As mentioned in results section 3.2.5, preservation of lipids is superior in Phases 1 and 2 samples over Phase 3 and 4 samples, and this could have resulted in samples from these earlier phases being more representative. Overall results from all four occupational phases have a narrow range of values, highlighting very specific pottery use over a long period of time. These narrow values suggest pottery use remains largely consistent over time while vessel form, decoration, and production change between each occupational period at the site.

[1] I frequently make the distinction between carnivorous fish and other fish in this chapter because the distinction is also often made by zooarchaeologists due to the size of the fish. Carnivorous fish are much larger than their herbivorous counterparts and catching them requires different fishing methods. Moreover, these large fish have different $\delta^{13}C$ values as can be seen in table 5.1.

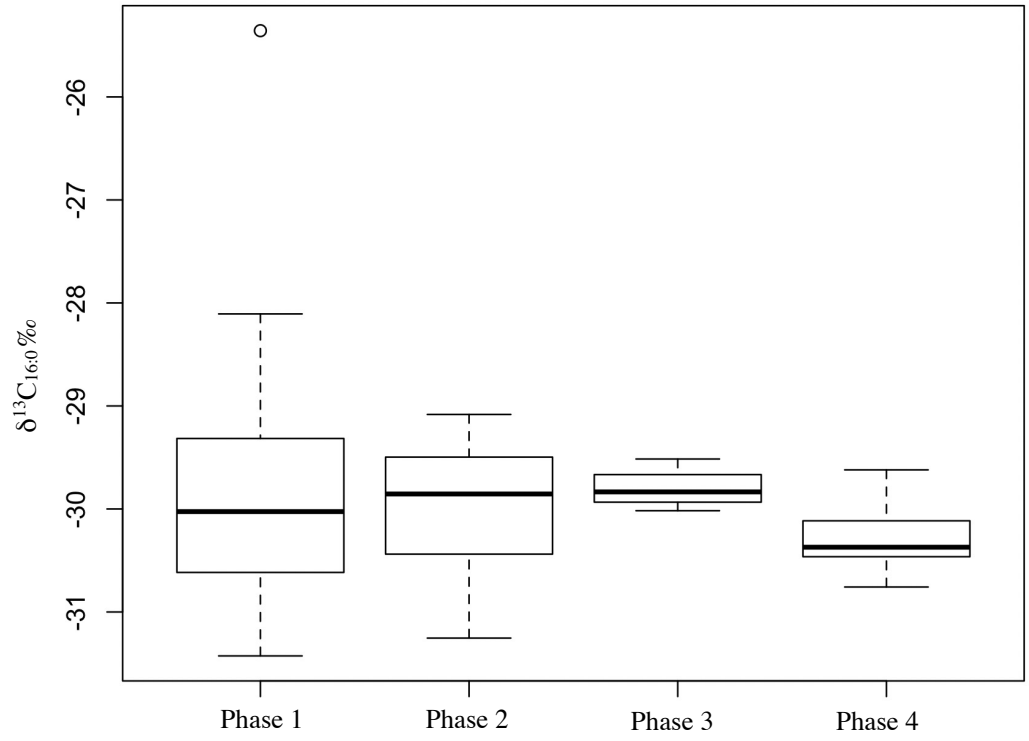


Figure 4.3. Distribution of $\delta^{13}C$ values of $C_{16:0}$ monocarboxylic acid extracted from four Neolithic phases of pottery from the site of Houtaomuga

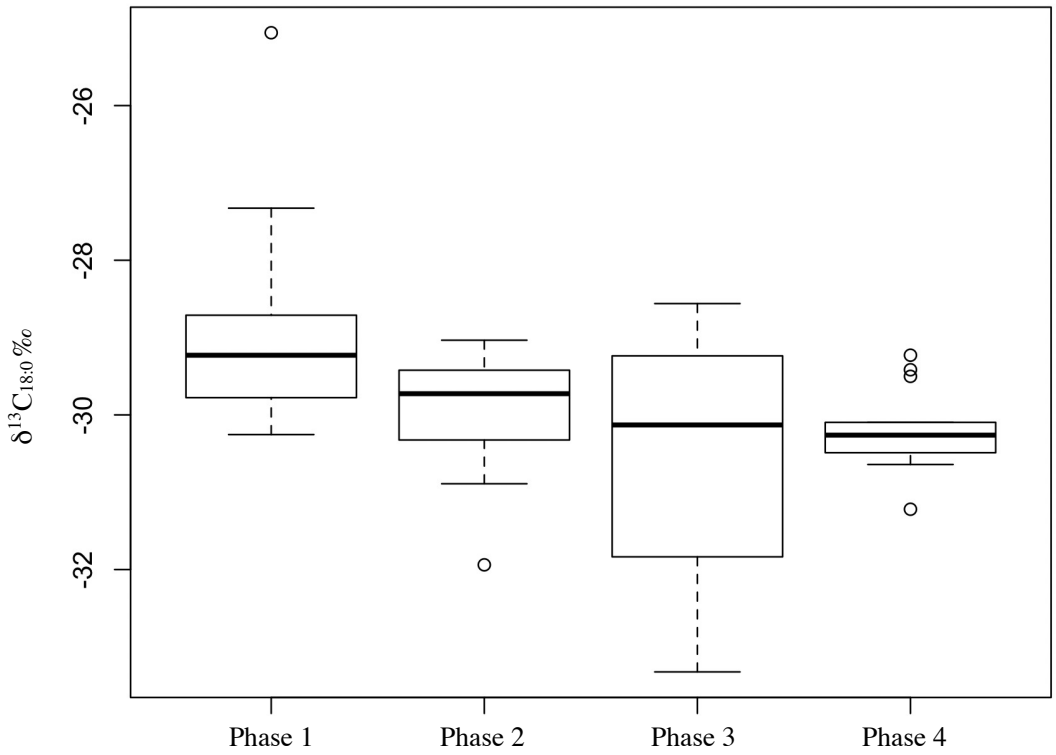


Figure 4.4. Distribution of $\delta^{13}C$ values of $C_{18:0}$ monocarboxylic acid extracted from four Neolithic phases of pottery from the site of Houtaomuga

$\delta^{13}C$ values of $C_{18:0}$ from Phase 3 show a wider distribution, however, this data set consists of four values and is likely, not representative. Furthermore, one of these samples (11 DHA III G001 (1) S2) has been discussed previously as it shows a use pattern different from the majority of the data set, and this sample exhibits $\delta^{13}C_{18:0}$ values consistent with ruminant fats rather than freshwater resources. In figure 4.5 below, this value has been removed and distribution narrowed significantly. This distribution chart better reflects the range of values identified and the specific pottery usage, meaning pottery was not used for a wide range of foods.

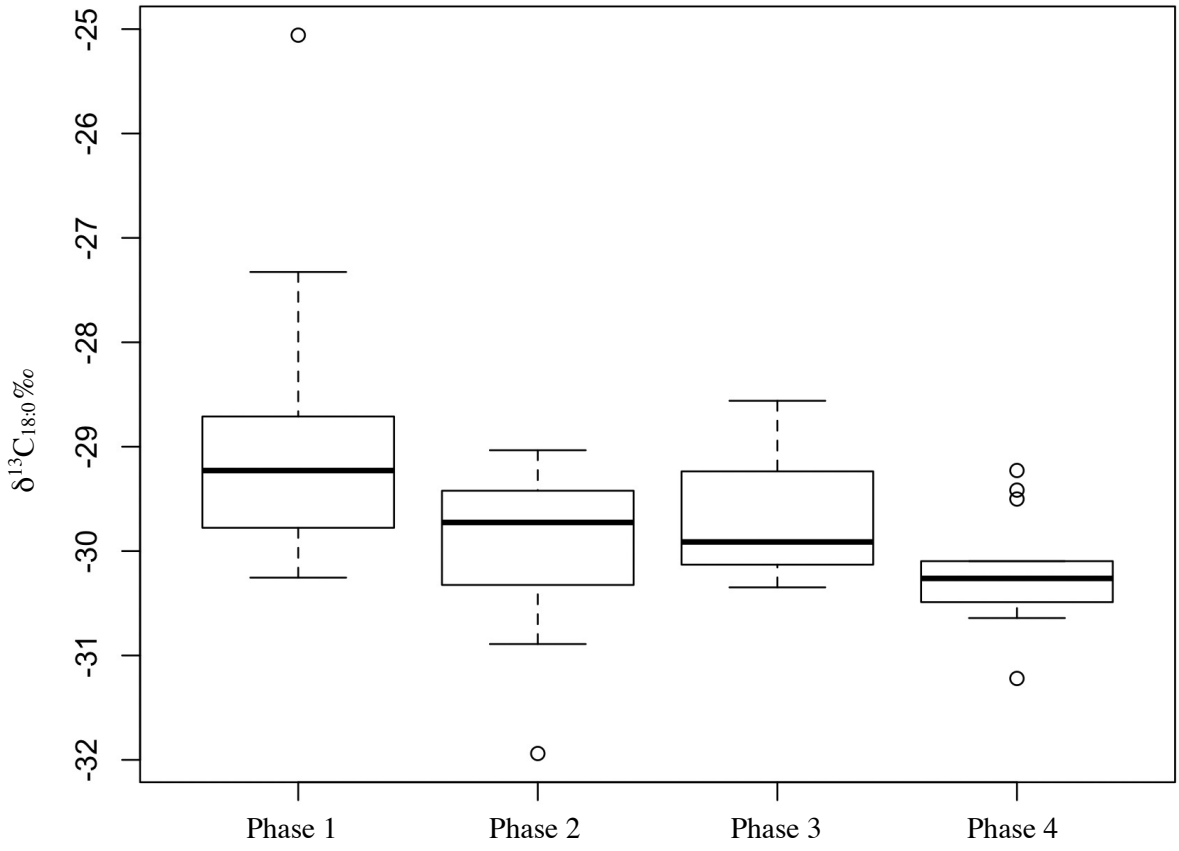


Figure 4.5. Distribution of $\delta^{13}C$ values of $C_{18:0}$ monocarboxylic acid extracted from four Neolithic phases of pottery from the site of Houtaomuga with outlier removed from Phase 3.

4.2 Changes between features

To identify differences in spatial use across the site, pottery residue results presented in Chapter 3 were compared between features and regarding feature proximity. A map of features at the site can be seen in section 1.1.5.3.2. This type of interpretation is often used to identify activity

areas within the archaeological site and can often be compared with other lines of evidence to better understand the overall site structure. Pottery use across all features is highly similar as discussed in previous sections, making this interpretation difficult. Results between each feature were compared by phase to see if any identifying patterns emerged. Nonetheless, all features show a wide spread of values that would not imply designated activity areas or specific feature use. An example of this non-specific and highly variable feature use can be seen in figure 4.6 showing $\Delta^{13}C$ values against $\delta^{13}C$ $C_{16:0}$ monocarboxylic acid values for Phase 1 spilt by feature. Figures showing results by feature for the remaining phases, 2, 3 and 4 can be seen in appendix IV. These results all cluster in the range of values for aquatic non-ruminant sources and display values of $\delta^{13}C_{16:0}$ monocarboxylic acid consistent with C_3 plant use. These results further exemplify the notion that pottery use at Houtaomuga was highly specific across all areas of the site and between occupational phases.

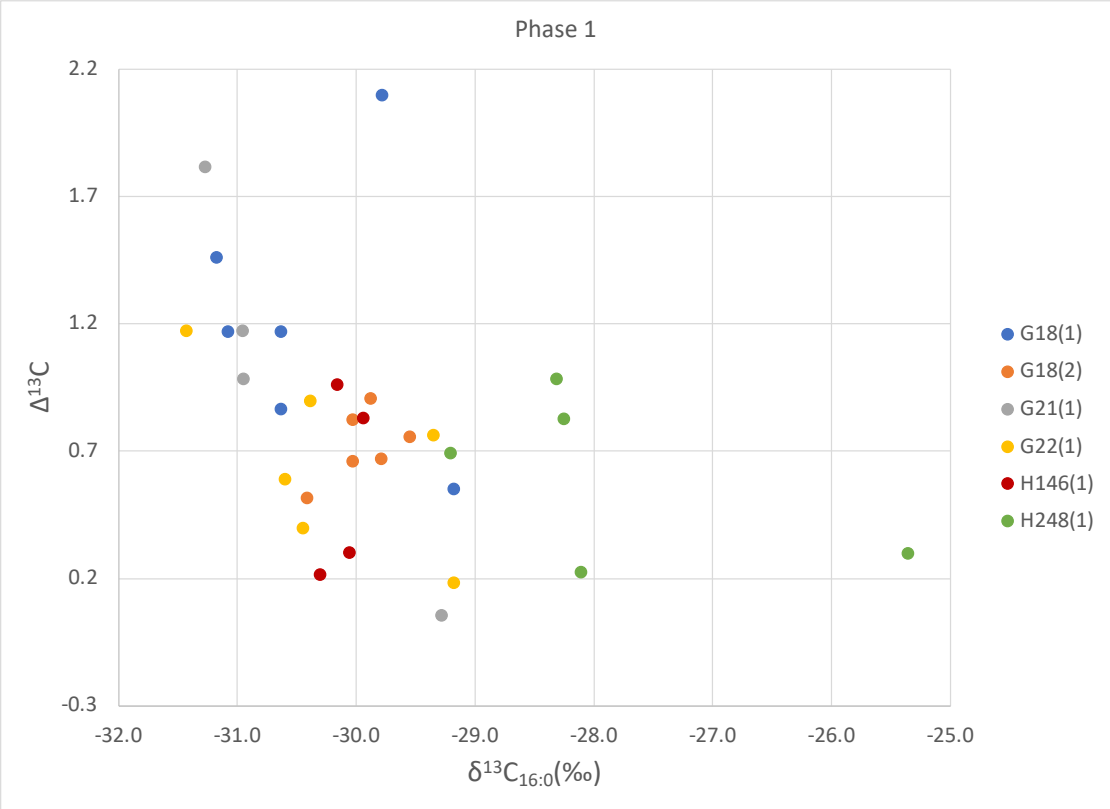


Figure 4.6. Plot of $\Delta^{13}C$ ($\delta^{13}C_{18:00}$ - $\delta^{13}C_{16:00}$) values against $\delta^{13}C_{16:00}$ values from Phase 1 from the site of Houtaomuga allowing for the distinction between various animal fats by feature.

4.3 Comparison with previous data

Kunikita et al., (2017) undertook research on charred residues from pottery from Houtaomuga (n=21) for radiocarbon dating and dietary reconstruction based on bulk carbon and nitrogen isotope composition and C/N ratios. This method of analysis is rudimentary for identifying previous pottery contents because of the ambiguity of different isotopic endpoints for many food types and diagenesis (Heron and Craig, 2015). While this dietary reconstruction is more uncertain and measured different indices than those in this study, I will present a discussion of these results because they consider the same site in question.

Nitrogen isotope ratios for these samples were higher than those for terrestrial animals and C_3 plants (inner vessel surface samples $\delta^{15}N$ 7.6-10.6‰, outer vessel surface samples $\delta^{15}N$ 7.5-16.0‰) with carbon isotope ratios being below -25‰, suggesting no influence from marine resources (Figure 4.7) (Kunikita et al., 2017). Overall, carbon isotope ratios for these samples are similar to those analysed in this thesis (-25.2 – -32.8; figure 4.7 and 4.8), despite being from bulk measurement as opposed to the single compound measurements taken in this study. This carbon isotope value range falls specifically in the scope for freshwater fish resources, suggesting these residues contained primarily freshwater fish lipids (Kunikita et al., 2017). Similar to the results of this work, Kunikita et al., (2017) found little evidence of changes in pottery use between the cultural phases. Kunikita et al., (2017) analysed 2 samples from Phase 1 and these samples exhibited $\delta^{13}C$ values of 32.8‰ and 31.3‰. These values are significantly depleted in $\delta^{13}C$ and are similar to the results of this thesis showing more depleted carbon isotope values for Phase 1 overall.

Figure 4.7 and 4.8 show data on pottery use from Houtaomuga and the nearby site of Shuangta as collected from Kunikita et al., (2017). Figure 5.7 shows $\delta^{15}N$ against $\delta^{13}C$ values while figure 5.8 shows $\delta^{13}C$ values against C/N ratios. Both figures have marked cluster regions for modern reference materials showing various food types. Kunikita et al., (2017) noted the wide range of C/N ratios and suggested this was likely the result of resource mixing with the pots and assemblage as a whole. Samples cluster in the C_3 plants and terrestrial mammal region in figure 5.8 concerning these C/N ratios. In this thesis, GC-MS and GC-C-IRMS analysis can further differentiate between these two resources. Results from these analyses show little evidence for terrestrial mammal fats in the form of cholesterol or branched odd chain monocarboxylic acids

and ruminant animal fats based on $\Delta^{13}C$ values (>-1.1). This may suggest that depleted single compound carbon isotope ratios in this study exhibit a complex mix of C_3 plants and freshwater resources. The results from this thesis also emphasize C_3 plant processing in pottery, and this resource group likely played a larger role in the diet than previously understood.

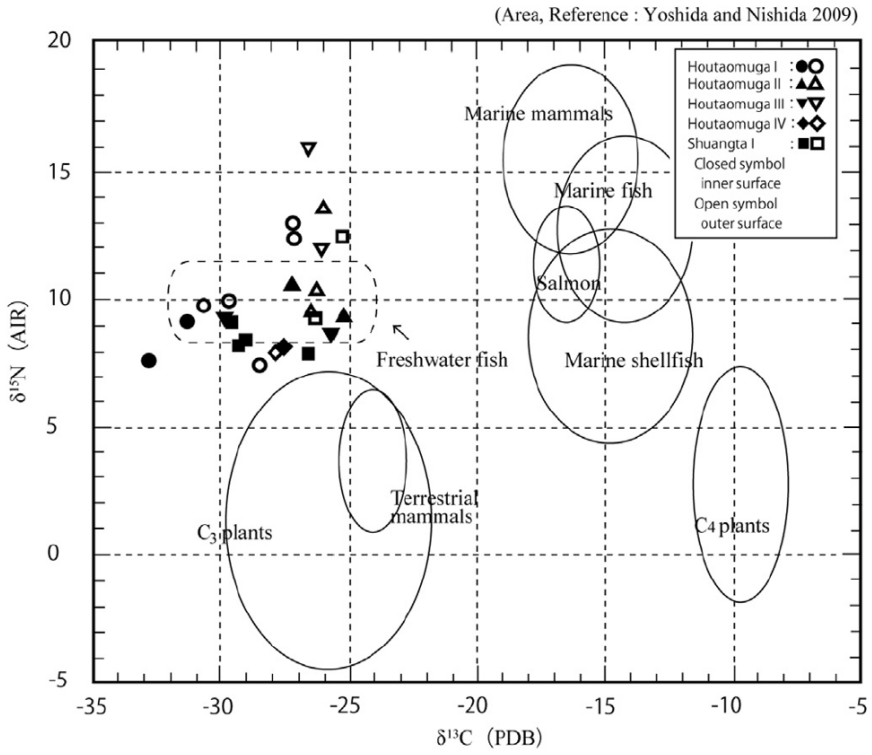


Figure 4.7. Carbon and Nitrogen isotope ratios of charred residues on pottery from Kunikita et al., (2017, p. 67) showing a comparison of data points with bulk isotope values for modern reference materials.

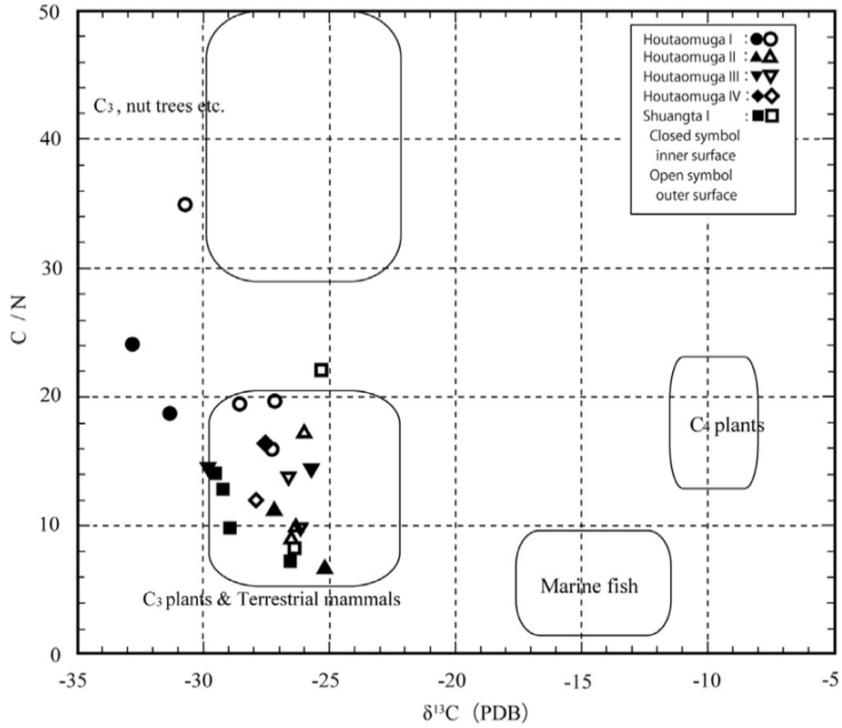


Figure 4.8. Carbon isotope ratios and C/N values of charred residues on pottery from Kunikita et al., (2017, p. 67) showing a comparison of data points with bulk isotope values for modern reference materials.

4.4 Resins, Hydrocarbons, Diterpenoids, and Lithics

This thesis included molecular analysis of lipid residues as described in section 3.2. Results from this analysis include evidence for various hydrocarbons and diterpenoids indicative of burning or deriving from wood smoke as discussed in sections 3.2.1 and 3.2.2. This result along with analysis from Liu et al (2016) and Wang and Sebillaud (2019) indicating pottery vessels from Houtaomuga exhibit extensive sooting around the exterior of vessels suggests these pots were often used for cooking or heating products over an open flame. These compounds identified most likely do not derive from the firing stage of the production process alone. This determination is important because it is highly problematic to refer to a pottery vessel by its use (i.e., cooking pot or storage pot) without empirical evidence of said use.

One sample discussed in section 3.2.2 from Phase 2 exhibited a full suite of compounds indicative of a resin. This resin could be made within the pot or applied as a surface treatment to the pot to make the ceramic body less permeable. If the latter were true, we might expect to see more evidence of this in the assemblage as a whole and this is not the case for the results of this study. Additional pottery samples from this phase have a similar permeability and more than one would likely have shown evidence of this surface treatment. This could be due to sampling bias, and the few pots which did not receive this treatment were chosen or the resinous surface treatment degraded in other vessels over time. I believe this resin was most likely present as people at Houtaomuga used microblade technologies which were hafted into composite tools (Chen et al., 2005; Elston et al., 2011; Lu, 1998; Qu et al., 2013). This type of resin is often found in other contexts with similar lithic styles used by hunter-gatherers as a hafting agent (Kozowyk et al., 2020; Mazza et al., 2006). Future work would benefit from sampling organic residues from lithics at the site. What is specifically noteworthy about this particular result is that pottery may have served as a tool in producing this resin, as the process required long periods of heating, however, many hunter-gatherer groups do not use pottery meaning it is not a required technology for the task. This suggests that using pottery for this activity is a specific choice being made by past people at Houtaomuga in this instance.

Pottery vessels from the first four phases of Neolithic occupation at the site show evidence for pottery repair in the form of drilled holes, and an example of this from Phase 4 can be seen in figure 4.9 (P. Sebillaud, personal communication, March 2022). This repair is illustrated by

the reconstruction in figure 4.10 showing that these holes were often tied together with twine and often waterproofed and/or glued together using an organic substance such as wax, bitumen, pitch or a combination of these substances (Miloglav, 2020). This result suggests that people had the necessary organic substances for pottery repair as early as Phase 2 and could have undertaken the process. More pottery samples with these obvious signs of repair should be sampled for residue analysis to better understand the organic component of this phenomenon.



Figure 4.9. Phase 4 reconstructed pottery vessel from Houtaomuga with repair holes (Wang et al., 2016, p. 327).

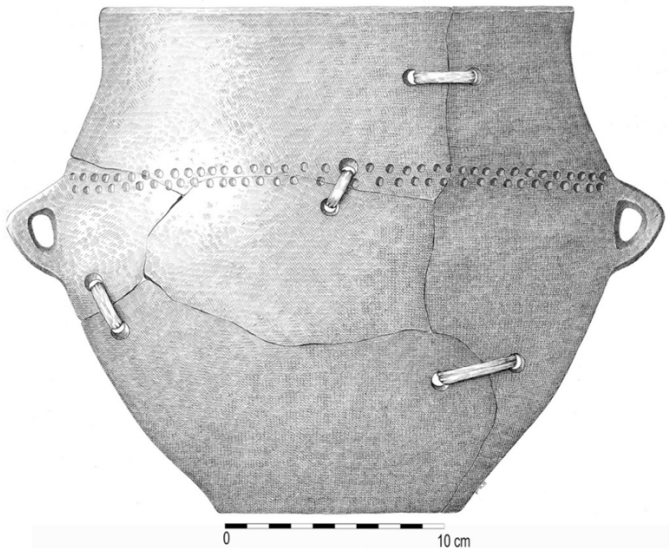


Figure 4.10. 1. Reconstruction of a repaired vessel from Damića gradina site in eastern Croatia with drilled holes which are tied together with fibre (Kudelić et al., 2018, p. 17)

4.5 Terrestrial Animals

The results of the residue analyses conducted in this study do not show evidence for the processing of terrestrial animals in pottery vessels across all phases of Neolithic occupation, except for possibly sample 11 DHA III G001 (1) S2 as discussed in section 4.1. The most common use-wear identified on the zooarchaeological record at Houtaomuga is smashing and chopping associated with marrow extraction, predominantly on auroch long bones (Luan, 2016; Song et al., 2016; Zhang, 2015). It has been suggested for other regions in China that pottery was heavily exploited by hunter-gatherer groups for cooking bone marrow either for bone grease production or soups, and that this was a driving force behind the adoption and continued popularity of vessel technology in other areas in China during the Late Palaeolithic (Patania and Jaffe, 2021). While the practice of marrow extraction is clearly present at Houtaomuga, residue analysis indicates this activity occurred without the use of pottery vessels, and these were instead used for plant and aquatic resources. This activity might have involved pottery at other sites in the Amur River Region of modern-day Russia according to residue results which show greater occurrences of ruminant animal fats, and this differentiation is discussed in section 4.8 (figure 4.11).

The most abundant species identified in the faunal assemblage at Houtaomuga during Phase 3 and 4 are auroch (Luan, 2016; Song et al., 2016). A slight change can be seen between Phase 3 and 4, where fewer fish and molluscs and more auroch were identified in the fourth Phase compared to the third, suggesting the beginnings of a shifting economy towards the Late Neolithic (Luan, 2016). This change is not seen in the pottery residues. This could be the result of a sampling bias, where the pottery used to cook auroch were not selected as a part of this study or this could imply differentiating cooking techniques. Auroch bones also have evidence for burning as if cooked over a fire, and this could be the preferred cooking method for this food type, while aquatic resources and plants are processed or cooked in a vessel. Zhang (2015) hypothesised that Houtaomuga was a seasonally occupied auroch hunting site during Phase 3 and 4 based on the increase in the contribution of auroch bones in these phase's faunal assemblages. Based on this additional new residue data added here, it seems that people are still favouring the cooking of aquatic resources in pottery. If the main reason for occupying the region during this time is for auroch hunting as Zhang (2015) has suggested, they are not frequently being cooked in pottery.

4.6 Aquatic Resources

Freshwater aquatic resources were the main product processed in pottery at Houtaomuga based on lipid residue analysis presented in this thesis. This is consistent with previous hypotheses about pottery usage at the site by other archaeologists (Kunikita et al., 2017; Wang and Sebillaud, 2019). The faunal assemblage at the site includes high amounts of fish and mollusc species which could easily have been obtained in the surrounding marshlands, lakes, and rivers rich environment (Sebillaud et al., 2021). Compounds deriving from fish oils often used as evidence for fish processing were not identified in the GC-MS results (Craig et al., 2007). These compounds are present in higher abundances in fish, particularly large carnivores, than molluscs and other small aquatic resources. It is likely these creatures were cooked in pottery more frequently at Houtaomuga than large fish, or large fish were always cooked with a large amount of C_3 plants, causing carbon mixing. Shells found at Houtaomuga during all phases of the Neolithic do not have traces of mechanical opening (i.e. with a tool of some type) and no traces of burning or sooting, and this suggests they were opened through cooking (P. Sebillaud, personal communication, March 2022). Results of isotopic analysis suggest this activity was frequently done in ceramic containers at the site. One individual from Phase 1 (tomb M45) has significant wear marks on their teeth, consistent with a diet which included sand (Xiao, 2014). This result has been interpreted as evidence for shelled creatures making up a significant proportion of the diets of Neolithic people at Houtaomuga, and isotopic results presented here support this (Xiao, 2014).

Use wear on the faunal assemblage at Houtaomuga shows grill marks and charring which is specifically prominent on mammalian long bones as mentioned above, and carnivorous fish (Song et al., 2016). This suggests one of the cooking methods for these larger fish was over an open flame instead of utilising vessel technology. This evidence in context with residue results presented in this thesis has some implications for pottery use and general cooking practices. Results of lipid residues deriving from aquatic resources might be showing more evidence for the processing of freshwater molluscs and other smaller animals rather than carnivorous fish due to increasingly depleted carbon isotope ratios. As seen in table 5.1, carnivorous fish are more enriched in $\delta^{13}C$ values, and if these animals were cooked in pots at Houtaomuga, it was

always in tandem with C_3 plant resources which are increasingly depleted in $\delta^{13}C$ values. These grill marks on these carnivorous fish highlight a separate cooking practice for these aquatic resources compared to results from residue analysis. There is a possibility that people living at Houtaomuga had separate preferences for preparing various aquatic resources and pottery was not used as a multitool for a wide variety of resources.

4.7 Plants

Residue analysis results from this thesis indicate that C_3 plants were frequently processed in pottery vessels present in all stages of occupation. This result comes from isotopic characterisation as well as molecular evidence in the form of squalene and monocarboxylic acids. Analysis of starches from grinding stones at Neolithic sites in northeast China show evidence for broad-spectrum plant use, and these tools were multifunctional and used for a variety of resources including both wild and domestic plants (Tao et al., 2011). Early phases show specific evidence for upland and wetland wild plant resources indicating plants made up a large proportion of the diet (Tang et al., 2020). Teeth from Houtaomuga have a high prevalence of calculus suggesting a high protein and low carbohydrate diet throughout (Zhang et al., 2016). Additionally, human teeth at Houtaomuga throughout the Neolithic are heavily worn, which could be related to the prevalence of plants in the diet in addition to sand inclusions (Xiao, 2014). This is consistent with zooarchaeological evidence at the site. Residue analysis in this study in tandem with previous research from Kunikita et al. (2017) demonstrates that pottery vessels were used primarily for freshwater aquatic resources and C_3 plants. According to research by Tang et al. (2020), these plants could have included green foxtail (*Setaria* sp.), millet tribe (*Paniceae*), soy (*Glycine* sp.), chenopod (*Chenopodium* sp.), knotweed (*Polygonum* sp.), cabbage family (*Brassicaceae*) and cattail (*Typha* sp.). All of these wild species, except *Paniceae,* fall into the C_3 plant category that is identified through isotopic characterization of residues and were likely a part of the former contents of pots at Houtaomuga. This plant use in pottery specifically is likely more common at Houtaomuga than at other Neolithic sites in Russia (figure 4.1), highlighting a unique broad-spectrum diet.

The earliest evidence for domesticated millets, a C_4 plant, in Northeast China dates to 8,700 cal BP, and starch grain analysis has shown evidence for foraging groups using millet and

Triticeae preferentially beginning in the LGM and the mid-Holocene (Yang et al., 2018). Nevertheless, there is no evidence of millet becoming a staple crop at Houtaomuga despite evidence that climatic fluctuation would favour such plants, and results from residue analysis in this thesis support this notion (Sebillaud et al., 2021; Tang et al., 2020; Wang and Sebillaud, 2019; Yang et al., 2018). While not a main component of the diet, millet was present, but very rare, at Houtaomuga during Phase 4 and possibly as early as Phase 3, however, pottery selected for this study shows no evidence for millet ever being cooked in pottery vessels at the site.

4.8 Discussion of Results in Broad Archaeological Context

The following chapter explores the relationship between results presented in chapter 4 within a broader archaeological context, prioritising contemporaneous hunter-gatherer sites in northeast Asia and residue analysis research on hunter-gatherer pottery. Chapter one contains a map with locations of sites mentioned here (figure 1.2) and visualisation of overlapping dates and culture periods for the region can be seen (figure 1.3). While it is important to understand results from Houtaomuga on their own, it is also vital to consider how results are similar or different from the surrounding region to contextualise these findings and better aid future research in this area involving a larger scope.

4.8.1 Comparison of results at Houtaomuga with Neolithic sites in the Amur River Region of Russia

Results from GC-MS and GC-C-IRMS analysis at Houtaomuga have been compared here with two published Neolithic sites located on the lower and middle sections of the Amur River in southern Russia. Pottery at these sites is similar to that of Houtaomuga with regional variation (Wang and Sebillaud, 2019). The site of Goncharka 1 is located 20 km upstream from the city of Khabarovsk (Shevkomud and Yanshina, 2012), making the site about 880 km (546.8 miles) away from Houtaomuga. The site is the most well-studied site for the Osipovka Culture Horizon and early pottery dates between 15,070 – 9,890 cal BP (Kuzmin, 2006; Shevkomud and Yanshina, 2012; Shoda et al., 2020). While Osipovka Culture pottery exhibits variability, vessels generally have thick walls, flat bottoms, and are tempered with various materials including gravel, grog, and plant fibres (Yanshina, 2017). Vessels also have external comb

marking similar to those at Houtaomuga (Wang and Sebillaud, 2019; Yanshina, 2017). The Gromatukha site is positioned on the Zeya River, a tributary joining the middle course of the Amur River and dates to 14,820-11,200 cal BP (Shoda et al., 2020). Pottery residues at both of these sites in Russia have been subject to GC-C-IRMS analysis, and due to their proximity, similar time range, pottery style, and environmental conditions, results are compared here.

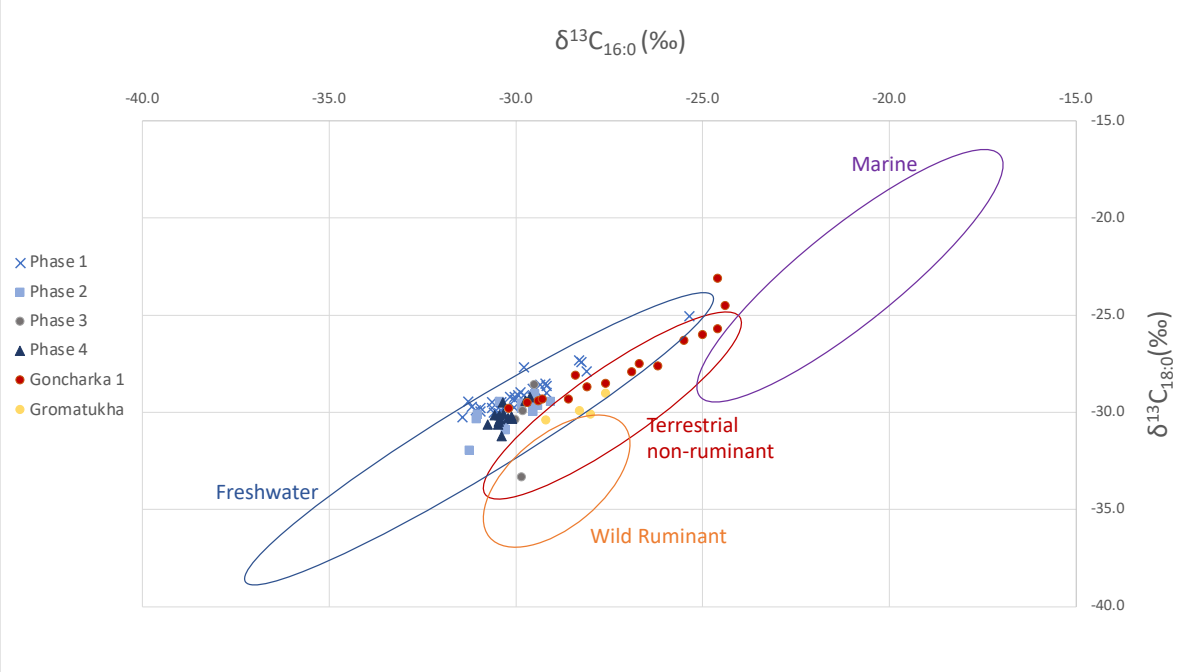


Figure 4.11. $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ monocarboxylic acids extracted from four Neolithic phases of pottery from the site of Houtaomuga and Gromatukha and Goncharka 1 from Russia (Shoda et al., 2020). The data are compared with reference ranges for authentic reference lipids from modern tissues and archaeological bone published by Lucquin et al. (2016) and Horiuchi et al. (2015). Samples from the sites in Russia include more terrestrial non-ruminant fats than Houtaomuga, showing a clear difference in pottery use between sites that are contemporaneous and nearby Houtaomuga.

Pottery use between Gromathukha and Goncharka 1 shows variability, with both areas having their own specific range of $\delta^{13}C$ values. There is some overlap with such use for Houtaomuga, however, the site seems to have its own distinct characteristics as well. Figure 4.11 showing the distribution of $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ n monocarboxylic acids from each site suggests Houtaomuga pottery was used for freshwater resources primarily, and these results cluster much closer together than those from the Amur. This suggests that pottery in the Amur region had a larger range of uses than at Houtaomuga, where samples have a much narrower range of values. Figure 5.12 shows $\Delta^{13}C$ ($\delta^{13}C_{18:00}$ - $\delta^{13}C_{16:00}$) values against $\delta^{13}C_{16:00}$ values from Houtaomuga Phase 1 to 4 and from Russia (Shoda et al., 2020). The Houtaomuga samples from

all phases are more depleted in $\delta^{13}C_{16:00}$, suggesting pottery was used for more C_3 plant processing than at sites in Russia. More samples from the sites in Russia exhibit $\Delta^{13}C$ values below the -1.1 threshold for ruminant fats, suggesting more of these pottery samples were used for processing these products at Houtaomuga. This could be related to hypotheses about bone grease (from large land mammals) production among hunter-gatherers during the Late Glacial period, and this activity might have been common in the Amur River region but absent from earlier Neolithic stages at Houtaomuga (Elston et al., 2011).

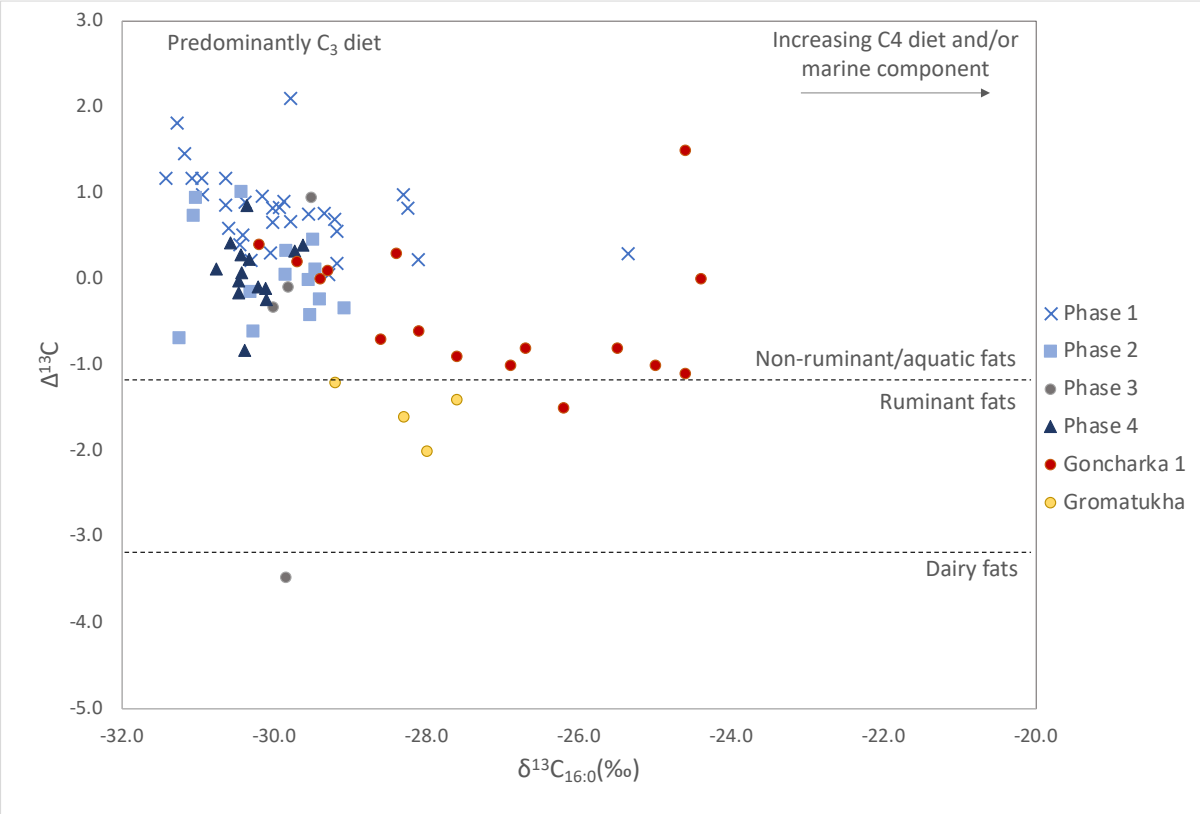


Figure 4.12. Plot of $\Delta^{13}C$ ($\delta^{13}C_{18:00}$ - $\delta^{13}C_{16:00}$) values against $\delta^{13}C_{16:00}$ values from Houtaomuga Phase 1 to 4 and contemporaneous sites Gromatukha and Goncharka 1 in Russia (Shoda et al., 2020) allowing for the distinction between various animal fats. Samples from the Russian sites are less depleted in $\delta^{13}C_{16:00}$ suggesting more carnivorous fish or C4 plants were processed in pottery. $\Delta^{13}C$ values of the samples from Russia are also lower, suggesting an increased use of ruminant fat compared to Houtaomuga.

The majority of samples from Houtaomuga and Gromatukha and Goncharka 1 in Russia are above this -1.1 $\Delta^{13}C$ threshold, however, suggesting a non-ruminant and/or aquatic fat source (figure 4.12). For the Amur sites, these values are less depleted in $\delta^{13}C_{16:00}$ and some of them contain lipid biomarkers indicative of fish i.e. ω-(o-alkylphenyl) alkanoic acids (APAA's)

containing 18 and 20 carbon atoms with at least one isoprenoid carboxylic acids (Evershed, 2008; Shoda et al., 2020). Such compounds were not identified in samples from Houtaomuga in this study. I believe this discrepancy to be related to a difference in pottery use and use of rivers and marshlands. Tissues from freshwater fish species have higher APAA concentrations and are less depleted in $\delta^{13}C_{16:00}$ and $\delta^{13}C_{18:00}$ than tissues from freshwater shellfish such as clams and molluscs. These differences in values are illustrated in table 4.1. The depletion in $\delta^{13}C_{16:00}$ may also be in part attributed to the cooking of C_3 plants which also display similar values (table 4.1) which may not be as available in the Amur region, processed using different means, or not preferred. It is likely the pottery at Houtaomuga was not used solely for processing plant remains, however, as they would be even more depleted in $\delta^{13}C_{16:00}$ and $\delta^{13}C_{18:00}$, and instead, a combination of plants and shellfish or shellfish alone were the main source of lipids extracted from the pottery. Four samples from Phase 1 at Houtaomuga are less depleted in $\delta^{13}C_{16:00}$ and are more similar to those from the Amur early phases. These samples may have been used for cooking fish as well, and do not fit as well into the main cluster of Houtaomuga results. Moreover, large carnivorous fish bones at Houtaomuga show evidence of charring, suggesting they were roasted over an open flame (Song et al., 2016). These results may suggest that fish were more often cooked using pottery in the Amur, but cooked over an open flame at Houtaomuga, where pottery was used more frequently for cooking shellfish and/or plants.

Table 4.1. Selection of modern reference samples and their respective $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ monocarboxylic acids used as a comparison in this study. For a full table, see appendix III

Class	Type	Taxa	$\delta^{13}C_{16:0}$	$\delta^{13}C_{18:0}$	Reference
Freshwater	Amur minnow	*Rhynchocypris lagowskii*	-27.43	-28.13	Craig et al. 2013
Freshwater	Topmouth gudgeon	*Pseudorasbora parva*	-26.95	-26.64	Craig et al. 2013
Freshwater	Crucian Carp	*Carassius sp.*	-28.64	-29.26	Lucquin et al 2016
Freshwater	Freshwater clam	-	-30.24	-30.03	Miyata et al 2015
Freshwater	Freshwater clam	*Cyrenidae*	-30.3	-29.7	Horiuchi et al. 2015
Freshwater	Salmon	*Salmonidae sp*	-26.4	-27.6	Horiuchi et al. 2015
Freshwater	Trout	*Salmonidae sp*	-26.7	27	Horiuchi et al. 2015
Plant	Walnut	-	-30.59	-31.8	Miyata et al 2015
Plant	Acorn	*Quercus sp.*	-35.15	-32.77	Miyata et al 2015
Plant	Chestnut	*Castanea sp.*	-34.64	-33.54	Miyata et al 2015
Plant	Acorn	*Quercus sp.*	-37.67	-35.71	Miyata et al 2015
Plant	Chesnut	*Castanea sp.*	-29.43	-28.71	Heron et al 2016
Plant	Horse chesnut	*Aesculus turbinata*	-31.82	-29.56	Heron et al 2016
Plant	Acorn (Fagus crenata blume)	*Fagus crenata*	-27.6	-28.35	Heron et al 2016
Plant	Acorn	*Quercus sp.*	-33.7	-34.14	Lucquin et al 2016
Plant	Chesnut	*Castanea sp.*	-33.35	-30.61	Heron et al 2016
Plant	acorn	*Quercus sp.*	-32.38	-33.33	Lucquin et al 2016
Plant	acorn	*Quercus sp.*	-34.77	-35.64	Lucquin et al 2016
Plant	acorn	*Quercus sp.*	-32.81	-34.32	Lucquin et al 2016
Plant	acorn	*Quercus sp.*	-31.81	-33.71	Lucquin et al 2016
Plant	White oak	*Quercus myrsinifolia*	-34.7	-33.4	Horiuchi et al. 2015

Samples from the Amur region are more enriched in $\delta^{13}C_{16:0}$ and $C_{18:0}$ than at Houtaomuga, and this is particularly prevalent for $C_{16:00}$. Figures 4.15 and 4.16 further illustrate this difference in values between the three sites by showing the distributions of their isotopic ratio values. Moreover, these distributions highlight the narrow range of values for samples from each phase at Houtaomuga despite this data set being the more extensive (for Houtaomuga n=63; Amur samples n=19). This may suggest that resources, people's diet, and food preferences at Houtaomuga are not as variable in the Amur region, or, more likely, pottery use at Houtaomuga is not as variable. This difference is most noticeable in comparing Phases 1 and 2 from Houtaomuga and Goncharka 1, especially considering the proximity of the sites and the similarities in pottery styles. Yanshina (2017) discussed regional differences between pottery shapes and decorations of the lower and middle Amur, and additional regional variability between these areas and Houtaomuga and other pottery from the Song-Nen Plain. Results from this research in combination with results from Shoda et al. (2020) suggest the regional differences in style also have regional differences in pottery use, and variability between hunter-gatherer groups in the Early Neolithic is prevalent and was important.

4.8.2 Comparison between Neolithic sites in Mainland East Asia and on the Japanese Archipelago

The following section discusses differences in pottery use throughout northeast Asia during the early stages of the Neolithic. Results compared here include samples from the Amur River region of Russia (n=19; Shoda et al. 2020) as discussed in section 1.1.6 as well as additional data from four sites on Sakhalin Island (n=12; Gibbs et al., 2017) and various Incipient and Initial Jōmon sites in Japan (n=561; Lucquin et al., 2018). A complete table of these samples and their respective dates can be found in appendix II. An illustration of these data is provided in figure 10.3 showing each value as compared to modern reference data for $\delta^{13}C_{16:00}$ and $\delta^{13}C_{18:00}$ (appendix III). Locations of these sites and their dates can be seen in figures 1.2 and 1.3.

The data points in figures 4.13 and 4.14 from the Japanese archipelago are more widely distributed than those at Houtaomuga, however, these data sets include far more sites, and this is expected. The strong marine component of this data set is noticeable and clearly shows this is an important product produced in vessels from this region during both Jōmon periods in question. This is expected due to the coastal location for most sites in Japan and on Sakhalin Island, and the mainland locations of Houtaomuga and sites in Russia would make a marine rich diet nearly impossible. It is noteworthy however that a large majority of these vessels are used for some type of aquatic resource, freshwater or otherwise. These samples also contain more pottery samples with terrestrial non-ruminant fats than at Houtaomuga, similar to pottery from the Amur region.

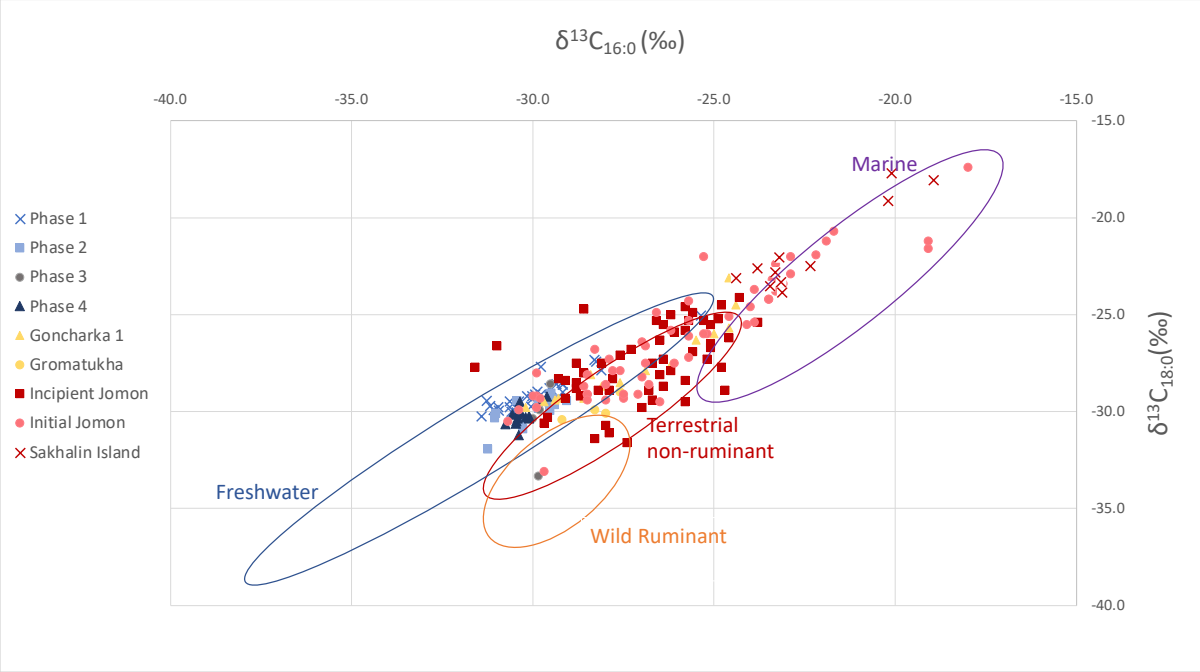


Figure 4.13. $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ monocarboxylic acids extracted from four Neolithic phases of pottery from the site of Houtaomuga, and from Russia (Shoda et al., 2020), three Neolithic sites on Sakhalin Island (Gibbs et al., 2017) and various Incipient and Initial Jōmon sites in Japan (Lucquin et al., 2018). The data are compared with reference ranges for authentic reference lipids from modern tissues and archaeological bone published by Lucquin et al. (2016) and Horiuchi et al. (2015). Samples from Houtaomuga exhibit a narrower cluster of values and do not include terrestrial non-ruminant and marine resources like other contemporaneous regions.

The $\Delta^{13}C$ ($\delta^{13}C_{18:00}$ - $\delta^{13}C_{16:00}$) proxy against $\delta^{13}C_{16:00}$ for all the northeast Asia sites in question illustrates the increase in $\delta^{13}C_{16:00}$ values related to pottery use in areas with marine resources (Figure 4.14). Additionally, the majority of samples include aquatic fats, further highlighting the importance of these resources on diet throughout the region (Figure 4.14). More sites from the Japanese Archipelago also plot in the region for ruminant animal fat in figure 4.14, and the use of this product is more widespread in this region. The tight cluster of samples from Houtaomuga in the region for aquatic fats and C_3 plant processing is notable, and this site has a narrow range of pottery uses compared to other regions.

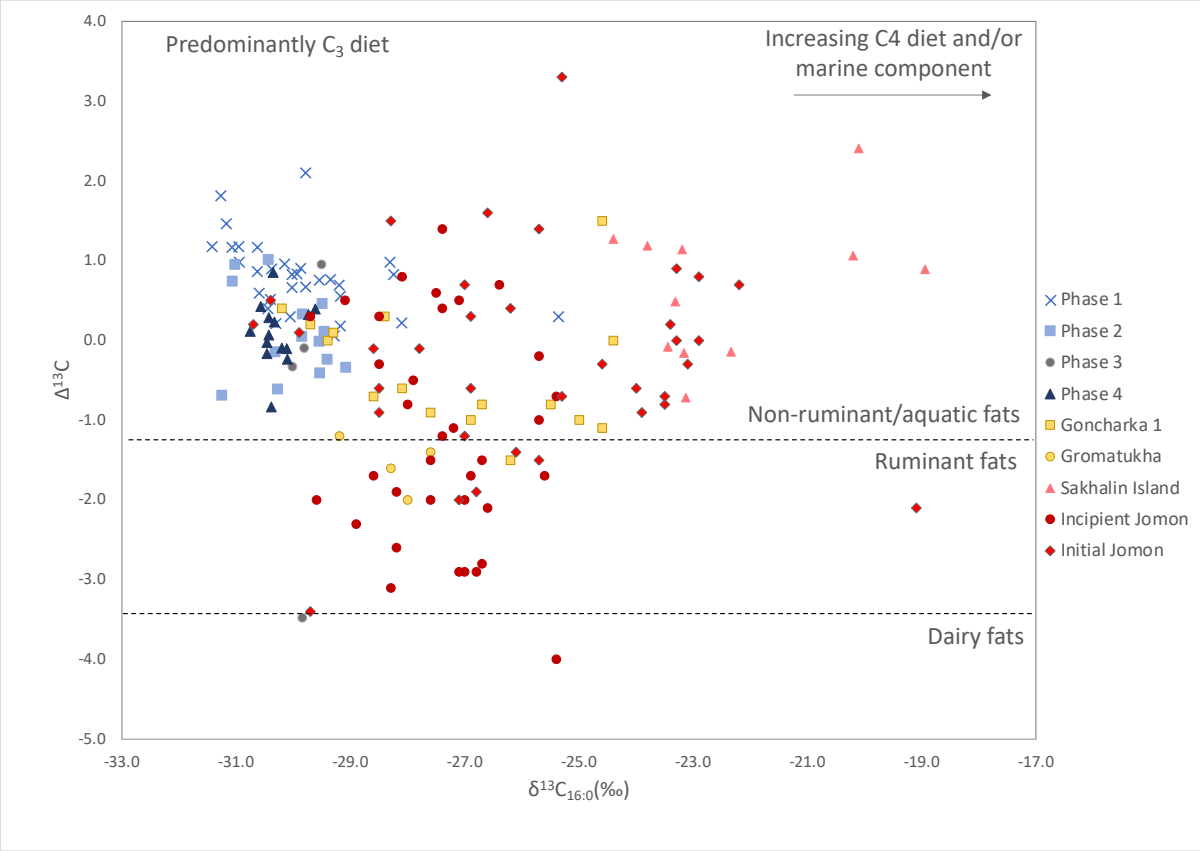


Figure 4.14. Plot of $\Delta^{13}C$ ($\delta^{13}C_{18:00}$ - $\delta^{13}C_{16:00}$) values against $\delta^{13}C_{16:00}$ values from Houtaomuga Phase 1 to 4 and from Russia (Shoda et al., 2020), three Neolithic sites on Sakhalin Island (Gibbs et al., 2017) and various Incipient and Initial Jōmon sites in Japan (Lucquin et al., 2018) allowing for the distinction between various animal fats.

The distribution of $\delta^{13}C_{16:00}$ and $\delta^{13}C_{18:00}$. $\delta^{13}C$ values from Houtaomuga fall into a narrow range in comparison to this additional data from around the region (figures 4.15 and 4.16). Data from Houtaomuga is more depleted in $\delta^{13}C$ values as a whole, likely a result of more C_3 plant and freshwater shellfish processing in pottery vessels compared to the surrounding region. Specifically, this differentiation is notable when comparing the first two phases at Houtaomuga to data distributions from the Japanese Archipelago, where data overlaps but similar values to those seen at Houtaomuga do not fall between the 25th to 75th percentiles of values. This suggests there may be some overlap between pottery use at some of these sites and Houtaomuga, but this is infrequent. Overall, pottery use at Houtaomuga includes a narrower range of products compared to the pottery of other regions.

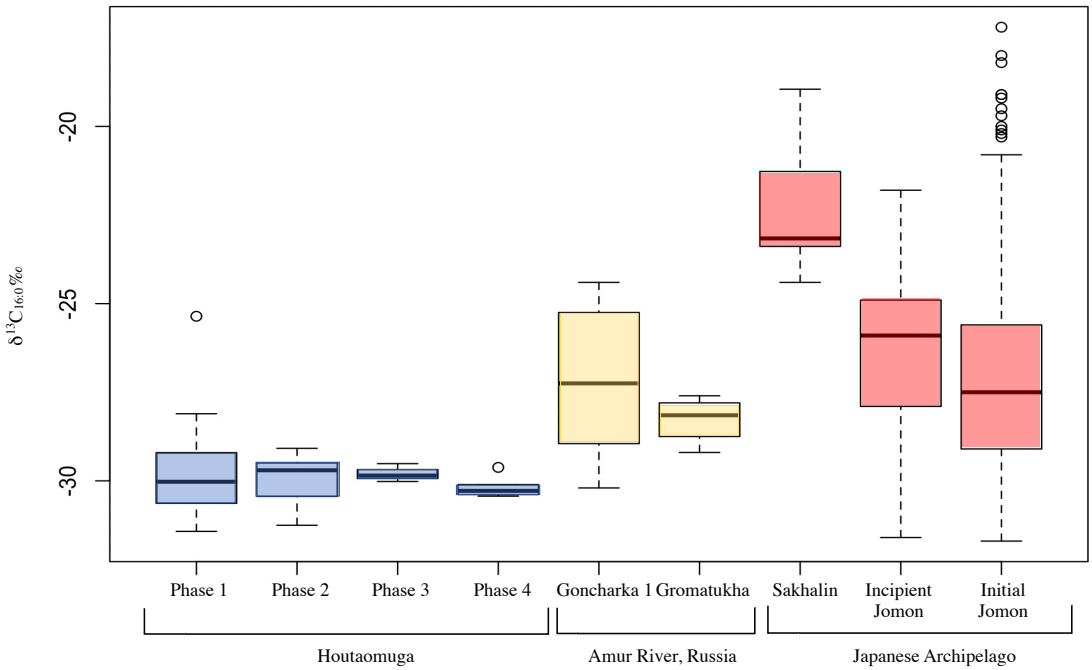


Figure 4.15. Distribution of $\delta^{13}C$ values of $C_{16:0}$ monocarboxylic acid extracted from four Neolithic phases of pottery from the site of Houtaomuga, two sites in Russia (Shoda et al., 2020), three Neolithic sites on Sakhalin Island (Gibbs et al., 2017) and various Incipient and Initial Jōmon sites in Japan (Lucquin et al., 2018).

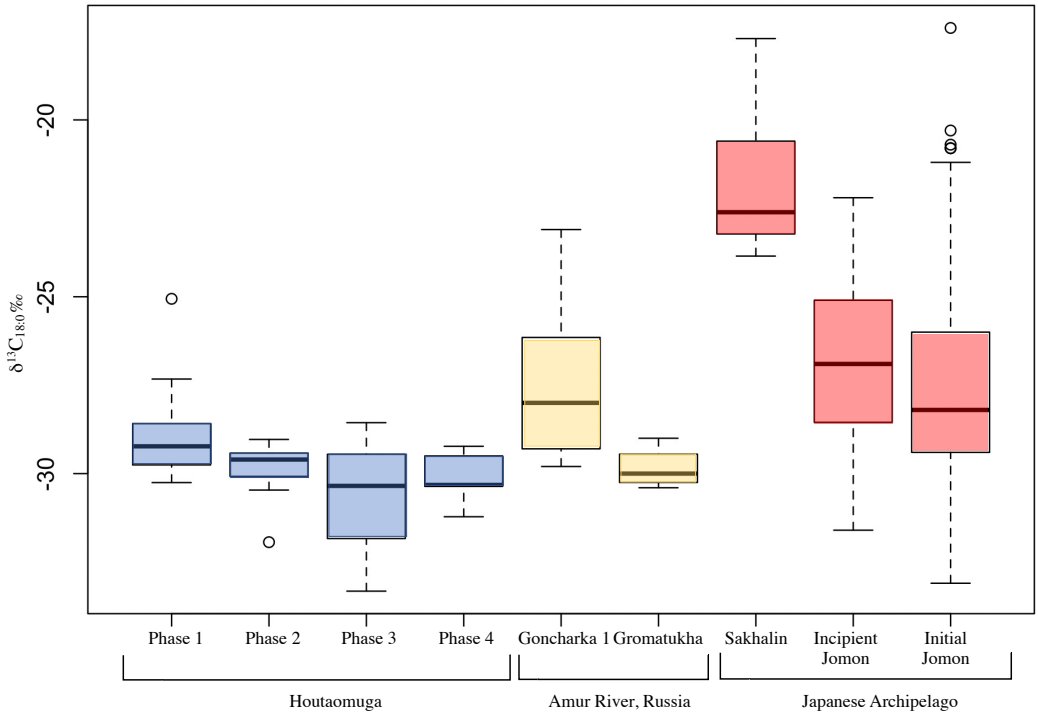


Figure 4.16. Distribution of $\delta^{13}C$ values of $C_{18:0}$ monocarboxylic acid extracted from four Neolithic phases of pottery from the site of Houtaomuga, two sites in Russia (Shoda et al., 2020), three Neolithic sites on Sakhalin Island (Gibbs et al., 2017) and various Incipient and Initial Jōmon sites in Japan (Lucquin et al., 2018).

As mentioned above, there is some overlap in similar $\delta^{13}C$ values between Jōmon sites on the Japanese Archipelago and Houtaomuga. Since Houtaomuga is a long distance from the coast, I expected this overlap to occur at sites that are also far from the coast in Japan, as marine lipids have significant effects on $\delta^{13}C$ values, and these effects are particularly noticeable among $\delta^{13}C$ values of $C_{16:0}$ monocarboxylic acid. To investigate this, sites published by Lucquin et al. (2018) with distributions of $\delta^{13}C$ values most similar to and/or within the same range of Houtaomuga were examined further. These sites included Setakitsunezuka, Tendogao, Tonohira, and Ohsikakubo. Sites examined and published by Lucquin et al. (2018) which were furthest from the coast were also examined and they include Ishihata, and Shimojuku. The distances from the coast and the respective time periods for each site can be seen in table 4.2.

Table 4.2. Table of sites mentioned in this section and the distances to the coast

Source	Site	Period	Distance to coast (km)
Keute 2022 (this study)	Houtaomuga	Houtaomuga 1-4	520
Lucquin et al. 2018	Setakitsunezuka	Initial Jomon	42
Lucquin et al. 2018	Tendogao	Initial Jomon	32
Lucquin et al. 2018	Tonohira	Initial Jomon	28
Lucquin et al. 2018	Ohshikakubo	Incipient Jomon	14
Lucquin et al. 2018	Ishihata	Incipient Jomon	84
Lucquin et al. 2018	Shimojuku	Incipient Jomon	113

The sites which are furthest from the coast in this instance have differentiating $\delta^{13}C$ values from Houtaomuga. These distributions of $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ monocarboxylic acid between the sites in question is illustrated in Figure 4.17 and 4.18. Sites with the most similar distributions to Houtaomuga are not far from coastal resources, by comparison, suggesting they are likely favouring C_3 plants. It is interesting to consider the Early Neolithic across northeast Asia is often referred to as the "aquatic Neolithic" where novel technology was needed to cook aquatic resources, however, this data could suggest the C_3 plant use in pottery is the most consistent across the region, while the aquatic resource exploitation is highly variable, and/or more depended on the environmental setting and cultural choices. Indeed, these preliminary results would need to be tested further using additional analyses at a wider range of sites to better understand this pattern.

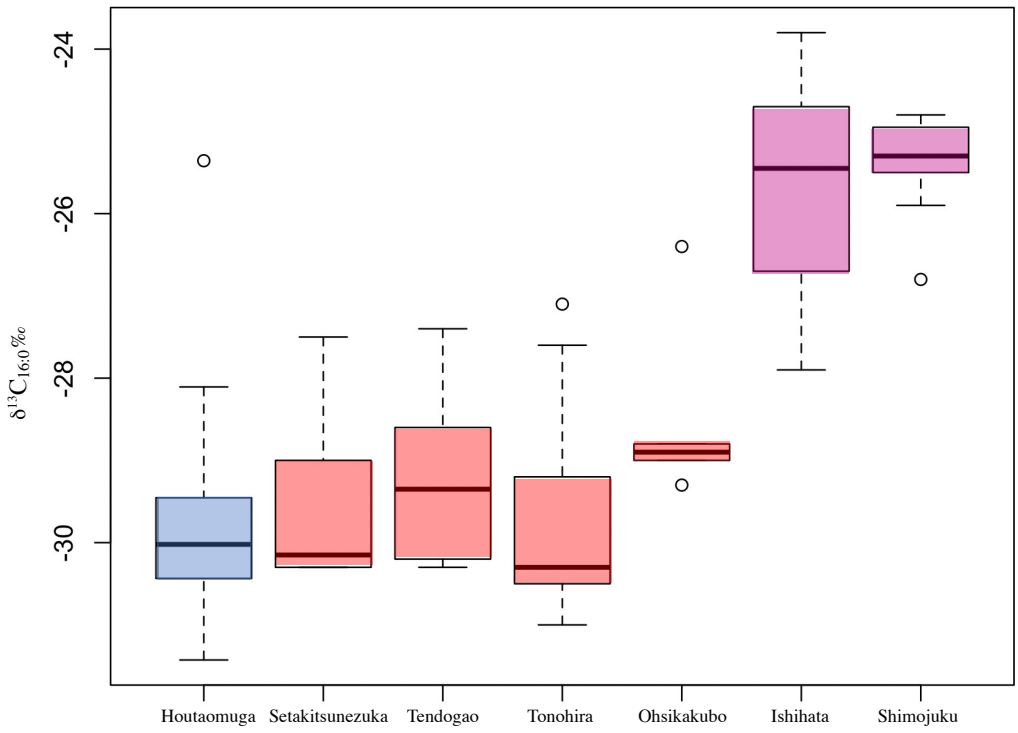


Figure 4.17. Distribution of $\delta^{13}C$ values of $C_{16:0}$ monocarboxylic acid extracted from all phases of pottery from the site of Houtaomuga (blue), select Incipient and Initial Jōmon sites in Japan (Lucquin et al., 2018). Sites with distributions of values similar to Houtaomuga are shown in red while values for sites in Japan from Lucquin et al. (2018) which are furthest from the coast are shown in magenta. These results show that pottery use between these regions is more similar in plant use than aquatic use.

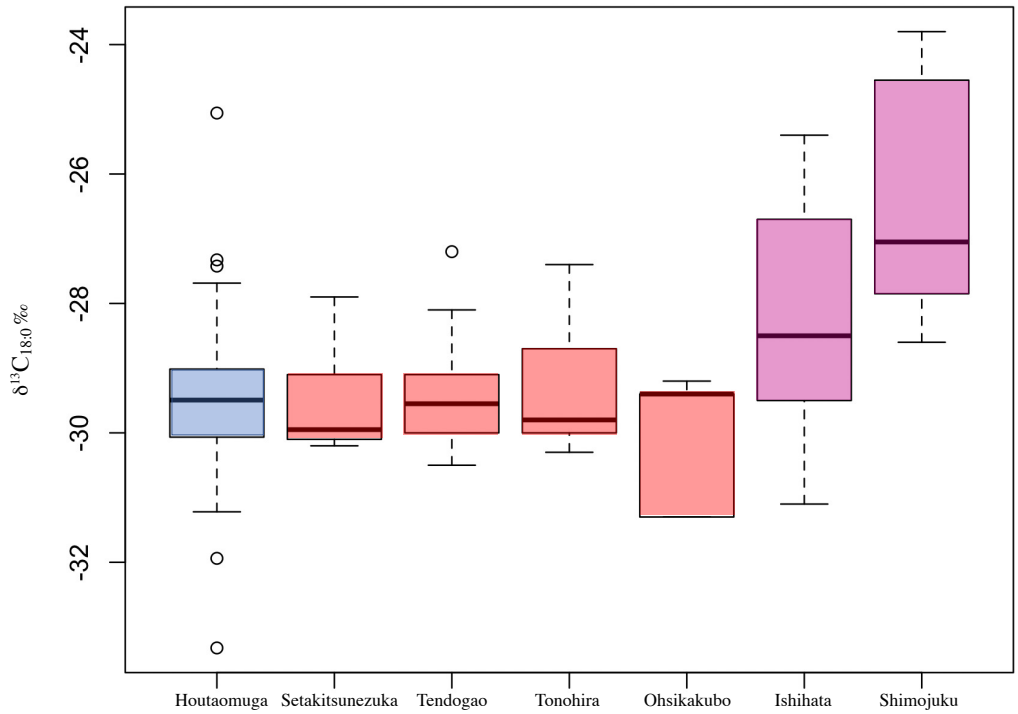


Figure 4.18. Distribution of $\delta^{13}C$ values of $C_{18:0}$ monocarboxylic acid extracted from all phases of pottery from the site of Houtaomuga (blue), select Incipient and Initial Jōmon sites in Japan (Lucquin et al., 2018). Sites with distributions of values similar to Houtaomuga are shown in red while values for sites in Japan from Lucquin et al. (2018) which are furthest from the coast are shown in magenta. These results show that pottery use between these regions is more similar in plant use than aquatic use.

4.8.3 Hunter-gatherer pottery use in relation to results at Houtaomuga

The location of Houtaomuga in the Lower Nen River Valley on the shores of Lake Xinhuang made it an ideal location to be a hunter-gatherer or hunter-gatherer-fisher seasonal settlement. Pottery, however, is not obligatory for this foraging-based economy, and this research shows that people living at the site chose to adopt the technology and use it to cook small freshwater aquatic animals and plants based on this research. It is possible they also used these vessels for cooking large fish; however, this would have only been in tandem with a significant number of plant resources. This is different to nearby sites near the Amur River in Russia where pottery is also commonly used for terrestrial animals and shows much less evidence for extensive plant use in vessels. Pottery use at Houtaomuga is different from most contemporaneous pottery yielding sites in Japan, which is not unexpected due to the varying environment, however, there is some overlap in increased wild plant exploitation. While these results at Houtaomuga

identifying aquatic processing are not themselves surprising, the narrow range of products cooked in pots at the site is. Pottery at Houtaomuga had a specific function regarding plants and small aquatic animals over a long period of time spanning from the first to the fourth occupational phase. This plant use in human diets at the site was suggested by Teng et al. (2020) and this research shows these foods were often cooked. For plants that require long cooking periods, pottery is more energy-efficient than other cooking methods including hot stone boiling.

I have mentioned previously (section 1.1) that pottery technology was not a necessary condition for fishing-forager economies. This notion makes the results of this study slightly more interesting, as we know cooking smaller aquatic shelled creatures does not require pottery, however, this is the preferred use of pottery at the site of Houtaomuga. This may highlight a specific choice made by the people living here who prefer a cooking style that can only be achieved with a pot, or a specific cuisine type that requires a pot. Food choice is often the result of various factors including the technologies available for the preparation and consumption of food (Fuller and Rowlands, 2011). Such choices in food use eventually result in "food-zones" – a collection of specific culinary practices which are the result of ritual, cosmological ideas, preferences, resource availability and technology (Fuller and Rowlands, 2011). The specific use of pottery at Houtaomuga which is unique to the region could suggest an early manifestation of a "food zone" where people have developed unique and intentional culinary practices.

Patania and Jaffe (2021) suggest that using pottery to cook soups during the early phases of pottery development in southern China had the ability to facilitate group cohesion through feasting. It is possible that using pottery for group feasting was a driving force for pottery popularity in northeast Asia, and these discrepancies between regions in lipid characterisation are due to bioavailable resources in the environment and regional food preferences. Patania and Jaffe (2021) specifically mention pottery being used for bone marrow extraction and use in these soups used for feasting. Potentially, this specific part of the practice is common in some areas while largely absent in others such as Houtaomuga, and this could be due to a variety of factors such as environmental conditions, resource availability, and partiality to plant or aquatic foods in certain areas. However, the lack of evidence for bone marrow production in pottery at Houtaomuga does not rule out the possibility of pottery being used for soups as

suggested. There is a possibility residue analysis results are highlighting the use of pottery for moist cooking technologies such as cooking soups or steaming molluscs. Pottery can retain fat and juices from food while they are cooking, retaining more nutrients and flavour in the final product or dish. To further understand this, more research across a wide geographic range would need to be undertaken which is beyond the scope of this work.

To further discuss the results of lipid residue analysis at Houtaomuga, I will discuss similarities and differences to results presented in this thesis with other published works on hunter-gatherer pottery outside of northeast Asia to better demonstrate what these results add to our understanding of hunter-gatherer pottery use. These examples come from a wide variety of geographic locations and varying local environments. In discussing them here I do not aim to view them comparatively, but instead, show the wide variety of hunter-gatherer lifeways even when similar technology may be employed.

Results from single-compound stable carbon isotope data of Ertebølle (EBK) and Narva pottery samples from hunter-gatherer coastal sites in Sweden, Denmark, Germany and Estonia show $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ monocarboxylic acid which fall within a much broader range than at Houtaomuga (Papakosta et al., 2019). For $\delta^{13}C_{16:0}$ these values fall between -32 to -16 and for $\delta^{13}C_{16:0}$ between -32 to -14. This range includes marine recourse which will always increase the range of values as these sources are far more enriched in both values. However, the wide range compared to Houtaomuga is striking. Pottery in this area seems to have a broader range of uses including plants as well as terrestrial and aquatic animals. This is different from the very specific pottery uses shown here over a long timescale at Houtaomuga.

Further research by Courel et al. (2020) shed further light on hunter-gatherer pottery in Europe by analysing residues of a large number of pottery samples from around the Baltic. They found that hunter-gatherers in each region all had their own specific pottery uses, with the those living on the eastern Baltic mainly using pottery for aquatic resources, and non-ruminant terrestrial animals in the southern Baltic, and a wide range of uses in the Northern Baltic mainly including ruminant animal products (Courel et al., 2020). Courel et al. (2020) argue that these differences in pottery use cannot be attributed to differences in environmental settings or resource availability, because in the eastern Baltic, for example, faunal remains show that hunter-gatherers were eating a broad range of foods including terrestrial non-ruminant animals, but

also that they were selectively not cooking them in pottery and this technology was exclusively used for aquatic animals. Courel et al. (2020) suggest these differences in pottery use are evidence of an emergence of subregional cuisines and changes to culinary practices made available by the use of pottery including cooking and exploring different food combinations. This argument implies culturally specific cooking choices were present and had strong influences over pottery use among hunter-gatherers in the past, and this practice has been identified in the ethnographic record (Courel et al., 2020; Kelly, 2013). While it is important to consider the environmental setting in which human activities occurred, it is also important we not think about this environment as a constraint or a factor that decides and controls human activities, particularly regarding hunter-gatherers and this research by Courel et al. (2020) exemplifies this. We might think because people lived in a region where a certain resource is abundant, they would mainly consume it, but this does not necessarily mean people chose to prepare said resource a certain way or include it in their diet at all.

The process by which hunter-gatherers took up and used pottery was not straightforward and the motivations behind these choices were more complex than archaeologists have previously imagined (Courel et al., 2020). Lipid residues from hunter-gatherer pottery dating to 2,500 to 2,800 years ago found near coastal sites in Alaska was analysed by Anderson et al. (2017). Researchers assumed this pottery would be primarily used for marine resources when instead it was more often used for freshwater aquatic animals and some terrestrial mammals (Anderson et al., 2017). This activity also seemed to be influenced in some ways by hunter-gatherer foodscapes and culinary choices. This result is also similar to results from Incipient and Initial Jōmon sites in Japan analysed by Lucquin et al. (2018), where pottery was primarily used for cooking plants and freshwater resources, similar to Houtaomuga, despite their proximity to the coast as discussed in section 4.8.2.

Regarding pottery use at Houtaomuga and northeast Asia as a whole, similar phenomena of culinary choices could have been at play. At Houtaomuga, we know people had a broad-spectrum diet based on the faunal remains, analysis of teeth, and palaeobotanical analysis. However, pottery from the beginning of occupation at ~13,000 BP to Phase 4 at 5,000 BP was used in very narrow ways. We have evidence for multiple dietary practices, including marrow extraction from aurochs long bones and roasting of large aquatic and terrestrial animals over open flames. Similar to results in the Baltic region as described by Courel et al. (2020), this

evidence seems to suggest that people were developing culturally influenced culinary practices which may not have been available to them without pottery vessels which allow for specific cooking performance and efficacy, ability to combine foodstuffs in a way which may not be as easy without pottery. Pottery use at Houtaomuga could have been influenced by foodscapes or food zones and pre-existing culinary practices similar to hunter-gatherers who adopt pottery technology in the Baltic. In summary, people at Houtaomuga had access to and ate similar food types to people living in the Amur River region in Russia, but these groups were using pottery in very different ways and the motivations behind these choices is unknown but likely culturally influenced. Shoda et al. (2020) suggest that the differences in early pottery use between the Amur region and the Japanese archipelago imply that pottery adoption and use was much more complex than previously understood, and these additional results from Houtaomuga further complicate this picture.

Furthermore, it would seem obvious that people at Houtaomuga would cook plants and molluscs in pottery based on the surrounding marshland and lake environment, but without this research here we would not know that people were actively not using pottery for other resources they eat including auroch and eventually millet. This type of choice is difficult to identify archaeologically without other means. Future research could explore this result further and look at human bone isotopes at Houtaomuga to better understand what portion of the diet was terrestrial versus aquatic animals. Future work at the site and in northeast Asia as a whole would benefit from microbotanical analysis on deposits from inside pottery vessels. Musaubach and Berón (Musaubach and Berón, 2017) undertook this type of analysis to better understand pottery use among hunter-gatherers in the archaeological record in Western Pampa, Argentina and identified specific plant remains from these contexts (Musaubach and Berón, 2017). Such research would be extremely useful in northeast Asia to better understand how increased plant exploitation, pottery use, and domestication may be intertwined.

Chapter 5 Conclusion

This thesis aimed to improve methods for residue analysis and to apply these improved methods to answer pressing archaeological questions about the adoption of pottery vessel technology and human diets during the Neolithic at Houtaomuga. Through the first application of non-destructive sampling, this thesis demonstrates that hunter-gatherers living at Houtaomuga during this time had specific and preferential uses for their pottery which differ from pottery use in surrounding regions of northeast Asia. The further implications for these results and future directions are described here.

5.1 Implications of Non-destructive Residue Analysis

This research presents the first attempt to characterise lipid residues from an archaeological context by SFE followed by GC-MS and GC-C-IRMS. Previous work using supercritical fluid for the extraction of lipids from archaeological pottery has only used GC-MS for characterisation purposes. The main challenge of adding the IRMS analysis is that this technique measures isotopic ratios from single compounds rather than from bulk lipid extracts, and single compound analysis requires a higher threshold of single lipid concentration for analysis. In this study, 94 samples were selected for IRMS analysis by GC-MS pre-screening which showed high (>5µg/g) concentrations of palmitic and stearic acid (quantification methods can be seen in appendix IX). Of these 94 samples, 63 had high enough amounts of measurable carbon within these compounds for accurate results for a success rate of 68.1% (appendix V).

These results were compared with published results using similar analysis techniques coupled with a liquid extraction of lipids. Published results from the Neolithic of northeast Asia were chosen to make these results as comparable as possible. The success rate of these selected works can be seen in table 5.1. The success rate for samples from Houtaomuga that underwent the SFE-GC-C-IRMS workflow is higher than similar studies using a liquid extraction. This result is promising, as it does not suggest any severe decrease in lipid extraction or preservation throughout analysis and lab protocols. However, it is important to consider the studies in question are on different pottery samples from different environments and burial contexts which could affect lipid preservation overall between the sample sets. Furthermore, these

numbers have been collected through published results, and there is a possibility that more or fewer samples may not have been included in supplementary materials.

Table 5.1. Success rate in carbon preservation in this study compared to two similar studies in nearby regions

	Extraction Method	Number of IRMS Samples	Number of IRMS samples with palmitic and stearic acid concentrations above threshold of analysis	% IRMS Success
Keute 2022 (this study)	SFE	94	64	68.1
Lucquin et al. 2018	Liquid	110	42	38.2
Gibbs et al. 2017	Liquid	42	11	26.2

One study in particular by Shoda et al. (2020) includes pottery residue analysis from sites closest to Houtaomuga in distance and age. This study includes 28 samples that were analysed by GC-C-IRMS after undergoing extraction of lipids. It is unclear if this study is only presenting the samples which were above the threshold of analysis, or if all samples were above the threshold for analysis making the success rate 100%. This differentiation is not noted in the text or supplementary materials. Because of this uncertainty, this research was not included in the final comparative table of data.

The successful application of this workflow to archaeological ceramics presented in this thesis is the first of its kind to sample pottery for absorbed lipid residues for single compound isotopic characterisation without crushing the pottery into a powder. This workflow is minimally destructive, and the pottery can be retained after analysis, either for curation or future scientific sampling. Future work with archaeological ceramics which must be non-destructive or minimally destructive could duplicate this workflow. These results are highly beneficial to samples that are of a significant age or importance and cannot be subject to traditional methods of analysis which require the ceramic sherds to be crushed. This method would also be beneficial for interdisciplinary research. Since the ceramic sherd remains intact, other analyses such as petrography, XRD, or LA-ICP-MS could be undertaken on the ceramic to understand the inorganic qualities of the pottery.

5.2 Overall Implications of Results from Houtaomuga

Residue analysis of pottery from the site at Houtaomuga supports the hypothesis that people living in the region during the Neolithic relied on a forager-fisher economy and pottery was used to cook plants and aquatic resources. Pottery shows distinct evidence for heating via molecular and use-wear proxies and vessels at the site can confidently be described as cooking pots. One pottery sample was likely used for the processing of resin used for hafting microblades into composite tools; a process that requires long periods of heating which could benefit from the use of a pot but which a pot is not required. This resin could also have been used as glue for pottery repair, which did occur during all periods at the site. This specific pottery use remained consistent between all four of the Neolithic phases at Houtaomuga suggesting pottery had a narrow use for a long period of time at the site with little variation. Pottery was likely used more often for cooking small aquatic resources rather than large carnivorous fish, which were instead cooked over an open flame. This result is consistent with other archaeological data at the site indicating that shells in archaeological deposits show evidence of being opened without mechanical means. Furthermore, human teeth from Phase 1 exhibit evidence for wear consistent with a diet which includes sand, which could come from consistent eating of molluscs. Results from this thesis suggest that cooking plants in pottery at Houtaomuga occurred frequently. Pottery is not a technology required for cooking the resources identified in pottery at Houtaomuga, and different food types were cooked using different means over long periods, suggesting that the adoption and long-continued use of pottery at Houtaomuga could have motivations other than utilitarian.

There are stark differences in pottery use between the site of Houtaomuga and those from other northeast Asian regions including the Amur River Valley and Japan. Notably, pottery at Houtaomuga was not used for cooking terrestrial animal foods such as bone marrow extraction, despite evidence for these food products being consumed, particularly in large quantities in Phase 4. Hunter-gatherer pottery in nearby regions during the Neolithic were used for cooking these resources and overall have a broader range of functions than at Houtaomuga. Moreover, pottery production and vessel shapes changed between occupational periods at Houtaomuga, but this change is not accompanied by a change in pottery use. We know from the faunal assemblage that people at Houtaomuga did include these resources into their diet and extracted bone marrow from mammalian long bones. While diets at Houtaomuga were broad-spectrum,

pottery use was limited to specific practices, which suggests the beginnings of specific culinary choices made by people living here during the Neolithic.

Interestingly, pottery across northeast Asia has specific regional variations in vessel form and decoration, with northeast China, Russia, Korea and Japan each having their own distinct variation (Yanshina, 2017). Results from this study in tandem with results from previous research suggest these differences in styles also had regional differences in use. This result is interesting to think about in addition to culinary choices which might be made during these periods. Perhaps the use of pottery in northeast Asia along with pre-existing foodscapes, bioavailable resources, and taste preferences allowed people to begin creating specific regional cuisines. It is possible these visual differences in pottery might have signalled different uses or food types to the people living in this area in the past. Either way, the relationship between the visual and production differences and the differences in residue analysis in this area should be further explored. Indeed, these regional differences could also be partially related to differences in environmental settings. While this could be a factor to some degree, the resources being exploited between sites in Russia and Houtaomuga are nearly identical, but pottery use is different. These differences instead appear to be related to specific choices being made by people in the past about how to use pottery and what to cook in it.

Future research using paleobotanical analysis, specifically from charred residue deposits in pottery vessels and grinding stone in northeast China would also be complementary to this work and results would shed further light on specific plant use and cuisines. This would particularly be interesting to test at Houtaomuga to see if the narrow range of pottery functions we see in residue results are related to specific plant uses. Indeed, future research involving residue analysis at other Neolithic site in the surrounding region in northeast China would be highly beneficial to understand the range of specific pottery uses at Houtaomuga. A deeper investigation into the relationships between pottery styles and usage in the whole of northeast Asia is necessary to further understand the phenomenon of hunter-gatherer pottery during the Neolithic in the region. Unfortunately, a research project of this scale would be too extensive for one individual to undertake in a single thesis and in the future should include numerous scholars.

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Appendix I:
List of all samples

Field Number	Province	Year	City/Site	Zone	Feature	Layer	Sample	Weight (mg)	Phase
12 DHA III G18 (1)_S1	Jilin	12	DHA - Houtaomuga	III	G18	1	S1	1398	Phase 1
12 DHA III G18 (1)_S2	Jilin	12	DHA - Houtaomuga	III	G18	1	S2	1145	Phase 1
12 DHA III G18 (1)_S3	Jilin	12	DHA - Houtaomuga	III	G18	1	S3	1446	Phase 1
12 DHA III G18 (1)_S4	Jilin	12	DHA - Houtaomuga	III	G18	1	S4	920	Phase 1
12 DHA III G18 (1)_S5	Jilin	12	DHA - Houtaomuga	III	G18	1	S5	789	Phase 1
12 DHA III G18 (1)_S6	Jilin	12	DHA - Houtaomuga	III	G18	1	S6	1241	Phase 1
12 DHA III G18 (1)_S7	Jilin	12	DHA - Houtaomuga	III	G18	1	S7	1209	Phase 1
12 DHA III G18 (1)_S8	Jilin	12	DHA - Houtaomuga	III	G18	1	S8	1223	Phase 1
12 DHA III G18 (1)_S9	Jilin	12	DHA - Houtaomuga	III	G18	1	S9	2166	Phase 1
12 DHA III G18 (1)_S10	Jilin	12	DHA - Houtaomuga	III	G18	1	S10	1045	Phase 1
12 DHA III G18 (2)_S1	Jilin	12	DHA - Houtaomuga	III	G18	2	S1	1477	Phase 1
12 DHA III G18 (2)_S2	Jilin	12	DHA - Houtaomuga	III	G18	2	S2	1748	Phase 1
12 DHA III G18 (2)_S3	Jilin	12	DHA - Houtaomuga	III	G18	2	S3	1957	Phase 1
12 DHA III G18 (2)_S4	Jilin	12	DHA - Houtaomuga	III	G18	2	S4	1443	Phase 1

12 DHA III G18 (2)_S5	Jilin	12	DHA - Houtaomuga	III	G18	2	S5	1474	Phase 1
12 DHA III G18 (2)_S6	Jilin	12	DHA - Houtaomuga	III	G18	2	S6	1479	Phase 1
12 DHA III G18 (2)_S7	Jilin	12	DHA - Houtaomuga	III	G18	2	S7	2109	Phase 1
12 DHA III G18 (2)_S8	Jilin	12	DHA - Houtaomuga	III	G18	2	S8	1661	Phase 1
12 DHA III G18 (2)_S9	Jilin	12	DHA - Houtaomuga	III	G18	2	S9	1831	Phase 1
12 DHA III G18 (2)_S10	Jilin	12	DHA - Houtaomuga	III	G18	2	S10	2668	Phase 1
12 DHA III G18 (2)_S11	Jilin	12	DHA - Houtaomuga	III	G18	2	S11	1561	Phase 1
12 DHA III G18 (2)_S12	Jilin	12	DHA - Houtaomuga	III	G18	2	S12	1816	Phase 1
12 DHA III G18 (2)_S13	Jilin	12	DHA - Houtaomuga	III	G18	2	S13	1618	Phase 1
12 DHA III G18 (2)_S14	Jilin	12	DHA - Houtaomuga	III	G18	2	S14	2392	Phase 1
12 DHA III G18 (2)_S15	Jilin	12	DHA - Houtaomuga	III	G18	2	S15	2037	Phase 1
12 DHA III G18 (2)_S16	Jilin	12	DHA - Houtaomuga	III	G18	2	S16	1341	Phase 1
12 DHA III G18 (2)_S17	Jilin	12	DHA - Houtaomuga	III	G18	2	S17	2187	Phase 1
12 DHA III G18 (2)_S18	Jilin	12	DHA - Houtaomuga	III	G18	2	S18	1386	Phase 1
12 DHA III G18 (2)_S19	Jilin	12	DHA - Houtaomuga	III	G18	2	S19	1942	Phase 1

12 DHA III G18 (2)_S20	Jilin	12	DHA - Houtaomuga	III	G18	2	S20	1651	Phase 1
12 DHA III G18 (2)_S21	Jilin	12	DHA - Houtaomuga	III	G18	2	S21	3219	Phase 1
12 DHA III G18 (2)_S22	Jilin	12	DHA - Houtaomuga	III	G18	2	S22	2163	Phase 1
12 DHA III G18 (2)_S23	Jilin	12	DHA - Houtaomuga	III	G18	2	S23	2026	Phase 1
12 DHA III G18 (2)_S24	Jilin	12	DHA - Houtaomuga	III	G18	2	S24	2009	Phase 1
12 DHA III G18 (2)_S25	Jilin	12	DHA - Houtaomuga	III	G18	2	S25	2277	Phase 1
12 DHA III G18 (2)_S26	Jilin	12	DHA - Houtaomuga	III	G18	2	S26	2894	Phase 1
12 DHA III G18 (2)_S27	Jilin	12	DHA - Houtaomuga	III	G18	2	S27	1619	Phase 1
12 DHA III G18 (2)_S28	Jilin	12	DHA - Houtaomuga	III	G18	2	S28	1980	Phase 1
12 DHA III G18 (2)_S29	Jilin	12	DHA - Houtaomuga	III	G18	2	S29	1867	Phase 1
12 DHA III G18 (2)_S30	Jilin	12	DHA - Houtaomuga	III	G18	2	S30	1667	Phase 1
12 DHA III G21 (1)_S1	Jilin	12	DHA - Houtaomuga	III	G21	1	S1	891	Phase 1
12 DHA III G21 (1)_S2	Jilin	12	DHA - Houtaomuga	III	G21	1	S2	1237	Phase 1
12 DHA III G21 (1)_S3	Jilin	12	DHA - Houtaomuga	III	G21	1	S3	628	Phase 1
12 DHA III G21 (1)_S4	Jilin	12	DHA - Houtaomuga	III	G21	1	S4	994	Phase 1

12 DHA III G21 (1)_S5	Jilin	12	DHA - Houtaomuga	III	G21	1	S5	2019	Phase 1
12 DHA III G22 (1)_S1	Jilin	12	DHA - Houtaomuga	III	G22	1	S1	2241	Phase 1
12 DHA III G22 (1)_S2	Jilin	12	DHA - Houtaomuga	III	G22	1	S2	1202	Phase 1
12 DHA III G22 (1)_S3	Jilin	12	DHA - Houtaomuga	III	G22	1	S3	2397	Phase 1
12 DHA III G22 (1)_S4	Jilin	12	DHA - Houtaomuga	III	G22	1	S4	1336	Phase 1
12 DHA III G22 (1)_S5	Jilin	12	DHA - Houtaomuga	III	G22	1	S5	2145	Phase 1
12 DHA III G22 (1)_S6	Jilin	12	DHA - Houtaomuga	III	G22	1	S6	1500	Phase 1
12 DHA III G22 (1)_S7	Jilin	12	DHA - Houtaomuga	III	G22	1	S7	1036	Phase 1
12 DHA III G22 (1)_S8	Jilin	12	DHA - Houtaomuga	III	G22	1	S8	1834	Phase 1
12 DHA III G22 (1)_S9	Jilin	12	DHA - Houtaomuga	III	G22	1	S9	1726	Phase 1
12 DHA III G22 (1)_S10	Jilin	12	DHA - Houtaomuga	III	G22	1	S10	2422	Phase 1
12 DHA III H146 (1)_S1	Jilin	12	DHA - Houtaomuga	III	H146	1	S1	1814	Phase 1
12 DHA III H146 (1)_S2	Jilin	12	DHA - Houtaomuga	III	H146	1	S2	1142	Phase 1
12 DHA III H146 (1)_S3	Jilin	12	DHA - Houtaomuga	III	H146	1	S3	1283	Phase 1
12 DHA III H146 (1)_S4	Jilin	12	DHA - Houtaomuga	III	H146	1	S4	1857	Phase 1

12 DHA III H146 (1)_S5	Jilin	12	DHA - Houtaomuga	III	H146	1	S5	1316	Phase 1
12 DHA III H248 (1)_S1	Jilin	12	DHA - Houtaomuga	III	H248	1	S1	1523	Phase 1
12 DHA III H248 (1)_S2	Jilin	12	DHA - Houtaomuga	III	H248	1	S2	2584	Phase 1
12 DHA III H248 (1)_S3	Jilin	12	DHA - Houtaomuga	III	H248	1	S3	2072	Phase 1
12 DHA III H248 (1)_S4	Jilin	12	DHA - Houtaomuga	III	H248	1	S4	2104	Phase 1
12 DHA III H248 (1)_S5	Jilin	12	DHA - Houtaomuga	III	H248	1	S5	1550	Phase 1
11 DHA III G002 (1)_S1	Jilin	11	DHA - Houtaomuga	III	G002	1	S1	965	Phase 2
11 DHA III G002 (1)_S2	Jilin	11	DHA - Houtaomuga	III	G002	1	S2	2245	Phase 2
11 DHA III G002 (1)_S3	Jilin	11	DHA - Houtaomuga	III	G002	1	S3	1212	Phase 2
11 DHA III G002 (1)_S4	Jilin	11	DHA - Houtaomuga	III	G002	1	S4	1657	Phase 2
11 DHA III G002 (1)_S5	Jilin	11	DHA - Houtaomuga	III	G002	1	S5	935	Phase 2
11 DHA III G002 (1)_S6	Jilin	11	DHA - Houtaomuga	III	G002	1	S6	1530	Phase 2
11 DHA III G002 (1)_S7	Jilin	11	DHA - Houtaomuga	III	G002	1	S7	609	Phase 2
11 DHA III G002 (1)_S8	Jilin	11	DHA - Houtaomuga	III	G002	1	S8	1061	Phase 2
11 DHA III G002 (1)_S9	Jilin	11	DHA - Houtaomuga	III	G002	1	S9	2199	Phase 2

11 DHA III G002 (1)_S10	Jilin	11	DHA - Houtaomuga	III	G002	1	S10	741	Phase 2
11 DHA III G13_S1	Jilin	11	DHA - Houtaomuga	III	G13		S1	1808	Phase 2
11 DHA III H131 (1)_S1	Jilin	11	DHA - Houtaomuga	III	H131	1	S1	842	Phase 2
11 DHA III H131 (1)_S2	Jilin	11	DHA - Houtaomuga	III	H131	1	S2	1310	Phase 2
11 DHA III H131 (1)_S3	Jilin	11	DHA - Houtaomuga	III	H131	1	S3	1370	Phase 2
11 DHA III H131 (1)_S4	Jilin	11	DHA - Houtaomuga	III	H131	1	S4	863	Phase 2
11 DHA III H131 (1)_S5	Jilin	11	DHA - Houtaomuga	III	H131	1	S5	972	Phase 2
11 DHA III H131 (1)_S6	Jilin	11	DHA - Houtaomuga	III	H131	1	S6	1030	Phase 2
11 DHA III H131 (1)_S7	Jilin	11	DHA - Houtaomuga	III	H131	1	S7	474	Phase 2
11 DHA III H131 (1)_S8	Jilin	11	DHA - Houtaomuga	III	H131	1	S8	735	Phase 2
11 DHA III H131 (1)_S9	Jilin	11	DHA - Houtaomuga	III	H131	1	S9	1032	Phase 2
11 DHA III H131 (1)_S10	Jilin	11	DHA - Houtaomuga	III	H131	1	S10	1031	Phase 2
11 DHA III G001 (1)_S1	Jilin	11	DHA - Houtaomuga	III	G001	1	S1	1236	Phase 3
11 DHA III G001 (1)_S2	Jilin	11	DHA - Houtaomuga	III	G001	1	S2	1283	Phase 3
11 DHA III G001 (1)_S3	Jilin	11	DHA - Houtaomuga	III	G001	1	S3	1097	Phase 3

11 DHA III G001 (1)_S4	Jilin	11	DHA - Houtaomuga	III	G001	1	S4	1286	Phase 3
11 DHA III G001 (1)_S5	Jilin	11	DHA - Houtaomuga	III	G001	1	S5	664	Phase 3
11 DHA III G001 (1)_S6	Jilin	11	DHA - Houtaomuga	III	G001	1	S6	656	Phase 3
11 DHA III G001 (1)_S7	Jilin	11	DHA - Houtaomuga	III	G001	1	S7	1272	Phase 3
11 DHA III G001 (1)_S8	Jilin	11	DHA - Houtaomuga	III	G001	1	S8	637	Phase 3
11 DHA III G001 (1)_S9	Jilin	11	DHA - Houtaomuga	III	G001	1	S9	1135	Phase 3
11 DHA III G001 (1)_S10	Jilin	11	DHA - Houtaomuga	III	G001	1	S10	1361	Phase 3
11 DHA III G001 (1)_S11	Jilin	11	DHA - Houtaomuga	III	G001	1	S11	1570	Phase 3
11 DHA III G001 (1)_S12	Jilin	11	DHA - Houtaomuga	III	G001	1	S12	2090	Phase 3
11 DHA III G001 (1)_S13	Jilin	11	DHA - Houtaomuga	III	G001	1	S13	579	Phase 3
11 DHA III G001 (1)_S14	Jilin	11	DHA - Houtaomuga	III	G001	1	S14	773	Phase 3
11 DHA III G001 (1)_S15	Jilin	11	DHA - Houtaomuga	III	G001	1	S15	1739	Phase 3
11 DHA III G001 (1)_S16	Jilin	11	DHA - Houtaomuga	III	G001	1	S16	930	Phase 3
11 DHA III G001 (1)_S17	Jilin	11	DHA - Houtaomuga	III	G001	1	S17	989	Phase 3
11 DHA III G001 (1)_S18	Jilin	11	DHA - Houtaomuga	III	G001	1	S18	1645	Phase 3

11 DHA III G001 (1)_S19	Jilin	11	DHA - Houtaomuga	III	G001	1	S19	1980	Phase 3
11 DHA III G001 (1)_S20	Jilin	11	DHA - Houtaomuga	III	G001	1	S20	855	Phase 3
13 DHA I F008 (1)_S1	Jilin	13	DHA - Houtaomuga	I	F008	1	S1	1230	Phase 4
13 DHA I F008 (1)_S2	Jilin	13	DHA - Houtaomuga	I	F008	1	S2	1390	Phase 4
13 DHA I F008 (1)_S3	Jilin	13	DHA - Houtaomuga	I	F008	1	S3	887	Phase 4
13 DHA I F008 (1)_S4	Jilin	13	DHA - Houtaomuga	I	F008	1	S4	779	Phase 4
14 DHA I F010 (1)_S1	Jilin	14	DHA - Houtaomuga	I	F010	1	S1	1020	Phase 4
14 DHA I F010 (1)_S2	Jilin	14	DHA - Houtaomuga	I	F010	1	S2	663	Phase 4
14 DHA I F010 (1)_S3	Jilin	14	DHA - Houtaomuga	I	F010	1	S3	1122	Phase 4
14 DHA I F010 (1)_S4	Jilin	14	DHA - Houtaomuga	I	F010	1	S4	802	Phase 4
14 DHA I F010 (1)_S5	Jilin	14	DHA - Houtaomuga	I	F010	1	S5	1198	Phase 4
14 DHA I F013 (1)_S1	Jilin	14	DHA - Houtaomuga	I	F013	1	S1	1551	Phase 4
14 DHA I F013 (1)_S2	Jilin	14	DHA - Houtaomuga	I	F013	1	S2	768	Phase 4
14 DHA I F013 (1)_S3	Jilin	14	DHA - Houtaomuga	I	F013	1	S3	709	Phase 4
14 DHA I F013 (1)_S4	Jilin	14	DHA - Houtaomuga	I	F013	1	S4	1333	Phase 4

14 DHA I F016 (1)_S1	Jilin	14	DHA - Houtaomuga	I	F016	1	S1	903	Phase 4
14 DHA I F016 (1)_S2	Jilin	14	DHA - Houtaomuga	I	F016	1	S2	1112	Phase 4
14 DHA I F016 (1)_S3	Jilin	14	DHA - Houtaomuga	I	F016	1	S3	808	Phase 4
14 DHA I F016 (1)_S4	Jilin	14	DHA - Houtaomuga	I	F016	1	S4	1300	Phase 4
14 DHA I F016 (1)_S5	Jilin	14	DHA - Houtaomuga	I	F016	1	S5	779	Phase 4
14 DHA I F016 (2)_S1	Jilin	14	DHA - Houtaomuga	I	F016	2	S1	682	Phase 4
14 DHA I F016 (2)_S2	Jilin	14	DHA - Houtaomuga	I	F016	2	S2	845	Phase 4
14 DHA I F016 (3)_S1	Jilin	14	DHA - Houtaomuga	I	F016	3	S1	1702	Phase 4
14 DHA I F016 (3)_S2	Jilin	14	DHA - Houtaomuga	I	F016	3	S2	1097	Phase 4
14 DHA I F016 (3)_S3	Jilin	14	DHA - Houtaomuga	I	F016	3	S3	830	Phase 4
14 DHA I F016 (3)_S4	Jilin	14	DHA - Houtaomuga	I	F016	3	S4	778	Phase 4
14 DHA I F017 D1 (2)_S1	Jilin	14	DHA - Houtaomuga	I	F017 D1	2	S1	577	Phase 4
14 DHA I F017 D1 (2)_S2	Jilin	14	DHA - Houtaomuga	I	F017 D1	2	S2	1186	Phase 4
14 DHA I F017 D1 (2)_S3	Jilin	14	DHA - Houtaomuga	I	F017 D1	2	S3	901	Phase 4
14 DHA I F017 D1 (2)_S4	Jilin	14	DHA - Houtaomuga	I	F017 D1	2	S4	2156	Phase 4

14 DHA I F017 D1 (2)_S5	Jilin	14	DHA - Houtaomuga	I	F017 D1	2	S5	418	Phase 4
14 DHA I F018 (2)_S1	Jilin	14	DHA - Houtaomuga	I	F018	2	S1	2100	Phase 4
15 DHA I F021 (2)_S1	Jilin	15	DHA - Houtaomuga	I	F021	2	S1	1045	Phase 4
15 DHA I F021 (2)_S2	Jilin	15	DHA - Houtaomuga	I	F021	2	S2	1564	Phase 4
15 DHA I F023 (2)_S1	Jilin	15	DHA - Houtaomuga	I	F023	2	S1	984	Phase 4
15 DHA I F023 (2)_S2	Jilin	15	DHA - Houtaomuga	I	F023	2	S2	806	Phase 4
15 DHA I F023 (2)_S3	Jilin	15	DHA - Houtaomuga	I	F023	2	S3	985	Phase 4
15 DHA I F026 (2)_S1	Jilin	15	DHA - Houtaomuga	I	F026	2	S1	798	Phase 4
15 DHA I F026 (1)_S1	Jilin	15	DHA - Houtaomuga	I	FO26	1	S1	963	Phase 4
11 DHA III H085(1)_S1	Jilin	11	DHA - Houtaomuga	III	H085	1	S1	1700	Phase 4
11 DHA III H085(1)_S2	Jilin	11	DHA - Houtaomuga	III	H085	1	S2	1300	Phase 4
11 DHA III H085(1)_S3	Jilin	11	DHA - Houtaomuga	III	H085	1	S3	1000	Phase 4
11 DHA III H085(1)_S4	Jilin	11	DHA - Houtaomuga	III	H085	1	S4	800	Phase 4
11 DHA III H085(1)_S5	Jilin	11	DHA - Houtaomuga	III	H085	1	S5	1800	Phase 4
11 DHA III H085(1)_S6	Jilin	11	DHA - Houtaomuga	III	H085	1	S6	2015	Phase 4

11 DHA III H085(1)_S7	Jilin	11	DHA - Houtaomuga	III	H085	1	S7	636	Phase 4
11 DHA III H085(1)_S8	Jilin	11	DHA - Houtaomuga	III	H085	1	S8	1098	Phase 4
11 DHA III H085(1)_S9	Jilin	11	DHA - Houtaomuga	III	H085	1	S9	192	Phase 4
11 DHA III H085(1)_S10	Jilin	11	DHA - Houtaomuga	III	H085	1	S10	1734	Phase 4

Appendix II:
All compiled IRMS data from northeast Asia

Source	Sample Number	Site and/or Phase	$\delta^{13}C_{16:0}$	$\delta^{13}C_{18:0}$	$\Delta^{13}C$	Dates
Keute 2022	12 DHA III G18 (1)_S3	Houtaomuga Phase 1	-29.2	-28.6	0.6	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (1)_S4	Houtaomuga Phase 1	-29.8	-27.7	2.1	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (1)_S5	Houtaomuga Phase 1	-30.6	-29.8	0.9	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (1)_S6	Houtaomuga Phase 1	-31.1	-29.9	1.2	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (1)_S8	Houtaomuga Phase 1	-31.2	-29.7	1.5	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (1)_S9	Houtaomuga Phase 1	-30.6	-29.5	1.2	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (2)_S3	Houtaomuga Phase 1	-30.4	-29.9	0.5	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (2)_S4	Houtaomuga Phase 1	-30.0	-29.2	0.8	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (2)_S6	Houtaomuga Phase 1	-29.5	-28.8	0.8	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (2)_S7	Houtaomuga Phase 1	-30.0	-29.4	0.7	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (2)_S9	Houtaomuga Phase 1	-29.9	-29.0	0.9	13,000-11,000 cal BP
Keute 2022	12 DHA III G18 (2)_S10	Houtaomuga Phase 1	-29.8	-29.1	0.7	13,000-11,000 cal BP

Keute 2022	12 DHA III G21 (1)_S1	Houtaomuga Phase 1	-31.3	-29.5	1.8	13,000-11,000 cal BP
Keute 2022	12 DHA III G21 (1)_S3	Houtaomuga Phase 1	-30.9	-30.0	1.0	13,000-11,000 cal BP
Keute 2022	12 DHA III G21 (1)_S4	Houtaomuga Phase 1	-31.0	-29.8	1.2	13,000-11,000 cal BP
Keute 2022	12 DHA III G21 (1)_S5	Houtaomuga Phase 1	-29.3	-29.2	0.1	13,000-11,000 cal BP
Keute 2022	12 DHA III G22 (1)_S2	Houtaomuga Phase 1	-29.3	-28.6	0.8	13,000-11,000 cal BP
Keute 2022	12 DHA III G22 (1)_S3	Houtaomuga Phase 1	-30.6	-30.0	0.6	13,000-11,000 cal BP
Keute 2022	12 DHA III G22 (1)_S5	Houtaomuga Phase 1	-30.4	-29.5	0.9	13,000-11,000 cal BP
Keute 2022	12 DHA III G22 (1)_S8	Houtaomuga Phase 1	-29.2	-29.0	0.2	13,000-11,000 cal BP
Keute 2022	12 DHA III G22 (1)_S9	Houtaomuga Phase 1	-31.4	-30.3	1.2	13,000-11,000 cal BP
Keute 2022	12 DHA III G22 (1)_S10	Houtaomuga Phase 1	-30.4	-30.1	0.4	13,000-11,000 cal BP
Keute 2022	12 DHA III H146 (1)_S2	Houtaomuga Phase 1	-30.2	-29.2	1.0	13,000-11,000 cal BP
Keute 2022	12 DHA III H146 (1)_S3	Houtaomuga Phase 1	-29.9	-29.1	0.8	13,000-11,000 cal BP
Keute 2022	12 DHA III H146 (1)_S4	Houtaomuga Phase 1	-30.1	-29.8	0.3	13,000-11,000 cal BP

Keute 2022	12 DHA III H146 (1)_S5	Houtaomuga Phase 1	-30.3	-30.1	0.2	13,000-11,000 cal BP
Keute 2022	12 DHA III H248 (1)_S1	Houtaomuga Phase 1	-29.2	-28.5	0.7	13,000-11,000 cal BP
Keute 2022	12 DHA III H248 (1)_S2	Houtaomuga Phase 1	-28.3	-27.4	0.8	13,000-11,000 cal BP
Keute 2022	12 DHA III H248 (1)_S3	Houtaomuga Phase 1	-25.4	-25.1	0.3	13,000-11,000 cal BP
Keute 2022	12 DHA III H248 (1)_S4	Houtaomuga Phase 1	-28.3	-27.3	1.0	13,000-11,000 cal BP
Keute 2022	12 DHA III H248 (1)_S5	Houtaomuga Phase 1	-28.1	-27.9	0.2	13,000-11,000 cal BP
Keute 2022	11 DHA III G002 (1)_S1	Houtaomuga Phase 2	-31.1	-30.3	0.7	8,000-7,500 cal BP
Keute 2022	11 DHA III G002 (1)_S2	Houtaomuga Phase 2	-30.3	-30.9	-0.6	8,000-7,500 cal BP
Keute 2022	11 DHA III G002 (1)_S3	Houtaomuga Phase 2	-30.4	-29.4	1.0	8,000-7,500 cal BP
Keute 2022	11 DHA III G002 (1)_S4	Houtaomuga Phase 2	-29.5	-29.0	0.5	8,000-7,500 cal BP
Keute 2022	11 DHA III G002 (1)_S5	Houtaomuga Phase 2	-29.4	-29.6	-0.2	8,000-7,500 cal BP
Keute 2022	11 DHA III G002 (1)_S6	Houtaomuga Phase 2	-29.9	-29.5	0.3	8,000-7,500 cal BP
Keute 2022	11 DHA III G002 (1)_S8	Houtaomuga Phase 2	-31.3	-31.9	-0.7	8,000-7,500 cal BP

Keute 2022	11 DHA III G002 (1)_S10	Houtaomuga Phase 2	-29.5	-29.4	0.1	8,000-7,500 cal BP
Keute 2022	11 DHA III G13_S1	Houtaomuga Phase 2	-31.0	-30.1	1.0	8,000-7,500 cal BP
Keute 2022	11 DHA III H131 (1)_S1	Houtaomuga Phase 2	-29.9	-29.8	0.1	8,000-7,500 cal BP
Keute 2022	11 DHA III H131 (1)_S2	Houtaomuga Phase 2	-29.6	-29.6	0.0	8,000-7,500 cal BP
Keute 2022	11 DHA III H131 (1)_S3	Houtaomuga Phase 2	-29.5	-29.9	-0.4	8,000-7,500 cal BP
Keute 2022	11 DHA III H131 (1)_S6	Houtaomuga Phase 2	-30.3	-30.5	-0.1	8,000-7,500 cal BP
Keute 2022	11 DHA III H131 (1)_S8	Houtaomuga Phase 2	-29.1	-29.4	-0.3	8,000-7,500 cal BP
Keute 2022	11 DHA III G001 (1)_S1	Houtaomuga Phase 3	-29.5	-28.6	1.0	6,300-5,500 cal BP
Keute 2022	11 DHA III G001 (1)_S2	Houtaomuga Phase 3	-29.9	-33.3	-3.5	6,300-5,500 cal BP
Keute 2022	11 DHA III G001 (1)_S7	Houtaomuga Phase 3	-30.0	-30.3	-0.3	6,300-5,500 cal BP
Keute 2022	11 DHA III G001 (1)_S10	Houtaomuga Phase 3	-29.8	-29.9	-0.1	6,300-5,500 cal BP
Keute 2022	13 DHA I F008 (1)_S3	Houtaomuga Phase 4	-30.4	-31.2	-0.8	~ 5,000 cal BP
Keute 2022	13 DHA I F008 (1)_S4	Houtaomuga Phase 4	-30.4	-30.4	0.1	~ 5,000 cal BP

Keute 2022	14 DHA I F010 (1)_S1	Houtaomuga Phase 4	-30.2	-30.3	-0.1	~ 5,000 cal BP
Keute 2022	14 DHA I F010 (1)_S2	Houtaomuga Phase 4	-29.6	-29.2	0.4	~ 5,000 cal BP
Keute 2022	14 DHA I F010 (1)_S3	Houtaomuga Phase 4	-30.1	-30.3	-0.2	~ 5,000 cal BP
Keute 2022	14 DHA I F013 (1)_S1	Houtaomuga Phase 4	-30.1	-30.2	-0.1	~ 5,000 cal BP
Keute 2022	14 DHA I F013 (1)_S3	Houtaomuga Phase 4	-30.4	-30.2	0.3	~ 5,000 cal BP
Keute 2022	14 DHA I F013 (1)_S4	Houtaomuga Phase 4	-30.4	-29.5	0.9	~ 5,000 cal BP
Keute 2022	14 DHA I F017 D1 (2)_S1	Houtaomuga Phase 4	-29.7	-29.4	0.3	~ 5,000 cal BP
Keute 2022	14 DHA I F017 D1 (2)_S2	Houtaomuga Phase 4	-30.6	-30.2	0.4	~ 5,000 cal BP
Keute 2022	14 DHA I F017 D1 (2)_S4	Houtaomuga Phase 4	-30.5	-30.5	0.0	~ 5,000 cal BP
Keute 2022	15 DHA I F021 (2)_S1	Houtaomuga Phase 4	-30.3	-30.1	0.2	~ 5,000 cal BP
Keute 2022	11 DHA III H085(1)_S3	Houtaomuga Phase 4	-30.8	-30.6	0.1	~ 5,000 cal BP
Keute 2022	11 DHA III H085(1)_S4	Houtaomuga Phase 4	-30.5	-30.6	-0.2	~ 5,000 cal BP
Shoda et al. 2020	GCK02	Goncharka 1	-26.2	-27.6	-1.5	Osipovka (15,070-10,650 cal BP)

Shoda et al. 2020	GCK07	Goncharka 1	-28.6	-29.3	-0.7	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	GCK08	Goncharka 1	-26.9	-27.9	-1	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	GCK09	Goncharka 1	-24.4	-24.5	0	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	GCK10	Goncharka 1	-28.4	-28.1	0.3	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	Amur 1	Goncharka 1	-26.7	-27.5	-0.8	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	Amur 2	Goncharka 1	-28.1	-28.7	-0.6	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	Amur3	Goncharka 1	-25.5	-26.3	-0.8	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	Amur4	Goncharka 1	-24.6	-25.7	-1.1	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	Amur 5	Goncharka 1	-25	-26	-1	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	Amur 6	Goncharka 1	-30.2	-29.8	0.4	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	Amur 7	Goncharka 1	-29.4	-29.4	0	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	Amur 8	Goncharka 1	-29.3	-29.3	0.1	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	Amur 9	Goncharka 1	-27.6	-28.5	-0.9	Osipovka (15,070-10,650 cal BP)

Shoda et al. 2020	Amur 10	Goncharka 1	-24.6	-23.1	1.5	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	Amur 11	Goncharka 1	-29.7	-29.5	0.2	Osipovka (15,070-10,650 cal BP)
Shoda et al. 2020	GMT01	Gromatukha	-28	-30.1	-2	Gromatukha (15,120-13,840 cal BP)
Shoda et al. 2020	GMT02	Gromatukha	-27.6	-29	-1.4	Gromatukha (15,120-13,840 cal BP)
Shoda et al. 2020	GMT04	Gromatukha	-29.2	-30.4	-1.2	Gromatukha (15,120-13,840 cal BP)
Shoda et al. 2020	GMT06	Gromatukha	-28.3	-29.9	-1.6	Gromatukha (15,120-13,840 cal BP)
Gibbs et al. 2017	85	Slavnaya 4	-22.34	-22.48	-0.1	Early Neolithic (7300–7050 cal BC)
Gibbs et al. 2017	87	Slavnaya 4	-20.11	-17.7	2.4	Early Neolithic(7300–7050 cal BC)
Gibbs et al. 2017	96	Slavnaya 4	-20.2	-19.14	1.1	Early Neolithic (7300–7050 cal BC)
Gibbs et al. 2017	103	Slavnaya 5	-18.95	-18.06	0.9	Early Neolithic (6830–6070 cal BC)
Gibbs et al. 2017	104	Slavnaya 5	-23.32	-22.83	0.5	Early Neolithic (6830–6070 cal BC)
Gibbs et al. 2017	108	Slavnaya 4	-23.45	-23.53	-0.1	Middle Neolithic (6200–5000 cal BC)
Gibbs et al. 2017	109	Slavnaya 4	-24.4	-23.13	1.3	Middle Neolithic (6200–5000 cal BC)

Gibbs et al. 2017	110	Slavnaya 4	-23.13	-23.85	-0.7	Middle Neolithic (6200–5000 cal BC)
Gibbs et al. 2017	112	Slavnaya 4	-23.16	-23.32	-0.2	Middle Neolithic (6200–5000 cal BC)
Gibbs et al. 2017	114	Slavnaya 4	-23.8	-22.61	1.2	Middle Neolithic (6200–5000 cal BC)
Gibbs et al. 2017	119	Chaivo 6	-23.2	-22.06	1.1	Middle Neolithic (6000–5630 cal BC)
Lucquin et al. 2018	KSJ-253	Kenshojo	-25.6	-24.9	0.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-254	Kenshojo	-28.6	-24.7	3.9	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-428	Kenshojo	-29.1	-28.4	0.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-429	Kenshojo	-28.8	-28.5	0.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-430	Kenshojo	-28.8	-27.5	1.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-431	Kenshojo	-29.3	-28.3	1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-432	Kenshojo	-28.7	-29.2	-0.5	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-433	Kenshojo	-28.1	-27.5	0.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-434	Kenshojo	-27.6	-27.1	0.5	Incipient Jomon (16,000–11,500 cal B.P.)

Lucquin et al. 2018	KSJ-435	Kenshojo	-27.9	-28.9	-1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-436	Kenshojo	-28.2	-28.9	-0.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-437	Kenshojo	-29.7	-30.6	-0.9	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-438	Kenshojo	-29.6	-30.3	-0.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-82	Kenshojo	-26.4	-25.5	0.8	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-83	Kenshojo	-25.8	-24.6	1.1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-84	Kenshojo	-26.5	-26.3	0.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-85	Kenshojo	-25.7	-25.3	0.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-86	Kenshojo	-28.6	-28	0.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-87	Kenshojo	-26.2	-25	1.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-88	Kenshojo	-27.3	-26.8	0.5	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-89	Kenshojo	-31.6	-27.7	3.8	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-90	Kenshojo	-28.8	-28.8	0	Incipient Jomon (16,000–11,500 cal B.P.)

Lucquin et al. 2018	KSJ-91 (81)	Kenshojo	-31	-26.6	4.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC01	Yukura Cave	-25.3	-25.3	0	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC02	Yukura Cave	-24.9	-25.2	-0.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC03	Yukura Cave	-25.8	-25.8	0	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC05	Yukura Cave	-26.1	-25.9	0.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC06	Yukura Cave	-24.3	-24.1	0.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC07	Yukura Cave	-26.7	-27.5	-0.8	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC08	Yukura Cave	-24.8	-24.5	0.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC09	Yukura Cave	-28.3	-31.4	-3.1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC10	Yukura Cave	-26.2	-27.9	-1.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC11	Yukura Cave	-26.6	-25.3	1.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC12	Yukura Cave	-25.6	-26.9	-1.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC13	Yukura Cave	-26.8	-28.9	-2.1	Incipient Jomon (16,000–11,500 cal B.P.)

Lucquin et al. 2018	YKC14	Yukura Cave	-26.4	-28.7	-2.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC15	Yukura Cave	-25.8	-28.4	-2.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC16	Yukura Cave	-27.4	-31.6	-4.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC17	Yukura Cave	-28	-30.7	-2.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YKC18	Yukura Cave	-25.1	-25.5	-0.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR01	Ishihata	-27	-29.8	-2.9	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR02	Ishihata	-24.7	-28.9	-4.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR04	Ishihata	-26.7	-29.4	-2.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR05	Ishihata	-25.1	-26.7	-1.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR06	Ishihata	-24.8	-27.7	-2.9	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR07	Ishihata	-26.5	-28.1	-1.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR08	Ishihata	-27.9	-31.1	-3.1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR09	Ishihata	-24.6	-26.2	-1.7	Incipient Jomon (16,000–11,500 cal B.P.)

Lucquin et al. 2018	IHR10	Ishihata	-23.8	-25.4	-1.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR11	Ishihata	-25.8	-29.5	-3.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR12	Ishihata	-25.2	-27.3	-2.1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	IHR14	Ishihata	-25.1	-26.5	-1.5	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YHS01	Shaguchi	-29.1	-29.3	-0.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YHS02	Shaguchi	-26.4	-27.3	-0.9	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YHS03	Shaguchi	-27.8	-28.3	-0.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YHS04	Shaguchi	-25.7	-25.7	0.1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YHS05	Shaguchi	-26.6	-27.5	-0.9	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YHS06	Shaguchi	-28.3	-29.3	-1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YHS07	Shaguchi	-28.9	-29.4	-0.5	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	YHS08	Shaguchi	-28.6	-29.1	-0.5	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ02	Shimojuku	-26.8	-27.9	-1.1	Incipient Jomon (16,000–11,500 cal B.P.)

Lucquin et al. 2018	SMJ04	Shimojuku	-24.9	-24.5	0.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ05	Shimojuku	-25.4	-28.6	-3.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ06	Shimojuku	-25.1	-24.6	0.5	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ07	Shimojuku	-24.9	-27.4	-2.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ08	Shimojuku	-25	-25.8	-0.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ10	Shimojuku	-25.6	-28.2	-2.5	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ12	Shimojuku	-24.8	-23.8	1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ13	Shimojuku	-25.3	-24.4	0.9	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ14	Shimojuku	-25.3	-27.3	-2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ15	Shimojuku	-25.9	-27.8	-1.9	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SMJ16	Shimojuku	-25.4	-26.8	-1.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	TGI01	Ichinotsuka	-27.1	-28.5	-1.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	TGI02	Ichinotsuka	-28.5	-28.9	-0.5	Incipient Jomon (16,000–11,500 cal B.P.)

Lucquin et al. 2018	TGI03	Ichinotsuka	-29	-29	0	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	TGI04	Ichinotsuka	-28.7	-28.5	0.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SSK101	Ohshikakubo	-28.8	-31.3	-2.5	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SSK102	Ohshikakubo	-29	-29.2	-0.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SSK103	Ohshikakubo	-28.9	-31.3	-2.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SSK104	Ohshikakubo	-29.3	-29.4	0	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	SSK105	Ohshikakubo	-26.4	-29.4	-3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T66Fi	Torihama	-28.8	-31	-2.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T68Fi	Torihama	-26.2	-25.5	0.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T69Fe	Torihama	-22.4	-22.2	0.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T73Fi	Torihama	-25.6	-25.5	0.1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T74Fi	Torihama	-24.2	-24.1	0.1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T77Fi	Torihama	-24.3	-24.2	0.1	Incipient Jomon (16,000–11,500 cal B.P.)

Lucquin et al. 2018	T124Fi	Torihama	-24.6	-23.6	1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T125Fi	Torihama	-23.8	-23.5	0.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T126Fi	Torihama	-25.5	-24.4	1.1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T130Fi	Torihama	-25.2	-24.1	1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T133Fi	Torihama	-24.1	-24.1	0	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T134Fi	Torihama	-24.6	-25.8	-1.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T135Fi	Torihama	-24.3	-24.2	0.1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T136Fi	Torihama	-24.2	-23.8	0.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T143Fe	Torihama	-24.9	-25.1	-0.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T144Fi	Torihama	-24.8	-24.4	0.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T145Fe	Torihama	-24.4	-24.3	0.1	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T146Fi	Torihama	-26.4	-27	-0.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T147Fe	Torihama	-24.3	-23.9	0.4	Incipient Jomon (16,000–11,500 cal B.P.)

Lucquin et al. 2018	T148Fi	Torihama	-24	-24.8	-0.8	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T149Fi	Torihama	-23.8	-24.1	-0.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T151Fi	Torihama	-27.9	-28.2	-0.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T152Fi	Torihama	-24.8	-26.5	-1.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T391Fi	Torihama	-25.3	-25.1	0.2	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T391S	Torihama	-24.6	-25.2	-0.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T392S	Torihama	-26.7	-27.4	-0.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T393S	Torihama	-25.9	-26.5	-0.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T395Fi	Torihama	-25.3	-23.9	1.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T395S	Torihama	-27.3	-28	-0.7	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T396Fi	Torihama	-25	-25.5	-0.5	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T396S	Torihama	-26.7	-28.2	-1.5	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T420S	Torihama	-24.3	-26	-1.7	Incipient Jomon (16,000–11,500 cal B.P.)

Lucquin et al. 2018	T421S	Torihama	-26.3	-28.7	-2.4	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T443Fi	Torihama	-22.8	-26.1	-3.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T444Fe	Torihama	-23.5	-25.1	-1.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	T445Fi	Torihama	-24.7	-26.3	-1.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KT3_17d_8b_F	Taisho 3	-25.4	-23.8	1.6	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KT3-5_F	Taisho 3	-21.8	-23.1	-1.3	Incipient Jomon (16,000–11,500 cal B.P.)
Lucquin et al. 2018	KSJ-01	11,500 B.P.)	-28.4	-28.1	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-02	Kenshojo	-25	-25.1	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-03	Kenshojo	-29.2	-29.6	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-04	Kenshojo	-28.1	-27.9	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-05	Kenshojo	-23.9	-26.6	-2.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-06	Kenshojo	-24.5	-24.8	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-07	Kenshojo	-23.4	-23.9	-0.5	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	KSJ-08	Kenshojo	-27.9	-28.4	-0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-09	Kenshojo	-28.4	-28.5	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-10	Kenshojo	-21.9	-23.3	-1.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-11	Kenshojo	-27.9	-28.2	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-12	Kenshojo	-26.6	-27.2	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-13	Kenshojo	-25.2	-26	-0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-14	Kenshojo	-28.4	-27.9	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-15	Kenshojo	-20.3	-22.6	-2.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-16	Kenshojo	-19.7	-23.4	-3.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-17	Kenshojo	-25.2	-26.4	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-18	Kenshojo	-25.4	-27	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-19	Kenshojo	-23.9	-24.5	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-20	Kenshojo	-23.3	-24.5	-1.2	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	KSJ-21	Kenshojo	-25.2	-26.5	-1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-22	Kenshojo	-22.2	-23.3	-1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-23	Kenshojo	-22.1	-21.8	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-24	Kenshojo	-24.3	-26	-1.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-25	Kenshojo	-27.1	-27.8	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-26	Kenshojo	-25.6	-25.7	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-27	Kenshojo	-20.1	-22.3	-2.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-28	Kenshojo	-26.1	-26.3	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-29	Kenshojo	-27.9	-28.5	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-30	Kenshojo	-24.7	-24.4	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-31	Kenshojo	-27.4	-27.4	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-32	Kenshojo	-28.4	-29.1	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-33	Kenshojo	-27.4	-25.4	2	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	KSJ-34	Kenshojo	-25	-25.6	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-35	Kenshojo	-20.2	-22	-1.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-36	Kenshojo	-25.2	-27	-1.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-37	Kenshojo	-25	-26.8	-1.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-38	Kenshojo	-28.8	-29.1	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-39	Kenshojo	-29.7	-29.2	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-40	Kenshojo	-25.6	-24.9	0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-41	Kenshojo	-23.3	-23.6	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-42	Kenshojo	-22.4	-23.1	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-420	Kenshojo	-29.8	-29.2	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-421	Kenshojo	-30.2	-31.1	-0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-422	Kenshojo	-29.1	-29	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-423	Kenshojo	-24	-25.6	-1.5	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	KSJ-424	Kenshojo	-25.2	-28.1	-2.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-425	Kenshojo	-24.7	-25.8	-1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-426	Kenshojo	-21.6	-24	-2.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-427	Kenshojo	-26.8	-26.8	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-43	Kenshojo	-27.4	-27	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-44	Kenshojo	-22.2	-23.7	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-45	Kenshojo	-25.5	-26.7	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-46	Kenshojo	-23.1	-26.6	-3.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-47	Kenshojo	-29.1	-28.7	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-48	Kenshojo	-26.4	-25.9	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-49	Kenshojo	-28.8	-28.7	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-50	Kenshojo	-28.8	-27.5	1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-51	Kenshojo	-25.8	-25.4	0.4	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	KSJ-52	Kenshojo	-20	-20.8	-0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-53	Kenshojo	-18.2	-20.8	-2.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-54	Kenshojo	-26.8	-26	0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-55	Kenshojo	-25.5	-25.4	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-56	Kenshojo	-29.5	-28.4	1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-57	Kenshojo	-22.6	-23.8	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-58	Kenshojo	-28.3	-27.1	1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-59	Kenshojo	-23.8	-22.5	1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-60	Kenshojo	-28.9	-28.9	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-61	Kenshojo	-26.1	-26.6	-0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-62	Kenshojo	-25.4	-26.3	-0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-63	Kenshojo	-28.9	-28.4	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-64	Kenshojo	-26.4	-26.7	-0.3	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	KSJ-65	Kenshojo	-24.5	-24.6	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-66	Kenshojo	-24	-24.2	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-67	Kenshojo	-23.1	-21.3	1.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-68	Kenshojo	-22.6	-23.2	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-69	Kenshojo	-27.4	-28	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-70	Kenshojo	-26	-24.8	1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-71	Kenshojo	-26.9	-27.6	-0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-72	Kenshojo	-28.4	-27.3	1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-73	Kenshojo	-22.8	-25.3	-2.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-74	Kenshojo	-26	-25.4	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-75	Kenshojo	-21.4	-23.7	-2.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-76	Kenshojo	-22.6	-20.8	1.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-77	Kenshojo	-25.8	-26.7	-0.9	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	KSJ-78	Kenshojo	-25.2	-24.9	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-79	Kenshojo	-22.7	-23.2	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KSJ-80	Kenshojo	-24.9	-26.3	-1.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC31	Yukura Cave	-27.1	-30	-2.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC32	Yukura Cave	-27.4	-28.6	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC33	Yukura Cave	-26.7	-28.2	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC34	Yukura Cave	-28.9	-31.2	-2.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC35	Yukura Cave	-27.9	-28.5	-0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC36	Yukura Cave	-28	-28.8	-0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC37	Yukura Cave	-25.4	-26.1	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC38	Yukura Cave	-26.6	-28.7	-2.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC41	Yukura Cave	-27	-29.9	-2.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC42	Yukura Cave	-28.6	-30.3	-1.7	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	YKC43	Yukura Cave	-25.7	-26.7	-1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC44	Yukura Cave	-26.8	-29.8	-2.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC45	Yukura Cave	-27.6	-29.1	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC46	Yukura Cave	-25.4	-29.4	-4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC47	Yukura Cave	-25.6	-27.4	-1.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC48	Yukura Cave	-29.6	-31.6	-2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC49	Yukura Cave	-27.6	-29.5	-2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC50	Yukura Cave	-25.7	-25.9	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC51	Yukura Cave	-27.4	-26.9	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	YKC53	Yukura Cave	-26.7	-29.5	-2.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TRK01	Torikake-Nishi	-28.1	-27.3	0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TRK02	Torikake-Nishi	-26.4	-25.7	0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TRK03	Torikake-Nishi	-27.4	-26	1.4	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	TRK04	Torikake-Nishi	-27.1	-26.6	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TRK07	Torikake-Nishi	-28.2	-30.8	-2.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TRK09	Torikake-Nishi	-28.5	-28.2	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TRK10	Torikake-Nishi	-29.7	-29.4	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TRK11	Torikake-Nishi	-29.1	-28.6	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TRK12	Torikake-Nishi	-28.2	-30.1	-1.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TRK13	Torikake-Nishi	-28.5	-28.8	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	EKK01	Kamikuroiwa	-27.5	-26.9	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	EKK02	Kamikuroiwa	-28.3	-31.5	-3.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	EKK03	Kamikuroiwa	-27.2	-28.3	-1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	EKK05	Kamikuroiwa	-27	-29.1	-2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	EKK06	Kamikuroiwa	-26.9	-28.6	-1.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO01	Nishinojo	-25.6	-25.5	0.2	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	NJO02	Nishinojo	-22.6	-23.8	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO04	Nishinojo	-23	-23.4	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO05	Nishinojo	-24.3	-24.3	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO06	Nishinojo	-23.3	-24	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO07	Nishinojo	-23.9	-23.9	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO08	Nishinojo	-25.8	-25.7	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO09	Nishinojo	-28	-26.9	1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO10	Nishinojo	-27.7	-27	0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO11	Nishinojo	-26.7	-26.6	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO12	Nishinojo	-27.8	-30	-2.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO15	Nishinojo	-19.5	-21.8	-2.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO16	Nishinojo	-25.3	-24.5	0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO17	Nishinojo	-28.4	-28.9	-0.5	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	NJO18	Nishinojo	-26.5	-27	-0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO19	Nishinojo	-25.2	-25.4	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NJO20	Nishinojo	-27.8	-27.9	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOK1	Tokisaki	-23.8	-24.7	-0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOK2	Tokisaki	-26.3	-28.1	-1.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOK3	Tokisaki	-26.2	-26	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOK4	Tokisaki	-27.1	-27.7	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOK7	Tokisaki	-25.8	-27.7	-1.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOK8	Tokisaki	-26	-27.9	-1.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOK9	Tokisaki	-26.2	-26.6	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOK11	Tokisaki	-25.9	-26	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOK12	Tokisaki	-25.9	-28.2	-2.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOK13	Tokisaki	-26.1	-26.6	-0.5	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	TOK14	Tokisaki	-25.4	-25.5	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI05	Ichinotsuka	-28.9	-29.2	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI06	Ichinotsuka	-26.8	-28.1	-1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI07	Ichinotsuka	-28.2	-28.3	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI09	Ichinotsuka	-26.6	-28.3	-1.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI11	Ichinotsuka	-27.5	-28.3	-0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI12	Ichinotsuka	-28.2	-27.8	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI13A	Ichinotsuka	-27	-28.3	-1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI13B	Ichinotsuka	-27	-27.5	-0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI15	Ichinotsuka	-29.3	-28.6	0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI16	Ichinotsuka	-29.4	-29.3	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI17	Ichinotsuka	-29.5	-29.5	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI18	Ichinotsuka	-28.9	-29.3	-0.4	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	TGI19	Ichinotsuka	-28.3	-29.3	-1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI20	Ichinotsuka	-29.3	-29.2	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI21A	Ichinotsuka	-28.9	-29.2	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI21B	Ichinotsuka	-27.9	-28.6	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI23	Ichinotsuka	-27.7	-28.6	-0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI25	Ichinotsuka	-26.4	-25.7	0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI29	Ichinotsuka	-27.1	-28.1	-1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI30	Ichinotsuka	-27.7	-28.6	-1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI31	Ichinotsuka	-28	-28.9	-0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI32A	Ichinotsuka	-27.6	-28.3	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI32B	Ichinotsuka	-27.8	-28.1	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI33A	Ichinotsuka	-29.1	-28.6	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI33B	Ichinotsuka	-30.1	-29.9	0.2	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	TGI33C	Ichinotsuka	-29.9	-29.7	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI34A	Ichinotsuka	-28.8	-29.2	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI34B	Ichinotsuka	-27.7	-28.1	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI34C	Ichinotsuka	-28.4	-28.7	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI34D	Ichinotsuka	-29.9	-29.6	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI34E	Ichinotsuka	-30	-29.4	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI34F	Ichinotsuka	-29.3	-29.6	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI35A	Ichinotsuka	-29.5	-29.4	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI35B	Ichinotsuka	-29.8	-29.7	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI35C	Ichinotsuka	-30.1	-29.9	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI36A	Ichinotsuka	-30.3	-29.9	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI36B	Ichinotsuka	-29.7	-29.5	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI36D	Ichinotsuka	-29.4	-29.3	0.1	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	TGI36E	Ichinotsuka	-29	-29.3	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI36F	Ichinotsuka	-29.1	-29.4	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI36G	Ichinotsuka	-29.6	-29.7	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI37A	Ichinotsuka	-29.7	-29.7	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI37B	Ichinotsuka	-28.8	-29.3	-0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI38A	Ichinotsuka	-29.2	-29.6	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI38B	Ichinotsuka	-29.3	-29.1	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI38C	Ichinotsuka	-29.1	-29.4	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI38D	Ichinotsuka	-28.4	-28.7	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI38E	Ichinotsuka	-29.1	-29.2	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI39A	Ichinotsuka	-28.3	-28	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI41A	Ichinotsuka	-29.8	-29.7	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI41B	Ichinotsuka	-28.8	-29.2	-0.4	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	TGI42A	Ichinotsuka	-30	-29.6	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI42B	Ichinotsuka	-30.4	-29.9	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI42C	Ichinotsuka	-30.1	-29.7	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI42D	Ichinotsuka	-29.9	-29.8	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI42E	Ichinotsuka	-30	-31.9	-1.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI42F	Ichinotsuka	-27.8	-28.9	-1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI42G	Ichinotsuka	-29.7	-29.8	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI43	Ichinotsuka	-27.4	-28	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGI44	Ichinotsuka	-28.3	-28.5	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGU45	Utsunomiya Seiryo highschool	-26	-27	-1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGU46	Utsunomiya Seiryo highschool	-26.4	-27.4	-1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGU47A	Utsunomiya Seiryo highschool	-26.1	-27.6	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGU47B	Utsunomiya Seiryo highschool	-26.6	-27.8	-1.2	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	TGU48A	Utsunomiya Seiryo highschool	-26.1	-27.2	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGU48B	Utsunomiya Seiryo highschool	-26.1	-26.8	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGU48C	Utsunomiya Seiryo highschool	-27.2	-27.6	-0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGY49	Yamazaki-Kita	-27.5	-27.7	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGY50A	Yamazaki-Kita	-29.5	-29.8	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGY50B	Yamazaki-Kita	-27.2	-29.5	-2.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TGY50C	Yamazaki-Kita	-28.8	-29	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	AW-H24_1_F	Awazu	-28.4	-28	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	AW-H24_23_F	Awazu	-30.1	-27.7	2.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	AW-H24_33_F	Awazu	-28.3	-26.1	2.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	AW-H24_61_F	Awazu	-26.4	-26.3	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	AW-H24_83_F	Awazu	-28.9	-27	1.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	AW-H24_84_F	Awazu	-28.2	-30.4	-2.3	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	HAI-1	Haizuka	-31.7	-32.8	-1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HAI-2	Haizuka	-28.1	-29.4	-1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HAI-3	Haizuka	-26.8	-26.4	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HAI-4	Haizuka	-29.7	-29.8	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HAI-5	Haizuka	-26.3	-28	-1.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HAI-6	Haizuka	-31.7	-29.4	2.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HAI-7	Haizuka	-28.2	-28.6	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HAI-8	Haizuka	-28.9	-28.6	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HAI-9	Haizuka	-19.2	-24.7	-5.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HAI-10	Haizuka	-27.3	-30.1	-2.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SET-1	Setakitsunezuka	-29	-28.9	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SET-2	Setakitsunezuka	-30.2	-30.2	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SET-3	Setakitsunezuka	-30.3	-30	0.3	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	SET-4	Setakitsunezuka	-30.3	-30.1	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SET-5	Setakitsunezuka	-30.1	-30.1	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SET-6	Setakitsunezuka	-30.2	-30.1	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SET-7	Setakitsunezuka	-27.5	-29.1	-1.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SET-8	Setakitsunezuka	-30.3	-29.7	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SET-9	Setakitsunezuka	-29.8	-29.9	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SET-10	Setakitsunezuka	-28.1	-27.9	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TEN-1	Tendogao	-29.9	-29.7	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TEN-2	Tendogao	-28.1	-27.2	0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TEN-3	Tendogao	-30.3	-29.7	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TEN-4	Tendogao	-28.6	-28.1	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TEN-5	Tendogao	-27.4	-29.3	-2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TEN-6	Tendogao	-28.7	-29.1	-0.4	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	TEN-7	Tendogao	-30.2	-30.5	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TEN-8	Tendogao	-29.4	-29.4	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TEN-9	Tendogao	-29.3	-30	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TEN-10	Tendogao	-30.3	-30.1	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TON-1	Tonohira	-29.4	-29.6	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TON-2	Tonohira	-27.1	-27.4	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TON-3	Tonohira	-30.5	-30	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TON-4	Tonohira	-27.6	-28.2	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TON-5	Tonohira	-30.9	-29.9	1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TON-6	Tonohira	-30.3	-29.7	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TON-7	Tonohira	-31	-30.2	0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TON-8	Tonohira	-30.5	-30.3	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TON-9	Tonohira	-29.2	-28.7	0.5	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	TON-10	Tonohira	-30.3	-30	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-16	Jozuka	-29	-29.2	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-31	Jozuka	-29.2	-30.7	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-99	Jozuka	-30.6	-30	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-190	Jozuka	-29.6	-30	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-331	Jozuka	-29.4	-30.2	-0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-2683	Jozuka	-27.7	-28.8	-1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-4197	Jozuka	-29.4	-29	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-4675	Jozuka	-27.1	-26	1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-4873	Jozuka	-27.8	-27.5	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-4897	Jozuka	-30.5	-29.9	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-7583	Jozuka	-29.2	-29.5	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-8959	Jozuka	-29.9	-29.4	0.5	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	JOZ-18519	Jozuka	-30.3	-29.8	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-18595	Jozuka	-27.1	-28.4	-1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-20028	Jozuka	-29.1	-30.1	-1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-20814	Jozuka	-30.8	-30.7	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-43824	Jozuka	-30.7	-30.1	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-45557	Jozuka	-29.9	-30.8	-0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-46672	Jozuka	-30.5	-29.7	0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-48728	Jozuka	-30.1	-29.1	1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-51663	Jozuka	-30	-30.5	-0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-52303	Jozuka	-27.9	-28.3	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-53755	Jozuka	-29.9	-29.9	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-53806	Jozuka	-29.6	-30.7	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-60222	Jozuka	-29.4	-30.1	-0.6	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	JOZ-281XX	Jozuka	-27.8	-28.8	-1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-601X3	Jozuka	-30.4	-28.6	1.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	JOZ-7X90	Jozuka	-29.5	-30	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NAG-1039	Nagasakobira	-29.6	-29.7	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NAG-1341	Nagasakobira	-29.5	-29.6	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NAG-4536	Nagasakobira	-29.2	-29.6	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NAG-5100	Nagasakobira	-29.5	-29	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NAG-5862	Nagasakobira	-28.8	-29.7	-0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NAG-6005	Nagasakobira	-29.1	-29.6	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NAG-6039	Nagasakobira	-27.6	-29.3	-1.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NAG-6540	Nagasakobira	-29.4	-29.8	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NAG-6714	Nagasakobira	-29	-29.7	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NAG-7784	Nagasakobira	-28.7	-29.5	-0.8	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	KM-508F	Kiriki Mimitori	-27.4	-30.1	-2.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-28	Maebaru	-27.7	-28.4	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-2915	Maebaru	-27.5	-27.8	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-3097	Maebaru	-25	-25.3	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-3470	Maebaru	-29.2	-29.1	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-3689	Maebaru	-27.9	-30	-2.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-44079	Maebaru	-26.8	-28.5	-1.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-4710	Maebaru	-24.2	-25.5	-1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-4765	Maebaru	-29.5	-29.1	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-4971	Maebaru	-25	-26.4	-1.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-5327	Maebaru	-29.1	-28.9	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-6203	Maebaru	-26.3	-26.5	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-6462	Maebaru	-27.4	-27.8	-0.4	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	MAE-7169	Maebaru	-27	-28.3	-1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-7232	Maebaru	-29	-29.1	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-7780	Maebaru	-25.7	-25.5	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-8880	Maebaru	-28.1	-28.4	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-8955	Maebaru	-26	-25.7	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-9240	Maebaru	-17.2	-23.6	-6.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-11458	Maebaru	-29.2	-29.4	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-11609	Maebaru	-27.2	-28.2	-1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-16196	Maebaru	-29.4	-29.1	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-17443	Maebaru	-29.9	-29.6	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-18039	Maebaru	-20.8	-24	-3.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-28427	Maebaru	-27.9	-29.2	-1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-32110	Maebaru	-24.1	-24.3	-0.2	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	MAE-35921	Maebaru	-28.2	-29.8	-1.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-42995	Maebaru	-23.3	-24.8	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-44027	Maebaru	-23.2	-23.4	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-45150	Maebaru	-28.7	-28.8	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-49328	Maebaru	-25.1	-26.8	-1.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-49378	Maebaru	-24.8	-26.6	-1.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-51293	Maebaru	-23.1	-26	-3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-53336	Maebaru	-26.1	-26.4	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-53602	Maebaru	-25.3	-26.6	-1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-56171	Maebaru	-26.6	-29.8	-3.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-57694	Maebaru	-27	-24.1	2.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-59537	Maebaru	-28	-28.6	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-60697	Maebaru	-27.1	-29.4	-2.3	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	MAE-61009	Maebaru	-26.9	-28.2	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	MAE-29B	Maebaru	-24.1	-25.2	-1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	UEN-266	Uenohara	-28.5	-28.6	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	UEN-487	Uenohara	-23.2	-25.1	-1.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	UEN-1157	Uenohara	-28	-28.3	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	UEN-4718	Uenohara	-26.6	-25.1	1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	UEN-5658	Uenohara	-29.9	-29.7	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	UEN-12037	Uenohara	-29.9	-29.8	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	UEN-260d	Uenohara	-27.4	-25.7	1.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	UEN-258d	Uenohara	-26.3	-28.9	-2.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ01	Shobuzaki	-29.3	-28.9	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ02	Shobuzaki	-26.4	-27.6	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ03	Shobuzaki	-24.6	-25.8	-1.2	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	SBZ04	Shobuzaki	-24.7	-26.1	-1.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ05	Shobuzaki	-27.3	-27.7	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ06	Shobuzaki	-23.3	-24.8	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ07	Shobuzaki	-25.7	-25.9	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ08	Shobuzaki	-26.2	-26	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ09	Shobuzaki	-29.1	-29.1	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ10	Shobuzaki	-24.1	-25.3	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ11	Shobuzaki	-23.3	-24.4	-1.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ12	Shobuzaki	-28	-29.4	-1.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ13	Shobuzaki	-24.3	-26.3	-2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	SBZ14	Shobuzaki	-28.8	-30.4	-1.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM181S	Higashimyou	-24.6	-25	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM478F	Higashimyou	-26	-27	-1	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	HGM011F	Higashimyou	-22.2	-24.1	-1.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM324F	Higashimyou	-29.4	-29.7	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM01	Higashimyou	-30	-29.4	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM02	Higashimyou	-30	-29.8	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM03	Higashimyou	-30.4	-29.6	0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM04	Higashimyou	-31.6	-29.1	2.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM05	Higashimyou	-29.9	-32.4	-2.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM06	Higashimyou	-21.1	-20.3	0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM08	Higashimyou	-28.3	-23.7	4.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM10	Higashimyou	-24	-28.3	-4.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM11	Higashimyou	-23.5	-23.8	-0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM12	Higashimyou	-29.4	-29.2	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM13	Higashimyou	-26	-25.4	0.6	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	HGM15	Higashimyou	-23.8	-25.3	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM16	Higashimyou	-27.9	-27.3	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM17	Higashimyou	-25.7	-25.3	0.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM18	Higashimyou	-29.9	-29.2	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM19	Higashimyou	-28.5	-29.1	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM20	Higashimyou	-30	-29.2	0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM21	Higashimyou	-25.2	-26	-0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM22	Higashimyou	-26.5	-29.5	-3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM23	Higashimyou	-28	-28.6	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM24	Higashimyou	-25.7	-26.1	-0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM26	Higashimyou	-29.9	-28	1.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM27	Higashimyou	-29.8	-29.3	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM28	Higashimyou	-19.1	-21.6	-2.5	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	HGM29	Higashimyou	-27.5	-29.3	-1.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM30	Higashimyou	-28	-29.4	-1.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM31	Higashimyou	-24.1	-25.5	-1.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM32	Higashimyou	-23.9	-25.4	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM33	Higashimyou	-27.6	-27.9	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM34	Higashimyou	-27.5	-29.1	-1.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM35	Higashimyou	-28.5	-28.1	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM38	Higashimyou	-26.2	-25.8	0.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM39	Higashimyou	-26.9	-26.6	0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM40	Higashimyou	-27.8	-27.9	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM03F	Higashimyou	-29.9	-29.8	0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM11F	Higashimyou	-23.5	-24.2	-0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	HGM13F	Higashimyou	-27	-26.4	0.7	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	T397S	Torihama	-28.5	-29.1	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T398S	Torihama	-28.6	-28.7	-0.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T268Fi	Torihama	-25.7	-24.3	1.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T270Fi	Torihama	-27.1	-29.1	-2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T271Fi	Torihama	-25.3	-22	3.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T272Fi	Torihama	-25.7	-27.2	-1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T274Fi	Torihama	-25.3	-26	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T275Fi	Torihama	-26.9	-27.5	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T446S	Torihama	-28.3	-26.8	1.5	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T447S	Torihama	-30.7	-30.5	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T448S	Torihama	-29.7	-33.1	-3.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T399S	Torihama	-28.5	-29.4	-0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	T400S	Torihama	-30.4	-29.9	0.5	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	KAK-05	Kakkumi	-26.6	-24.9	1.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-07	Kakkumi	-23.5	-24.2	-0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-08	Kakkumi	-26.1	-27.5	-1.4	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-12	Kakkumi	-24	-24.6	-0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-13	Kakkumi	-27	-28.2	-1.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-14	Kakkumi	-23.1	-23.4	-0.3	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-15	Kakkumi	-26.8	-28.6	-1.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-16	Kakkumi	-22.9	-22	0.8	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-17	Kakkumi	-23.4	-23.2	0.2	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-18	Kakkumi	-23.9	-23.7	-0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-19	Kakkumi	-22.9	-22.9	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-20	Kakkumi	-22.2	-21.9	0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KAK-21	Kakkumi	-24.6	-25.1	-0.3	Initial Jomon (11,500 - 7,000 B.P.)

Lucquin et al. 2018	KAK-22	Kakkumi	-23.3	-23.8	0	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	NIS236	Nisshin 3	-23.3	-22.4	0.9	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KT3-J32	Taisho 3	-19.1	-21.2	-2.1	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	KT3-J34	Taisho 3	-18	-17.4	0.6	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOS188	Tosamupolo	-21.9	-21.2	0.7	Initial Jomon (11,500 - 7,000 B.P.)
Lucquin et al. 2018	TOS189	Tosamupolo	-21.7	-20.7	1.1	Initial Jomon (11,500 - 7,000 B.P.)

Appendix III:
Reference IRMS data from northeast Asia used for comparison with archaeological results

Class	Type	Taxa	$\delta^{13}C_{16:0}$	$\delta^{13}C_{18:0}$	Reference
Boar	Wild boar	Sus scrofa	-26.13	-26.57	Craig et al. 2013
Boar	Wild boar	Sus scrofa	-27.71	-27.28	Craig et al. 2013
Boar	Wild boar	Sus scrofa	-28.33	-27.21	Lucquin et al 2016
Boar	Wild boar	Sus scrofa	-28.13	-28.87	Lucquin et al 2016
Boar	Wild boar	Sus scrofa	-27.75	-27.9	Lucquin et al 2016
Boar	Wild boar	Sus scrofa	-27.79	-29.22	Lucquin et al 2016
Boar	Wild boar	Sus scrofa	-29.49	-29.57	Lucquin et al 2016
Boar	Wild boar	Sus scrofa	-28.94	-29.5	Lucquin et al 2016
Boar	Wild boar	Sus scrofa	-28.27	-27.42	Lucquin et al 2016
Boar	Wild boar	Sus scrofa	-28.24	-27.27	Miyata et al 2015
Boar	Wild boar	Sus scrofa	-28.27	-27.37	Lucquin et al 2016
Boar	Wild boar	Sus scrofa	-28.65	-27.68	Lucquin et al 2016
Boar	Wild boar	Sus scrofa	-28.58	-27.79	Lucquin et al 2016
Boar	Wild boar	Sus scrofa	-28.3	-27.2	Horiuchi et al. 2015
Boar	Wild boar	Sus scrofa	-28.4	-27.1	Horiuchi et al. 2015
Boar	Wild boar	Sus scrofa	-26.4	-26.9	Horiuchi et al. 2015
Boar	Wild boar	Sus scrofa	-28.1	-27.4	Horiuchi et al. 2015
Plant	Walnut		-30.59	-31.8	Miyata et al 2015
Plant	Acorn	Quercus sp.	-35.15	-32.77	Miyata et al 2015
Plant	Chestnut	Castanea sp.	-34.64	-33.54	Miyata et al 2015
Plant	Acorn	Quercus sp.	-37.67	-35.71	Miyata et al 2015
Plant	Chesnut	Castanea sp.	-29.43	-28.71	Heron et al 2016
Plant	Horse chesnut	Aesculus turbinata	-31.82	-29.56	Heron et al 2016
Plant	Acorn (Fagus crenata blume)	Fagus crenata	-27.6	-28.35	Heron et al 2016
Plant	Acorn	Quercus sp.	-33.7	-34.14	Lucquin et al 2016
Plant	Chesnut	Castanea sp.	-33.35	-30.61	Heron et al 2016
Plant	acorn	Quercus sp.	-32.38	-33.33	Lucquin et al 2016
Plant	acorn	Quercus sp.	-34.77	-35.64	Lucquin et al 2016
Plant	acorn	Quercus sp.	-32.81	-34.32	Lucquin et al 2016
Plant	acorn	Quercus sp.	-31.81	-33.71	Lucquin et al 2016
Plant	White oak	Quercus myrsinifolia	-34.7	-33.4	Horiuchi et al. 2015
Freshwater	Amur minnow	Rhynchocypris lagowskii	-27.43	-28.13	Craig et al. 2013
Freshwater	Topmouth gudgeon	Pseudorasbora parva	-26.95	-26.64	Craig et al. 2013
Freshwater	Spotbill duck	Anas poecilorhyncha	-27.35	-27.94	Craig et al. 2013
Freshwater	Crucian Carp	Carassius sp.	-29.15	-29.3	Lucquin et al 2016
Freshwater	Crucian Carp	Carassius sp.	-29.18	-29.64	Lucquin et al 2016
Freshwater	Crucian Carp	Carassius sp.	-28.27	-29.91	Lucquin et al 2016
Freshwater	Crucian Carp	Carassius sp.	-28.76	-29.08	Lucquin et al 2016
Freshwater	Crucian Carp	Carassius sp.	-28.64	-29.26	Lucquin et al 2016
Freshwater	Freshwater clam		-30.24	-30.03	Miyata et al 2015
Freshwater	Scarce		-28.23	-28.19	Lucquin et al 2016
Freshwater	Eriocheir	Eriocheir sp.	-27.33	-27.94	Lucquin et al 2016
Freshwater	Freshwater clam		-28.62	-28.06	Lucquin et al 2016
Freshwater	Freshwater clam	Cyrenidae	-30.3	-29.7	Horiuchi et al. 2015

Appendix IV:
Lipid compound and contaminant concentrations by occupational Phase

Phase	Sample Number	Total Contaminants (µg/g)	Total All Lipid concentration (µg/g)	Average by Phase (µg/g)
Phase 1	**12 DHA III H248 (1)_S5**	342.8	5.5	2292.7
	12 DHA III G18 (1)_S1	0.0	11.0	
	12 DHA III G18 (1)_S4	0.0	27.6	
	12 DHA III G18 (2)_S3	35.9	29.7	
	12 DHA III G18 (2)_S5	29.4	48.7	
	12 DHA III G18 (1)_S7	0.0	139.6	
	12 DHA III H146 (1)_S5	466.4	156.3	
	12 DHA III G18 (1)_S3	65.8	167.7	
	12 DHA III G18 (2)_S4	30.8	184.1	
	12 DHA III G22 (1)_S4	1265.4	186.9	
	12 DHA III G22 (1)_S1	16523.0	244.6	
	12 DHA III H146 (1)_S3	867.3	264.4	
	12 DHA III H146 (1)_S2	922.0	286.2	
	12 DHA III G21 (1)_S5	762.0	293.0	
	12 DHA III G18 (2)_S10	12.7	409.7	
	12 DHA III G18 (2)_S9	54.4	501.3	
	12 DHA III G21 (1)_S1	696.3	533.1	
	12 DHA III G18 (2)_S12	155.1	567.7	
	12 DHA III G18 (1)_S10	181.0	568.5	
	12 DHA III G18 (2)_S26	111.1	648.8	
	12 DHA III G18 (2)_S7	45.8	717.2	
	12 DHA III G18 (1)_S5	86.7	719.2	
	12 DHA III G18 (2)_S2	0.0	740.2	
	12 DHA III G18 (2)_S18	359.1	742.7	
	12 DHA III H248 (1)_S2	614.0	768.2	
	12 DHA III G18 (2)_S8	439.5	775.5	
	12 DHA III G21 (1)_S3	272.1	780.6	
	12 DHA III G18 (2)_S1	111.8	895.3	
	12 DHA III G18 (2)_S13	75.0	954.4	
	12 DHA III G22 (1)_S10	435.2	985.8	
	12 DHA III G21 (1)_S2	420.5	1079.4	
	12 DHA III G18 (2)_S30	354.4	1093.6	
	12 DHA III G18 (2)_S6	62.3	1189.3	
	12 DHA III G18 (2)_S19	781.3	1287.0	

	12 DHA III G18 (2)_S17	897.6	1317.1	
	12 DHA III G18 (2)_S14	114.7	1551.7	
	12 DHA III G22 (1)_S5	2047.4	1597.3	
	12 DHA III G18 (2)_S20	6854.0	1678.8	
	12 DHA III G18 (2)_S25	595.2	1987.4	
	12 DHA III H248 (1)_S3	1303.2	2105.9	
	12 DHA III H248 (1)_S4	562.1	2120.1	
	12 DHA III G22 (1)_S8	776.9	2472.9	
	12 DHA III G18 (1)_S9	182.5	2493.4	
	12 DHA III G18 (2)_S16	9823.0	2768.6	
	12 DHA III G18 (1)_S6	492.7	2804.7	
	12 DHA III G22 (1)_S9	1405.1	2833.2	
	12 DHA III G18 (1)_S8	237.5	2979.4	
	12 DHA III G18 (2)_S15	225.2	3620.7	
	12 DHA III G22 (1)_S3	463.7	3639.3	
	12 DHA III H248 (1)_S1	1550.6	3677.7	
	12 DHA III H146 (1)_S1	3748.3	3867.0	
	12 DHA III G21 (1)_S4	1806.6	5008.8	
	12 DHA III G18 (2)_S23	1011.6	5045.7	
	12 DHA III G22 (1)_S2	565.0	5340.2	
	12 DHA III G18 (2)_S21	112.0	5999.5	
	12 DHA III G18 (2)_S29	5829.5	6103.1	
	12 DHA III G22 (1)_S7	661.2	6219.0	
	12 DHA III G18 (2)_S24	876.4	8770.0	
	12 DHA III G18 (2)_S27	2058.5	10629.8	
	12 DHA III G18 (2)_S11	175.4	11644.3	
	12 DHA III H146 (1)_S4	27462.9	13574.3	
Phase 2	**11 DHA III G002 (1)_S3**	129.7	24.3	
	11 DHA III H131 (1)_S7	2348.7	51.3	
	11 DHA III H131 (1)_S9	2395.7	107.4	
	11 DHA III G002 (1)_S9	393.8	269.0	
	11 DHA III H131 (1)_S8	2013.5	309.4	
	11 DHA III H131 (1)_S3	1272.5	359.0	1352.2
	11 DHA III H131 (1)_S2	1761.5	397.9	
	11 DHA III H131 (1)_S5	1469.5	407.2	
	11 DHA III G002 (1)_S5	124.9	451.6	
	11 DHA III H131 (1)_S6	2017.1	490.1	
	11 DHA III G002 (1)_S8	383.7	515.5	

	11 DHA III G002 (1)_S4	202.6	629.0	
	11 DHA III G002 (1)_S7	324.9	758.4	
	11 DHA III G002 (1)_S1	216.7	1051.8	
	11 DHA III H131 (1)_S4	1340.9	1066.7	
	11 DHA III H131 (1)_S1	888.7	1136.1	
	11 DHA III G002 (1)_S10	321.3	1177.4	
	11 DHA III G002 (1)_S6	210.5	1285.2	
	11 DHA III G002 (1)_S2	589.2	1453.1	
	11 DHA III G13_S1	1207.5	4087.1	
	11 DHA III H131 (1)_S10	16879.6	12368.5	
Phase 3	**11 DHA III G001 (1)_S3**	461.9	158.8	
	11 DHA III G001 (1)_S11	250.7	166.2	
	11 DHA III G001 (1)_S16	79.4	177.5	
	11 DHA III G001 (1)_S8	324.8	194.4	
	11 DHA III G001 (1)_S17	186.9	218.8	
	11 DHA III G001 (1)_S9	361.0	232.0	
	11 DHA III G001 (1)_S6	261.7	315.3	733.5
	11 DHA III G001 (1)_S4	665.7	409.6	
	11 DHA III G001 (1)_S5	227.1	615.0	
	11 DHA III G001 (1)_S2	363.6	986.9	
	11 DHA III G001 (1)_S1	562.2	1040.3	
	11 DHA III G001 (1)_S10	814.8	1945.2	
	11 DHA III G001 (1)_S7	646.4	3076.0	
Phase 4	**14 DHA I F013 (1)_S2**	83.7	32.9	
	14 DHA I F016 (3)_S4	33.1	34.0	
	15 DHA I F023 (2)_S2	117.7	57.2	
	14 DHA I F016 (2)_S2	84.9	70.2	
	14 DHA I F016 (3)_S2	130.4	76.4	
	14 DHA I F013 (1)_S3	57.8	92.3	
	14 DHA I F013 (1)_S1	132.3	98.7	
	11 DHA III H085(1)_S8	300.7	104.0	375.5
	14 DHA I F016 (3)_S1	89.9	107.0	
	15 DHA I F023 (2)_S1	222.4	142.8	
	14 DHA I F017 D1 (2)_S2	124.5	148.7	
	14 DHA I F017 D1 (2)_S4	87.9	158.8	
	11 DHA III H085(1)_S7	188.2	168.1	
	14 DHA I F017 D1 (2)_S1	78.0	177.4	
	13 DHA I F008 (1)_S1	287.3	180.1	

	11 DHA III H085(1)_S2	56.4	186.3	
	14 DHA I F017 D1 (2)_S5	168.3	204.2	
	14 DHA I F010 (1)_S5	79.3	205.0	
	15 DHA I F021 (2)_S1	296.4	210.2	
	11 DHA III H085(1)_S1	138.1	260.1	
	14 DHA I F018 (2)_S1	164.5	273.6	
	13 DHA I F008 (1)_S2	369.6	276.6	
	14 DHA I F010 (1)_S4	84.3	278.7	
	14 DHA I F013 (1)_S4	415.2	325.2	
	14 DHA I F010 (1)_S2	177.1	558.8	
	11 DHA III H085(1)_S4	107.3	748.9	
	14 DHA I F010 (1)_S1	216.1	817.5	
	14 DHA I F010 (1)_S3	318.8	921.0	
	13 DHA I F008 (1)_S3	686.8	1135.5	
	11 DHA III H085(1)_S3	143.4	1740.0	
	13 DHA I F008 (1)_S4	815.6	1850.3	

Appendix V:
Lists of samples with inadequate preservation for analysis

Table AV-1. Samples from Houtaomuga which did not yield lipid concentrations above the threshold of analysis for compound specific IRMS (peak abundance >50000; palmitic and stearic acids)

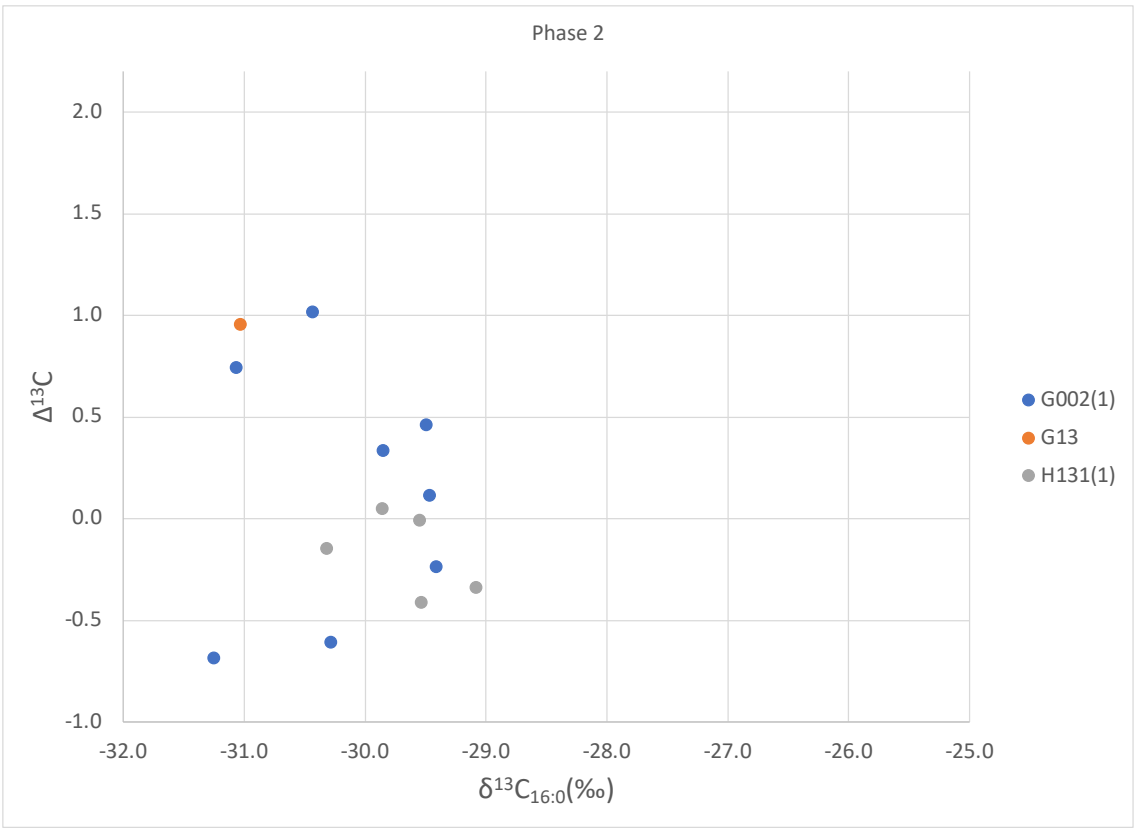
Field Number	Phase
12 DHA III G18 (1)_S1	Phase 1
12 DHA III G18 (1)_S7	Phase 1
12 DHA III G18 (2)_S1	Phase 1
12 DHA III G18 (2)_S2	Phase 1
12 DHA III G18 (2)_S5	Phase 1
12 DHA III G18 (2)_S8	Phase 1
12 DHA III G18 (2)_S11	Phase 1
12 DHA III G18 (2)_S12	Phase 1
12 DHA III G18 (2)_S13	Phase 1
12 DHA III G18 (2)_S14	Phase 1
12 DHA III G18 (2)_S15	Phase 1
12 DHA III G18 (2)_S16	Phase 1
12 DHA III G18 (2)_S17	Phase 1
12 DHA III G18 (2)_S18	Phase 1
12 DHA III G18 (2)_S19	Phase 1
12 DHA III G18 (2)_S20	Phase 1
12 DHA III G18 (2)_S21	Phase 1
12 DHA III G18 (2)_S22	Phase 1
12 DHA III G18 (2)_S23	Phase 1
12 DHA III G18 (2)_S24	Phase 1
12 DHA III G18 (2)_S25	Phase 1
12 DHA III G18 (2)_S26	Phase 1
12 DHA III G18 (2)_S27	Phase 1
12 DHA III G18 (2)_S28	Phase 1
12 DHA III G18 (2)_S29	Phase 1
12 DHA III G18 (2)_S30	Phase 1
12 DHA III G22 (1)_S1	Phase 1
12 DHA III G22 (1)_S4	Phase 1
12 DHA III G22 (1)_S6	Phase 1
11 DHA III G002 (1)_S9	Phase 2
11 DHA III H131 (1)_S7	Phase 2
11 DHA III H131 (1)_S9	Phase 2
11 DHA III G001 (1)_S3	Phase 3
11 DHA III G001 (1)_S11	Phase 3
11 DHA III G001 (1)_S12	Phase 3

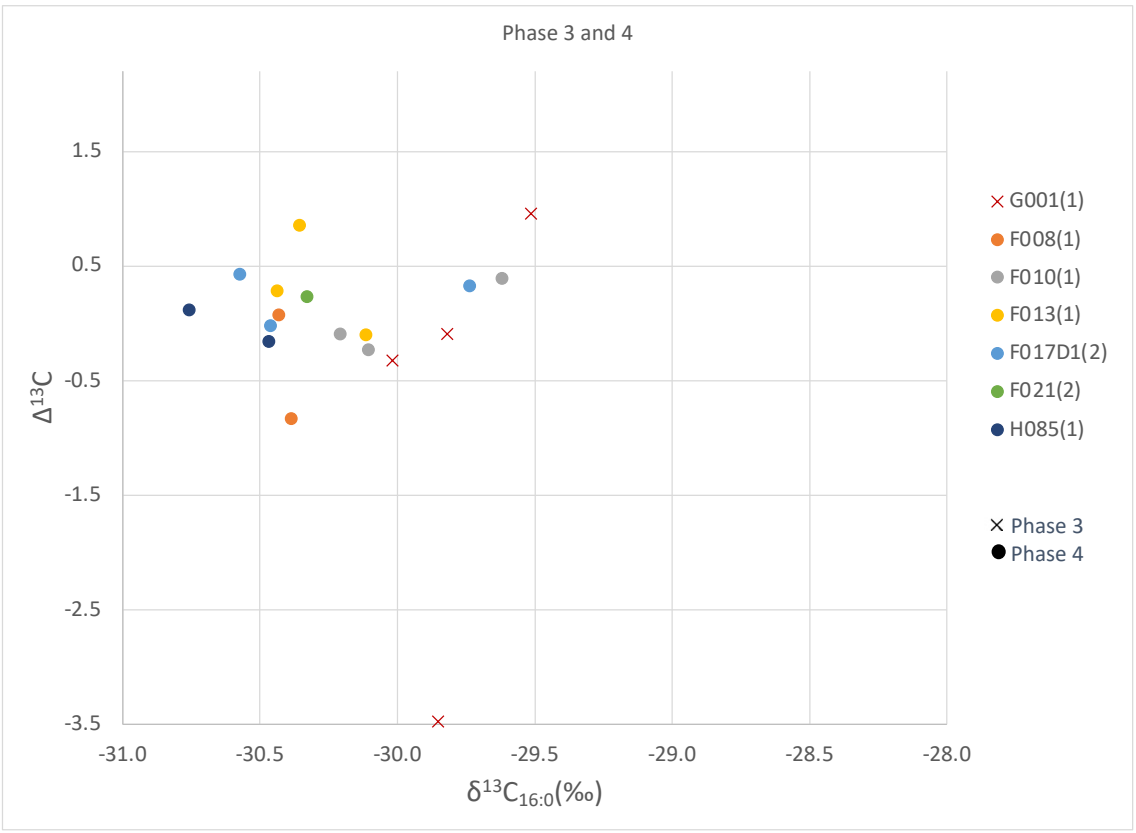
11 DHA III G001 (1)_S13	Phase 3
11 DHA III G001 (1)_S14	Phase 3
11 DHA III G001 (1)_S15	Phase 3
11 DHA III G001 (1)_S17	Phase 3
11 DHA III G001 (1)_S18	Phase 3
11 DHA III G001 (1)_S19	Phase 3
11 DHA III G001 (1)_S20	Phase 3
14 DHA I F016 (1)_S1	Phase 4
14 DHA I F016 (1)_S2	Phase 4
14 DHA I F016 (1)_S3	Phase 4
14 DHA I F016 (1)_S4	Phase 4
14 DHA I F016 (1)_S5	Phase 4
14 DHA I F016 (2)_S1	Phase 4
14 DHA I F016 (3)_S3	Phase 4
14 DHA I F016 (3)_S4	Phase 4
14 DHA I F017 D1 (2)_S3	Phase 4
15 DHA I F021 (2)_S2	Phase 4
15 DHA I F023 (2)_S2	Phase 4
15 DHA I F023 (2)_S3	Phase 4
15 DHA I F026 (2)_S1	Phase 4
15 DHA I F026 (1)_S1	Phase 4
11 DHA III H085(1)_S5	Phase 4
11 DHA III H085(1)_S6	Phase 4
11 DHA III H085(1)_S9	Phase 4
11 DHA III H085(1)_S10	Phase 4

Table AV-2. Samples which did not exhibit adequate carbon preservation for IRMS analysis

Field Number	Phase
12 DHA III G18 (1)_S10	Phase 1
12 DHA III G21 (1)_S2	Phase 1
12 DHA III G22 (1)_S7	Phase 1
12 DHA III H146 (1)_S1	Phase 1
11 DHA III G002 (1)_S7	Phase 2
11 DHA III H131 (1)_S4	Phase 2
11 DHA III H131 (1)_S5	Phase 2
11 DHA III H131 (1)_S10	Phase 2
11 DHA III G001 (1)_S4	Phase 3
11 DHA III G001 (1)_S5	Phase 3
11 DHA III G001 (1)_S6	Phase 3
11 DHA III G001 (1)_S8	Phase 3
11 DHA III G001 (1)_S9	Phase 3
11 DHA III G001 (1)_S16	Phase 3
13 DHA I F008 (1)_S1	Phase 4
13 DHA I F008 (1)_S2	Phase 4
14 DHA I F010 (1)_S4	Phase 4
14 DHA I F010 (1)_S5	Phase 4
14 DHA I F013 (1)_S2	Phase 4
14 DHA I F016 (2)_S2	Phase 4
14 DHA I F016 (3)_S1	Phase 4
14 DHA I F016 (3)_S2	Phase 4
14 DHA I F016 (3)_S4	Phase 4
14 DHA I F017 D1 (2)_S5	Phase 4
14 DHA I F018 (2)_S1	Phase 4
15 DHA I F023 (2)_S1	Phase 4
11 DHA III H085(1)_S1	Phase 4
11 DHA III H085(1)_S2	Phase 4
11 DHA III H085(1)_S7	Phase 4
11 DHA III H085(1)_S8	Phase 4

Appendix VI:
IRMS Data Split by feature and occupational phase


Phase 2
2.0
1.5
1.0
0.5
0.0
-0.5
-1.0
Δ13C
-32.0
-31.0
-30.0
-29.0
-28.0
-27.0
-26.0
-25.0
δ13C16:0(‰)
G002(1)
G13
H131(1)


Phase 3 and 4
1.5
0.5
-0.5
-1.5
-2.5
-3.5
Δ13C
-31.0
-30.5
-30.0
-29.5
-29.0
-28.5
-28.0
δ13C16:0(‰)
G001(1)
F008(1)
F010(1)
F013(1)
F017D1(2)
F021(2)
H085(1)
Phase 3
Phase 4

Appendix VII:
R Interface code

R version 3.6.0 (2019-04-26) -- "Planting of a Tree"
Copyright (C) 2019 The R Foundation for Statistical Computing
Platform: x86_64-apple-darwin15.6.0 (64-bit)

R is free software and comes with ABSOLUTELY NO WARRANTY.
You are welcome to redistribute it under certain conditions.
Type 'license()' or 'licence()' for distribution details.

  Natural language support but running in an English locale

R is a collaborative project with many contributors.
Type 'contributors()' for more information and
'citation()' on how to cite R or R packages in publications.

Type 'demo()' for some demos, 'help()' for on-line help, or
'help.start()' for an HTML browser interface to help.
Type 'q()' to quit R.

[R.app GUI 1.70 (7657) x86_64-apple-darwin15.6.0]

[Workspace restored from /Users/jenniferkeute/.RData]
[History restored from /Users/jenniferkeute/.Rapp.history]

2021-12-21 16:05:11.394 R[5563:15694388] Warning: Expected min height of view: (<NSPopoverTouchBarItemButton: 0x7faccbc6e3a0>) to be less than or equal to 30 but got a height of 32.000000. This error will be logged once per view in violation.
> hphase116 <- c(-29.18,-29.7845,-30.6355,-31.0805,-31.1755,-30.6345,-29.5485,-30.0255,-31.273,-30.945,-29.283,-29.3485,-30.383,-29.1785,-31.4265,-30.4495,-30.16,-29.937,-30.055,-30.302,-29.205,-28.2505,-25.357,-28.31,-28.107)
> hphase216 <- c(-30.4385,-29.4965,-29.4115,-29.8515,-31.2535,-31.0325,-29.5515,-29.539,-30.3205,-29.0825)
> hphase316 <- c(-29.515,-29.851,-30.017)
> hphase416 <- c(-30.386,-30.4315,-30.2085,-29.6205,-30.1045,-30.3565)
> gon16 <- c(-26.2,-28.6,-26.9,-24.4,-28.4,-26.7,-28.1,-25.5,-24.6,-25.0,-30.2,-29.4,-29.3,-27.6,-24.6,-29.7)
> gm16 <- c(-28.0,-27.6,-29.2,-28.3)
> sak16 <- c(-22.34,-20.11,-20.2,-18.95,-23.32,-23.45,-24.4,-23.13,-23.16,-23.8,-23.2)
> inc16 <- c(-25.6,-28.6,-29.1,-28.8,-28.8,-29.3,-28.7,-28.1,-27.6,-27.9,-28.2,-29.7,-29.6,-26.4,-25.8,-26.5,-25.7,-28.6,-26.2,-27.3,-31.6,-28.8,-31,-25.3,-24.9,-25.8,-26.1,-24.3,-26.7,-24.8,-28.3,-26.2,-26.6,-25.6,-26.8,-26.4,-25.8,-27.4,-28,-25.1,-27,-24.7,-26.7,-25.1,-24.8,-26.5,-27.9,-24.6,-23.8,-25.8,-25.2,-25.1,-29.1,-26.4,-27.8,-25.7,-26.6,-28.3,-28.9,-28.6,-26.8,-24.9,-25.4,-25.1,-24.9,-25,-25.6,-24.8,-25.3,-25.3,-25.9,-25.4,-27.1,-28.5,-29,-28.7,-28.8,-29,-28.9,-29.3,-26.4,-28.8,-26.2,-22.4,-25.6,-24.2,-24.3,-24.6,-23.8,-25.5,-25.2,-24.1,-24.6,-24.3,-24.2,-24.9,-24.8,-

24.4,-26.4,-24.3,-24,-23.8,-27.9,-24.8,-25.3,-24.6,-26.7,-25.9,-25.3,-27.3,-25,-26.7,-24.3,-26.3,-22.8,-23.5,-24.7,-25.4,-21.8)
> int16 <- c(-28.4,-25,-29.2,-28.1,-23.9,-24.5,-23.4,-27.9,-28.4,-21.9,-27.9,-26.6,-25.2,-28.4,-20.3,-19.7,-25.2,-25.4,-23.9,-23.3,-25.2,-22.2,-22.1,-24.3,-27.1,-25.6,-20.1,-26.1,-27.9,-24.7,-27.4,-28.4,-27.4,-25,-20.2,-25.2,-25,-28.8,-29.7,-25.6,-23.3,-22.4,-29.8,-30.2,-29.1,-24,-25.2,-24.7,-21.6,-26.8,-27.4,-22.2,-25.5,-23.1,-29.1,-26.4,-28.8,-28.8,-25.8,-20,-18.2,-26.8,-25.5,-29.5,-22.6,-28.3,-23.8,-28.9,-26.1,-25.4,-28.9,-26.4,-24.5,-24,-23.1,-22.6,-27.4,-26,-26.9,-28.4,-22.8,-26,-21.4,-22.6,-25.8,-25.2,-22.7,-24.9,-27.1,-27.4,-26.7,-28.9,-27.9,-28,-25.4,-26.6,-27,-28.6,-25.7,-26.8,-27.6,-25.4,-25.6,-29.6,-27.6,-25.7,-27.4,-26.7,-28.1,-26.4,-27.4,-27.1,-28.2,-28.5,-29.7,-29.1,-28.2,-28.5,-27.5,-28.3,-27.2,-27,-26.9,-25.6,-22.6,-23,-24.3,-23.3,-23.9,-25.8,-28,-27.7,-26.7,-27.8,-19.5,-25.3,-28.4,-26.5,-25.2,-27.8,-23.8,-26.3,-26.2,-27.1,-25.8,-26,-26.2,-25.9,-25.9,-26.1,-25.4,-28.9,-26.8,-28.2,-26.6,-27.5,-28.2,-27,-27,-29.3,-29.4,-29.5,-28.9,-28.3,-29.3,-28.9,-27.9,-27.7,-26.4,-27.1,-27.7,-28,-27.6,-27.8,-29.1,-30.1,-29.9,-28.8,-27.7,-28.4,-29.9,-30,-29.3,-29.5,-29.8,-30.1,-30.3,-29.7,-29.4,-29,-29.1,-29.6,-29.7,-28.8,-29.2,-29.3,-29.1,-28.4,-29.1,-28.3,-29.8,-28.8,-30,-30.4,-30.1,-29.9,-30,-27.8,-29.7,-27.4,-28.3,-26,-26.4,-26.1,-26.6,-26.1,-26.1,-27.2,-27.5,-29.5,-27.2,-28.8,-28.4,-30.1,-28.3,-26.4,-28.9,-28.2,-31.7,-28.1,-26.8,-29.7,-26.3,-31.7,-28.2,-28.9,-19.2,-27.3,-29,-30.2,-30.3,-30.3,-30.1,-30.2,-27.5,-30.3,-29.8,-28.1,-29.9,-28.1,-30.3,-28.6,-27.4,-28.7,-30.2,-29.4,-29.3,-30.3,-29.4,-27.1,-30.5,-27.6,-30.9,-30.3,-31,-30.5,-29.2,-30.3,-29,-29.2,-30.6,-29.6,-29.4,-27.7,-29.4,-27.1,-27.8,-30.5,-29.2,-29.9,-30.3,-27.1,-29.1,-30.8,-30.7,-29.9,-30.5,-30.1,-30,-27.9,-29.9,-29.6,-29.4,-27.8,-30.4,-29.5,-29.6,-29.5,-29.2,-29.5,-28.8,-29.1,-27.6,-29.4,-29,-28.7,-27.4,-27.7,-27.5,-25,-29.2,-27.9,-26.8,-24.2,-29.5,-25,-29.1,-26.3,-27.4,-27,-29,-25.7,-28.1,-26,-17.2,-29.2,-27.2,-29.4,-29.9,-20.8,-27.9,-24.1,-28.2,-23.3,-23.2,-28.7,-25.1,-24.8,-23.1,-26.1,-25.3,-26.6,-27,-28,-27.1,-26.9,-24.1,-28.5,-23.2,-28,-26.6,-29.9,-29.9,-27.4,-26.3,-29.3,-26.4,-24.6,-24.7,-27.3,-23.3,-25.7,-26.2,-29.1,-24.1,-23.3,-28,-24.3,-28.8,-24.6,-26,-22.2,-29.4,-30,-30,-30.4,-31.6,-29.9,-21.1,-28.3,-24,-23.5,-29.4,-26,-23.8,-27.9,-25.7,-29.9,-28.5,-30,-25.2,-26.5,-28,-25.7,-29.9,-29.8,-19.1,-27.5,-28,-24.1,-23.9,-27.6,-27.5,-28.5,-26.2,-26.9,-27.8,-29.9,-23.5,-27,-28.5,-28.6,-25.7,-27.1,-25.3,-25.7,-25.3,-26.9,-28.3,-30.7,-29.7,-28.5,-30.4,-26.6,-23.5,-26.1,-24,-27,-23.1,-26.8,-22.9,-23.4,-23.9,-22.9,-22.2,-24.6,-23.3,-23.3,-19.1,-18,-21.9,-21.7)
> boxplot(hphase116,hphase216,hphase316,hphase416,gon16,gm16,sak16,inc16,int16)
2021-12-21 16:09:56.755 R[5563:15694388] Warning: Expected min height of view: (<NSButton: 0x7faccbff0310>) to be less than or equal to 30 but got a height of 32.000000. This error will be logged once per view in violation.
2021-12-21 16:10:36.491 R[5563:15694388] +[NSSavePanel _warmUp] attempted warmup
> hphase118 <- c(-28.628,-27.686,-29.772,-29.9105,-29.7165,-29.4665,-28.7935,-29.365,-29.458,-29.9615,-29.2285,-28.5855,-29.485,-28.995,-30.2545,-30.054,-29.199,-29.1075,-29.755,-30.0865,-28.513,-27.423,-25.058,-27.3265,-27.884)
> hphase218 <- c(-29.422,-29.034,-29.646,-29.5165,-31.939,-30.078,-29.559,-29.948,-30.4655,-29.4185)
> hphase318 <- c(-28.5605,-33.3235,-30.3475)
> hphasee418 <- c(-31.22,-30.3605,-30.302,-29.228,-30.34,-29.5015)
> g18 <-- c(27.6,29.3,27.9,24.5,28.1,27.5,28.7,26.3,25.7,26.0,29.8,29.4,29.3,28.5,23.1,29.5)
> gm18 <--c(30.1,29.0,30.4,29.9)

```
> inc18 <- c(-24.9,-24.7,-28.4,-28.5,-27.5,-28.3,-29.2,-27.5,-27.1,-28.9,-28.9,-30.6,-30.3,-25.5,-
24.6,-26.3,-25.3,-28,-25,-26.8,-27.7,-28.8,-26.6,-25.3,-25.2,-25.8,-25.9,-24.1,-27.5,-24.5,-31.4,-
27.9,-25.3,-26.9,-28.9,-28.7,-28.4,-31.6,-30.7,-25.5,-29.8,-28.9,-29.4,-26.7,-27.7,-28.1,-31.1,-
26.2,-25.4,-29.5,-27.3,-26.5,-29.3,-27.3,-28.3,-25.7,-27.5,-29.3,-29.4,-29.1,-27.9,-24.5,-28.6,-
24.6,-27.4,-25.8,-28.2,-23.8,-24.4,-27.3,-27.8,-26.8,-28.5,-28.9,-29,-28.5,-31.3,-29.2,-31.3,-29.4,-
29.4,-31,-25.5,-22.2,-25.5,-24.1,-24.2,-23.6,-23.5,-24.4,-24.1,-24.1,-25.8,-24.2,-23.8,-25.1,-24.4,-
24.3,-27,-23.9,-24.8,-24.1,-28.2,-26.5,-25.1,-25.2,-27.4,-26.5,-23.9,-28,-25.5,-28.2,-26,-28.7,-
26.1,-25.1,-26.3,-23.8,-23.1)
> int18 <- c(-28.1,-25.1,-29.6,-27.9,-26.6,-24.8,-23.9,-28.4,-28.5,-23.3,-28.2,-27.2,-26,-27.9,-
22.6,-23.4,-26.4,-27,-24.5,-24.5,-26.5,-23.3,-21.8,-26,-27.8,-25.7,-22.3,-26.3,-28.5,-24.4,-27.4,-
29.1,-25.4,-25.6,-22,-27,-26.8,-29.1,-29.2,-24.9,-23.6,-23.1,-29.2,-31.1,-29,-25.6,-28.1,-25.8,-24,-
26.8,-27,-23.7,-26.7,-26.6,-28.7,-25.9,-28.7,-27.5,-25.4,-20.8,-20.8,-26,-25.4,-28.4,-23.8,-27.1,-
22.5,-28.9,-26.6,-26.3,-28.4,-26.7,-24.6,-24.2,-21.3,-23.2,-28,-24.8,-27.6,-27.3,-25.3,-25.4,-23.7,-
20.8,-26.7,-24.9,-23.2,-26.3,-30,-28.6,-28.2,-31.2,-28.5,-28.8,-26.1,-28.7,-29.9,-30.3,-26.7,-29.8,-
29.1,-29.4,-27.4,-31.6,-29.5,-25.9,-26.9,-29.5,-27.3,-25.7,-26,-26.6,-30.8,-28.2,-29.4,-28.6,-30.1,-
28.8,-26.9,-31.5,-28.3,-29.1,-28.6,-25.5,-23.8,-23.4,-24.3,-24,-23.9,-25.7,-26.9,-27,-26.6,-30,-
21.8,-24.5,-28.9,-27,-25.4,-27.9,-24.7,-28.1,-26,-27.7,-27.7,-27.9,-26.6,-26,-28.2,-26.6,-25.5,-
29.2,-28.1,-28.3,-28.3,-28.3,-27.8,-28.3,-27.5,-28.6,-29.3,-29.5,-29.3,-29.3,-29.2,-29.2,-28.6,-
28.6,-25.7,-28.1,-28.6,-28.9,-28.3,-28.1,-28.6,-29.9,-29.7,-29.2,-28.1,-28.7,-29.6,-29.4,-29.6,-
29.4,-29.7,-29.9,-29.9,-29.5,-29.3,-29.3,-29.4,-29.7,-29.7,-29.3,-29.6,-29.1,-29.4,-28.7,-29.2,-28,-
29.7,-29.2,-29.6,-29.9,-29.7,-29.8,-31.9,-28.9,-29.8,-28,-28.5,-27,-27.4,-27.6,-27.8,-27.2,-26.8,-
27.6,-27.7,-29.8,-29.5,-29,-28,-27.7,-26.1,-26.3,-27,-30.4,-32.8,-29.4,-26.4,-29.8,-28,-29.4,-28.6,-
28.6,-24.7,-30.1,-28.9,-30.2,-30,-30.1,-30.1,-30.1,-29.1,-29.7,-29.9,-27.9,-29.7,-27.2,-29.7,-28.1,-
29.3,-29.1,-30.5,-29.4,-30,-30.1,-29.6,-27.4,-30,-28.2,-29.9,-29.7,-30.2,-30.3,-28.7,-30,-29.2,-
30.7,-30,-30,-30.2,-28.8,-29,-26,-27.5,-29.9,-29.5,-29.4,-29.8,-28.4,-30.1,-30.7,-30.1,-30.8,-29.7,-
29.1,-30.5,-28.3,-29.9,-30.7,-30.1,-28.8,-28.6,-30,-29.7,-29.6,-29.6,-29,-29.7,-29.6,-29.3,-29.8,-
29.7,-29.5,-30.1,-28.4,-27.8,-25.3,-29.1,-30,-28.5,-25.5,-29.1,-26.4,-28.9,-26.5,-27.8,-28.3,-29.1,-
25.5,-28.4,-25.7,-23.6,-29.4,-28.2,-29.1,-29.6,-24,-29.2,-24.3,-29.8,-24.8,-23.4,-28.8,-26.8,-26.6,-
26,-26.4,-26.6,-29.8,-24.1,-28.6,-29.4,-28.2,-25.2,-28.6,-25.1,-28.3,-25.1,-29.7,-29.8,-25.7,-28.9,-
28.9,-27.6,-25.8,-26.1,-27.7,-24.8,-25.9,-26,-29.1,-25.3,-24.4,-29.4,-26.3,-30.4,-25,-27,-24.1,-
29.7,-29.4,-29.8,-29.6,-29.1,-32.4,-20.3,-23.7,-28.3,-23.8,-29.2,-25.4,-25.3,-27.3,-25.3,-29.2,-
29.1,-29.2,-26,-29.5,-28.6,-26.1,-28,-29.3,-21.6,-29.3,-29.4,-25.5,-25.4,-27.9,-29.1,-28.1,-25.8,-
26.6,-27.9,-29.8,-24.2,-26.4,-29.1,-28.7,-24.3,-29.1,-22,-27.2,-26,-27.5,-26.8,-30.5,-33.1,-29.4,-
29.9,-24.9,-24.2,-27.5,-24.6,-28.2,-23.4,-28.6,-22,-23.2,-23.7,-22.9,-21.9,-25.1,-23.8,-22.4,-21.2,-
17.4,-21.2,-20.7)
> boxplot(hphase118,hphase218,hphase318,hphase418,g18,gm18,inc18,int18)
Error in boxplot.default(hphase118, hphase218, hphase318, hphase418, g18,  :
  object 'hphase418' not found
> boxplot(hphase118,hphase218,hphase318,hphasee418,g18,gm18,inc18,int18)
>
```

Appendix VIII:
GC-MS Method Testing Calculations

Peak Asymmetry

Peak asymmetry was calculated to discuss applicability of the tested methods. Peak Asymmetry was calculated by measuring peak width at 5% height (A+B) as split by the midway point into values A for the front width of the peak and B for the back width. In Gaussian peaks which are symmetrical, these values are the same. Peak asymmetry is calculated by dividing value B by value A and values less than 1 show tailing while values over 1 show peak fronting. In Gaussian peaks, this value equals 1.

The selection of tables presented below show peak asymmetry for target compounds in heritage science from sample LFS-7 and CFS2-4 from South Africa and reference materials. Conclusions about adequate methods for GC-MS testing in section 3.1.3 were determined based off these calculations.

Beeswax: Lauryl Palmitate Peak Comparison		RT	Front RT	Back RT	A	B	A+B	Asymmetry (B/A)
Rxi-65TG	Method A	33.58	33.48	33.64	0.10	0.06	0.16	0.60
Rxi-65TG	Method B	35.14	35.08	35.29	0.07	0.14	0.21	2.15
Rxi-65TG	Method C	29.19	29.13	29.26	0.06	0.07	0.13	1.16
Rxi-65TG	Method D	33.51	33.44	33.69	0.07	0.18	0.25	2.73
DB-5ms	Method A	41.43	41.32	41.72	0.12	0.29	0.40	2.48
DB-5ms	Method B	41.48	41.38	41.63	0.10	0.15	0.25	1.58
DB-5ms	Method C	36.63	36.51	36.93	0.13	0.30	0.42	2.38
DB-5ms	Method D	39.81	39.55	40.39	0.26	0.59	0.85	2.25

CFS2-4: Palmitic Acid TMS Peak Comparison		RT	Front RT	Back RT	A	B	A+B	Asymmetry (B/A)
Rxi-65TG	Method A	19.39	19.28	19.46	0.12	0.07	0.18	0.58
Rxi-65TG	Method B	19.91	19.85	20.02	0.07	0.11	0.17	1.60
Rxi-65TG	Method C	19.91	19.85	20.02	0.07	0.11	0.17	1.68
Rxi-65TG	Method D	19.92	19.85	20.02	0.08	0.10	0.17	1.25
DB-5ms	Method A	23.42	23.36	23.49	0.07	0.07	0.13	1.00
DB-5ms	Method B	23.76	23.67	23.88	0.09	0.13	0.21	1.45
DB-5ms	Method C	23.78	23.70	23.90	0.08	0.13	0.20	1.62
DB-5ms	Method D	24.25	24.15	24.37	0.10	0.13	0.22	1.30

CFS2-4: Stearic Acid TMS Peak Comparison		RT	Front RT	Back RT	A	B	A+B	Asymmetry (B/A)
Rxi-65TG	Method A	21.10	21.01	21.23	0.10	0.13	0.22	1.30
Rxi-65TG	Method B	21.65	21.60	21.78	0.06	0.13	0.18	2.16
Rxi-65TG	Method C	21.10	21.01	21.22	0.09	0.12	0.21	1.30
Rxi-65TG	Method D	21.68	21.61	21.72	0.07	0.04	0.11	0.56
DB-5ms	Method A	24.70	24.65	24.80	0.05	0.10	0.14	2.00
DB-5ms	Method B	25.59	25.51	25.65	0.08	0.07	0.14	0.87
DB-5ms	Method C	25.61	25.54	25.74	0.08	0.13	0.20	1.62
DB-5ms	Method D	26.05	25.98	26.12	0.07	0.08	0.14	1.15

Peak Resolution

Peak resolution was calculated to understand success of the tested methods. Peak resolution was calculated using the following equation to understand success of the tested methods. *Tr* refers to the retention items of both peaks and *W* is the width at 5% height.

$$R=(Tr2-Tr1)/0.5*(W1+W2)$$

For adequate peak resolution, R should equal more than 1.5. In the following selection of tables, peak resolution is calculated between target compounds from sample LFS-7 and CFS2-4 from South Africa and Beeswax reference standard on each method and stationary phase. Conclusions about adequate methods for GC-MS testing in section 3.1.3 were determined based off these calculations.

LFS-7		Peak 1 RT (Heneiosane)	Peak 2 RT (C28 Alcohol)	Peak 1 Width	Peak 2 Width	Resolution
Rxi-65TG	Method A	27.0	27.2	0.2	0.3	1.0
Rxi-65TG	Method B	27.6	27.7	0.1	0.1	1.5
Rxi-65TG	Method C	27.6	27.8	0.2	0.1	1.7
Rxi-65TG	Method D	27.6	27.7	0.1	0.1	1.5
DB-5ms	Method A	28.5	28.7	0.1	0.1	1.5
DB-5ms	Method B	32.2	32.5	0.2	0.2	1.6
DB-5ms	Method C	32.2	32.4	0.1	0.1	2.3
DB-5ms	Method D	31.0	31.3	0.2	0.2	1.4

CFS2-4		Peak 1 RT (C16 TMS)	Peak 2 RT (Palmitelaidic acid)	Peak 1 Width	Peak 2 Width	Resolution
Rxi-65TG	Method A	19.4	19.5	0.2	0.1	0.6
Rxi-65TG	Method B	19.9	20.1	0.2	0.1	0.9
Rxi-65TG	Method C	19.9	20.0	0.2	0.1	0.6
Rxi-65TG	Method D	19.9	20.1	0.2	0.1	0.9
DB-5ms	Method A	23.4	23.3	0.1	0.1	1.0
DB-5ms	Method B	23.8	23.6	0.2	0.1	1.0
DB-5ms	Method C	23.8	23.7	0.2	0.1	0.8
DB-5ms	Method D	24.2	24.1	0.2	0.1	1.0

Beeswax		Tetracosanoic acid	Heneicosane	Peak 1 Width	Peak 2 Width	Resolution
Rxi-65TG	Method A	25.6	25.7	0.1	0.1	0.5
Rxi-65TG	Method B	26.2	26.2	0.2	0.0	0.6
Rxi-65TG	Method C	25.4	25.5	0.0	0.1	0.6
Rxi-65TG	Method D	26.6	26.6	0.0	0.1	0.4
DB-5ms	Method A	30.3	30.8	0.2	0.1	3.1
DB-5ms	Method B	30.3	30.8	0.2	0.1	3.3
DB-5ms	Method C	27.4	27.7	0.1	0.1	2.9
DB-5ms	Method D	29.5	29.8	0.1	0.1	2.7

Appendix IX:
Details of quantitation and blanks procedure for GC-MS

A-IX-1. Blanks Procedure

For SFE, one blank ceramic pellet for every 12 archaeological samples was subject to the same SFE procedure as archaeological samples. The extract of this extraction was then subject to the same procedures for analysis as outlined in section 2.2.2.3.

For GC-MS, archaeological and method validation samples were processed in groups of 6 with one derivatization blank. The derivatization blank was a clean glass vial with the same internal standards as samples added, including 10µl tridecanoic acid as IS 1 and 20µl hexadecane as IS2. These samples were subject to an identical GC-MS derivatization procedure as outlined in section 2.2.2. Samples were analyzed on the GC-MS in groups of 2, for a total of 12 samples and 2 blanks in each run group.

A-IX-2. Methods for lipid quantitation

All samples, including blanks as detailed above, received internal standards as quantitation markers. IS 1 – tridecanoic acid – was added at a concentration of 135µg/g. IS2 – hexadecane – was added at a concentration of 100µg/g. The ratio between these peaks' relative abundances and the relative peak abundance of the target compounds was calculated and the known concentrations of internal standards was used to quantify lipid abundances.

Appendix X:
Tables for figures 3.16-3.22

The following appendix includes tables of peak assignments which accompany figures 3.15-3.21. These figures show chromatograms obtained through GC-MS. RT stands for retention time for each compound illustrated and marked. IS 1 stands for internal standard 1 which is tridecanoic acid TMS ester. IS 2 stands for internal standard 2 which is hexadecane.

Table AX-1. Compounds illustrated in figure 3.16

RT	Compound
11.701	Hexanoic acid TMS
13.047	Heptanoic acid TMS
14.537	Octanoic acid TMS
15.893	Nonanoic acid TMS
17.182	Decanoic acid TMS
19.009	IS 2
19.576	Dodecanoic acid TMS
20.691	IS 1
21.759	Tetradecanoic acid TMS
22.778	n-Pentadecanoic acid TMS
23.778	Hexadecanoic acid TMS
24.691	Heptadecanoic acid TMS
25.624	Octadecanoic acid TMS
27.287	Eicosanoic acid TMS
28.422	16-Hydroxyoctadecanoic acid (TMS derivative)
28.701	cis-13-Docosenoic acid TMS
28.874	Docosanoic acid TMS
29.672	Tetracosane
30.345	Tetracosanoic acid TMS
31.085	C26 Alcohol
32.191	Octacosane
32.422	C28 Alcohol
33.691	C30 alcohol

Table AX-2. Compounds illustrated in figure 3.17

RT	Compound
10.883	Octanoic acid TMS
12.076	Nonanoic acid TMS
13.278	Decanoic acid TMS
14.258	IS 2
14.393	Undecanoic acid TMS
15.47	Dodecanoic acid TMS
16.489	IS 1
17.479	Tetradecanoic acid TMS
18.066	Pentadecanoic acid TMS
18.431	n-Pentadecanoic acid TMS
19.374	Hexadecanoic acid TMS
19.864	C16 Alcohol
20.22	Heptadecanoic acid TMS
21.114	Octadecanoic acid TMS
21.893	Nonadecanoic acid TMS
22.681	Eicosanoic acid TMS
23.451	Heneicosanoic acid TMS
23.758	16-Hydroxyoctadecanoic acid (TMS derivative)
24.191	cis-13-Docosenoic acid TMS
24.537	Tetracosane
25.124	18-Hydroxyoctadecanoic acid (TMS derivative)
25.566	Tetracosanoic acid TMS
26.951	Octacosane
27.172	C28 Alcohol
28.393	C30 alcohol
29.528	C32 Alcohol
30.595	C34 Alcohol

Table AX-3. Compounds illustrated in figure 3.18

RT	Compound
11.749	Hexanoic acid TMS
13.066	Heptanoic acid TMS
14.547	Octanoic acid TMS
15.903	Nonanoic acid TMS
17.181	Decanoic acid TMS
18.412	Undecanoic acid TMS
19.018	IS 2
19.57	Dodecanoic acid TMS
20.691	IS 1
21.73	Tetradecanoic acid TMS
22.614	n-Pentadecanoic acid TMS
23.412	Hexadecanoic acid TMS
24.076	Heptadecanoic acid TMS
24.71	Octadecanoic acid TMS
25.739	Eicosanoic acid TMS
26.383	16-Hydroxyoctadecanoic acid (TMS derivative)
26.556	cis-13-Docosenoic acid TMS
27.46	Docosanoic acid TMS
28.662	Tetracosane
28.528	Tetracosanoic acid TMS
28.662	C26 Alcohol
28.998	Octacosane
29.566	C28 Alcohol
30.633	C30 alcohol
31.96	Siloxane

Table AX-4. Compounds illustrated in figure 3.19

RT	Compound
11.403	Hexanoic acid TMS
13.018	Heptanoic acid TMS
13.566	Octanoic acid TMS
15.172	Butanedioic acid TMS
18.97	IS 2
20.653	IS 1
24.239	Phanthrene
26.105	Retene
27.153	Dehydroabietic acid TMS
28.374	7-oxo-dehrydroabietic acid TMS
29.451	Column bleed
31.383	Column bleed
36.31	Column bleed
39.018	Column bleed

Table AX-5. Compounds illustrated in figure 3.20

RT	Compound
11.403	Hexanoic acid TMS
13.037	Heptanoic acid TMS
14.355	Octanoic acid TMS
15.201	Butanedioic acid TMS
18.999	IS 2
20.681	IS 1
23.441	Phanthrene
23.73	Phanthrene
24.806	Retene
25.605	Dehydroabietic acid TMS
26.624	7-oxo-dehrydroabietic acid TMS
27.47	Column bleed
28.835	Column bleed

Table AX-6. Compounds illustrated in figure 3.21

RT	Compound
24.806	Phanthrene
24.931	Phanthrene
25.143	Retene
25.229	Dehydroabietic acid TMS
25.643	7-oxo-dehrydroabietic acid TMS
27.181	Column bleed
27.672	Column bleed

Table AX-7. Compounds illustrated in figure 3.22

RT	Compound
24.287	Phanthrene
24.412	Phanthrene
24.605	Retene
24.682	Dehydroabietic acid TMS
25.134	7-oxo-dehrydroabietic acid TMS
26.98	Column bleed
27.672	Column bleed

Appendix IX:
Samples analysed in this thesis