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Reconstruction of ancient environments using stable isotope analysis of archaeological charcoal from Sibudu Cave, KwaZulu-Natal

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Reconstruction of ancient environments using stable isotope analysis of archaeological charcoal from Sibudu Cave, KwaZulu-Natal

Volume I

Grant Hall



A thesis submitted to the Faculty of Science, University of the Witwatersrand, Johannesburg, in fulfilment of the requirements for the degree of Doctor of Philosophy

Johannesburg, 2010

DECLARATION

I declare that this thesis is my own unaided work. By thesis, it is understood to mean my contribution, as described in Chapter One, for submission for the degree of Doctor of Philosophy at the University of the Witwatersrand. The thesis has not been submitted before for any degree or examination at any other University.

_____

Grant Hall

_____ day of _____ 2010.

ABSTRACT

The stable carbon isotopic analysis of archaeological charcoal has the potential to provide an archive of environmental change during the Middle Stone Age occupation of Sibudu Cave, KwaZulu-Natal, South Africa. A wide array of evidence from the site suggests that profound environmental and cultural changes took place in the region through the past 75ka. The aim of this project is to develop a methodology to obtain such isotopic evidence and to test the validity of this evidence through comparison with additional proxy environmental data from the site.

Analogue data from modern tree species, *Mimusops caffra*, *Podocarpus latifolius* and *P. falcatus* provide an absolute annual chronological isotopic record of environmental response to prevailing climatic conditions. High precision radiocarbon dating of growth rings demonstrates that these species produce annual growth rings. Wholewood samples provide an accurate record of climatic conditions. Direct comparison of $\delta^{13}C$ value of *M. caffra* from northern KwaZulu-Natal with the historic climatic record, shows a strong inter-annual response to rainfall variability. Signal processing approaches allow the rainfall response to be separated from long-term anthropogenic influences.

The $\delta^{13}C$ values of *P. latifolius* trunk cores and corresponding branch samples, from KwaZulu-Natal, preserve the same environmental record, correlating with humidity and temperature data from the region. It is thus possible to obtain a record of past climatic conditions from the growth rings of branches. A *P. falcatus* disc from the Baviaans Kloof (Eastern Cape) provides a $\delta^{13}C$ time series for a moisture-restricted environment, responding to annual rainfall variation and provides a contrasting analogue to the KwaZulu-Natal trees.

Branch samples from *P. latifolius*, representative of the size class of wood fuel likely utilised by MSA inhabitants of Sibudu Cave, were combusted under oxidising and reducing conditions. Their respective $\delta^{13}C$ values are more negative with respect to the source material, but reflect the same response to prevailing climatic conditions. An experiment determined the range of isotopic variability in products released during various stages of combustion. The results indicate that combustion

temperature has a significant affect on the carbon isotope signature of the various products released during combustion, but the isotopic composition of the remaining wood tissue remains relatively constant, preserving seasonal and inter-annual isotopic trends. These modern analyses indicate that it is possible to obtain an isotopic record of past climatic conditions from archaeological charcoal.

The isotopic variability of archaeological *Podocarpus* and *Celtis* charcoal, from the MSA layers of Sibudu Cave reflect past environments. During the Howiesons Poort (65ka-62ka) the $\delta^{13}C$ charcoal data indicate a cool, humid forested environment predominated. $\delta^{13}C$ data from the post-Howiesons Poort (~58ka) and late MSA (~48ka) occupations show that conditions shifted to a dry, open grassland/woodland mosaic community with remnants of riverine forest. These interpretations were validated through comparison with additional faunal, botanical and sedimentological proxy data from Sibudu.

The environmental record from Sibudu Cave and palaeoenvironmental proxy data from seven South African MSA sites provide evidence for the local manifestation of large scale climate events between 70ka and 50ka. During Marine Isotope Stage (MIS) 4 it appears that southern Africa experienced conditions similar to those during the Last Glacial Maximum. At Sibudu the environment changed from a predominantly forested community to more open grass/wood land mosaic. Such environmental change is thought to be due to a weakening of the Agulhas Current and eastward shift of the Agulhas Retroflection resulting in lower sea surface temperatures and a corresponding decrease in humidity and rainfall. Adverse environmental conditions ultimately led to Sibudu being abandoned from about 37ka until approximately 1000 BP.

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Weather Service was instrumental in providing me with all the necessary climatic data that I requested.

I would like to thank Zenobia Jacobs for permission to use the currently available Optically Stimulated Luminescence dates for Sibudu Cave. Marc Pienaar assisted me with a number of complex statistical analyses and associated figures for one of the papers presented in the thesis. A final thank you goes to all the members of the ACACIA team (Lucy Allott, Andrew Herries, Marlize Lombard, Wayne Glenny, Ina Plug, Jamie Clark, Chet Cain, Paolo Villa, Sally Reynolds and others) who have worked at Sibudu Cave producing a wealth of data which has been of invaluable use to my research.

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Table C.1. Annualised weather data (rainfall, temperature and relative humidity) from 2005 to 1928. The dataset is composed of weather data from three stations, Durban, Mount Edgecombe and Virginia Airport. Relative humidity data were only available until 1983 (South African Weather Service).

Table C.2. Annualised climatic data (rainfall, temperature and relative humidity) from 2004 to1927. The dataset is composed of weather data from weather stations in Joubertina and Kareedouw in the Eastern Cape. Relative humidity and temperature data were only available until early 1997 (South African Weather Service).

Table C.3. Summary table for all sources of palaeoenvironmental proxies cited. The type of site (e.g. archaeological, deep sea core), location, age model or dating methods, proxy data types, a brief indication of environmental changes for the period 70-50ka and references are provided for each site.

CHAPTER 1. INTRODUCTION

1.1. Introduction

The reconstruction of past environments from archaeological sites is best formulated through the analysis of a wide range of proxy evidence. Stable carbon isotope analysis of plant material has been well established as a means of obtaining such proxy environmental evidence. Although the human inhabitants of a site select and utilise a range of plants and animals, the isotopic signatures of such resources are not affected by this selection, but are influenced by the environment in which they naturally occurred or grew. An analysis of the stable isotope composition, such as carbon, of such material (e.g. bone, enamel, wood or charcoal) has the potential to provide independent proxy data on the environments from whence they came.

Archaeological charcoal assemblages are assumed to have developed largely due to anthropogenic selection of suitable woody species for fuel, although certain of these species may also be used for construction purposes such as posts and lintels. Such material may recovered directly from easily distinguished hearth features or from associated deposits of ash and charcoal that were formed during the cleaning of hearths. These selected species are, however, at least partly a representation of the vegetation communities in the site vicinity. Changes in the species composition of charcoal assemblages through time will thus be a reflection of either change in the overall vegetation communities in the area in response to climatic changes or a shift in preference for fuel wood types. The carbon isotopic composition of archaeologically recovered charcoals have the potential to provide an archive of past climates, reflecting subtle isotope variations as the plants respond to a changing environment.

The current trend in archaeological research is to apply a multi-disciplinary approach as the strongest type of archaeological evidence is multi-faceted. Thus a variety of palaeoenvironmental data providing complementary evidence is compelling. Research at Sibudu Cave has produced a suite of environmental data from sediments, botanical and faunal remains. This array of palaeoenvironmental evidence has provided detailed insights into how the local environment around the site has changed at various periods during the past 75ka. Carbon isotope analysis of archaeologically recovered,

identified charcoal will provide another environmental proxy data set for the existing compilation.

1.2. Project aims

The principal aim of this project is:

To develop a methodology to obtain palaeoenvironmental evidence from the stable carbon isotope analysis of archaeological charcoal from the Middle Stone Age (MSA) occupation of Sibudu Cave and to test the validity of this evidence with additional proxy environmental data from the site.

In order to attain this goal a number of requirements need to be satisfied.
These are:

- **A range of analogue data from modern tree species from KwaZulu-Natal must provide an absolute annual chronological isotopic record of environmental response to prevailing climatic conditions.**
- **An environmental proxy (e.g., a response to rainfall or humidity) recorded in a tree ring sequence must be similarly reflected in the equivalent rings of branches from the same tree.**
- **This response must be conserved in the branch during charcoal formation, the processes involved and how these may affect the isotope proxy must be understood and accounted for where possible.**

1.3. Thesis structure

This PhD thesis is composed of a series of papers written during the period of my registration at the University of the Witwatersrand. This is in accordance with guidelines governing the submission of a thesis by publication. The four papers included in the thesis are in various stages of publication, two have been submitted to journals and are currently under review and two have been published. The rationale for the submission of a thesis by publication is the relatively rapid dissemination of data/results in a rapidly expanding and highly topical field, namely global climate change. The four papers are presented in such a manner as to provide a logical

progression of the research conducted. In order to maintain a standardised format throughout the thesis and for ease of reading, each paper is **not** presented in the various house styles of each journal, but rather as individual chapters (Chapters 4-7), linked together. A list of all relevant references for each paper is presented at the end of each chapter. The associated figures and tables for each chapter are grouped together in Volume II of the thesis to allow easier access to the relevant figures and tables for each chapter. At the beginning of each paper chapter a short introduction is provided outlining the rationale for methods used and the relevance of the data and results in the greater project goal.

Chapter 2 provides an introduction and background into charcoal analysis and stable carbon isotope research and a review of recent applications of carbon isotope analysis as a source of proxy palaeoenvironmental records from tree rings and other plant material (charcoal, fossils and leaves). Further discussion of the methods used in charcoal identification and analysis are provided as the focus of this project is on the potential of stable carbon isotope analysis of archaeological charcoal as an environmental proxy. A literature review of a number of relevant international stable carbon isotope studies, as well as a review of southern African studies is included.

Chapter 3 provides a background to Sibudu Cave and a summary of the most recent research carried out at the site. Sibudu is proving to be one of the more significant MSA sites in southern Africa in that it has a deep, well-preserved and well-dated sequence of material that has been subject to a multi-disciplinary analysis providing detailed information on environmental and cultural change during the MSA. The final occupants of the site were from local Iron Age populations and there are no indications of a Later Stone Age occupation. The emphasis is not on the cultural aspects of the MSA occupations, but rather on the available environmental proxy evidence derived from a multi-disciplinary research programme. The modern local environment, geology and dating of the site and the various methods used to define the environmental proxies are discussed. The environmental changes through time are presented.

Chapter 4 comprises a paper published in *The Holocene,* explaining the necessity for modern analogue environmental data from tree ring sequences to understand how

indigenous coast forest trees from KwaZulu-Natal respond to prevailing climatic and environmental conditions. The paper covers applicable sampling methods, sample pre-treatment, the development of annually resolved time series, carbon isotope analysis and the interpretation of the proxy environmental data obtained from *Mimusops caffra*. The results allowed me to establish part of the final methodology that was used for the project. I was responsible for all sample collection, preparation and isotopic analysis, as well as preparation and running of three samples for high precision radiocarbon dating. I generated the majority of the figures and the initial draft of the paper. Dr. Stephan Woodborne assisted with the generation of the radiocarbon dates, data interpretation and editing. Marc Pienaar carried out the spectral analysis using wavelets and produced figures 4.4 and 4.5 (Volume II).

In Chapter 5, a paper published in *Chemical Geology* presents additional modern proxy environmental isotope records from two *Podocarpus* species (*Podocarpus latifolius* and *P. falcatus*) and the results of a series of experiments investigating the alteration of carbon isotope signals during the combustion process. A coherent environmental signal is preserved in both branch growth rings and charcoal. The results of this paper complete the development of methods and they show that it is possible to obtain meaningful environmental signals from the carbon isotope composition of modern and archaeological charcoal. I carried out all sample collection, preparation and analyses and generated all figures and the initial draft. Dr. Stephan Woodborne assisted with data interpretation, editing and was responsible for the development of some the combustion experiments which I carried out. Prof. Mary Scholes and Prof. Lyn Wadley were involved in editing.

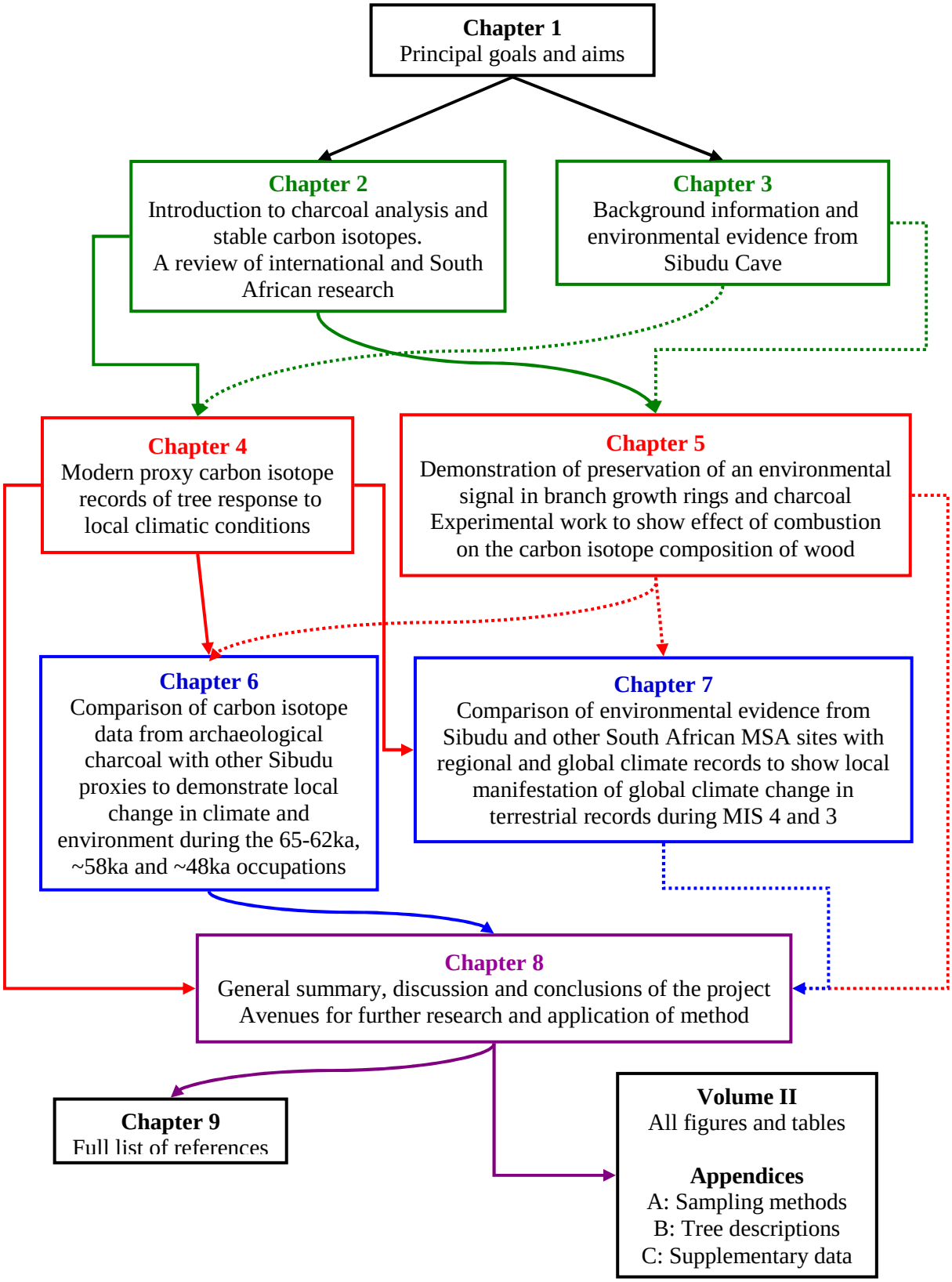
In Chapter 6, manuscript in preparation, the proxy environmental evidence from the carbon isotope composition of the archaeological charcoal from Sibudu is compared with other proxy palaeoenvironmental evidence. It is also demonstrated that there is little or no post-depositional contamination of the archaeological charcoal from Sibudu Cave. The local climatic and environmental changes seen during the Howiesons Poort (65ka-62ka), post-Howiesons Poort (~58ka) and late MSA (~48ka) occupations are presented. I put the synthesis of the Sibudu proxy environmental data together, comparing it with my charcoal isotope data, generated all figures and tables

to create the initial draft. Prof. Lyn Wadley, Dr. Stephan Woodborne and Prof. Mary Scholes assisted with interpretation and editing.

Chapter 7, a paper submitted to *Palaeoecology of Africa*, provides a synthesis of all the results for the project. The archaeological charcoal carbon isotope data and associated proxy evidence from Sibudu Cave along with complementary environmental evidence from other MSA sites in South Africa are compared with a range of regional and global records of climate change. This allows a demonstration of the local manifestation of large scale climate change reflected in terrestrial records during Marine Isotope Stage (MIS) 4 and MIS 3. I put the synthesis of the proxy environmental data from the various MSA sites and other locations together, generated all figures and tables to create the initial draft. Dr. Stephan Woodborne provided in-depth assistance with understanding and relating the non-archaeological environmental evidence to the archaeological material, as well as editing. Prof. Lyn Wadley and Prof. Mary Scholes assisted with interpretation and editing.

In Chapter 8 a summary discussion of the results and conclusions from the above mentioned papers is presented. The merits and contributions of this research are discussed and suggestions for further avenues of research are indicated. A series of three appendices is provided in Volume II. Due to size limitations of publications it is not possible to include extensive details of sampling strategies/methods. This supplementary information is included in Appendix A which provides a detailed explanation of all sampling methods, pre-treatment of wood and charcoal samples for carbon isotope analysis and details of the high-precision radiocarbon dating techniques used. Appendix B contains detailed botanical descriptions and environmental information of the selected modern tree species used in the project. Appendix C contains supplementary data including the range of annualised weather data used in the analyses of the various ecophysiological responses of the modern tree species, as well as a summary of the global and regional records of climate change referred to in Chapter 7.

A flowchart showing the links between the various chapters follows:


Chapter 1
Principal goals and aims
Chapter 2
Introduction to charcoal analysis and stable carbon isotopes.
A review of international and South African research
Chapter 3
Background information and environmental evidence from Sibudu Cave
Chapter 4
Modern proxy carbon isotope records of tree response to local climatic conditions
Chapter 5
Demonstration of preservation of an environmental signal in branch growth rings and charcoal
Experimental work to show effect of combustion on the carbon isotope composition of wood
Chapter 6
Comparison of carbon isotope data from archaeological charcoal with other Sibudu proxies to demonstrate local change in climate and environment during the 65-62ka, ~58ka and ~48ka occupations
Chapter 7
Comparison of environmental evidence from Sibudu and other South African MSA sites with regional and global climate records to show local manifestation of global climate change in terrestrial records during MIS 4 and 3
Chapter 8
General summary, discussion and conclusions of the project
Avenues for further research and application of method
Chapter 9
Full list of references
Volume II
All figures and tables
Appendices
A: Sampling methods
B: Tree descriptions
C: Supplementary data

CHAPTER 2. CHARCOAL ANALYSIS AND STABLE CARBON ISOTOPES

2.1. Charcoal analysis

With improved sampling methods, better understanding of wood anatomy and statistical analytical methods, the contribution of charcoal analysis to archaeological science is increasing. Trees are both adapted to, and responsive to, their environment. These adaptations and environmental responses are recorded in the anatomy (e.g. vessel size) and chemistry (e.g. isotopic composition) of the woody tissues making up the tree. These provide clues to the niche/environment in which the tree grew. There is growing evidence that certain of these physical and chemical characteristics may be preserved in charcoal. Charcoal may be subjected to a range of analyses including; the identification of taxa, anatomical variability and chemical changes. The identification of woody taxa from charcoal from both archaeological contexts and soil profiles may, under ideal conditions, allow researchers to study:

- Former natural ecosystems
- Changes in vegetation communities over time
- The effect of human activities on plants
- The evolution of plant communities
- The importance of wild and anthropogenic fires on vegetation structure and composition (Figueiral and Mossbrugger, 2000).

It is vital to be able to determine whether trends indicated by the charcoal analyses represent environmental effects or are due to anthropogenic factors. In practice it is often difficult to separate the reasons for the accumulation of charcoal and what the assemblages represent. Improved microscopy techniques and equipment have also improved the accuracy and the speed of identification.

There are various terms and definitions used by researchers working with charcoal in related fields. In keeping with the terminology used by archaeologists, the following definitions are used in this study:

Charcoal – the highly recalcitrant by-product of predominantly wood burning in an oxygen-poor atmosphere (Cohen-Ofri *et al*., 2006; Gundale and Deluca, 2006), but other plant material such as grass, fruit and seeds may also form charcoal. Charcoal is formed at temperatures between 350°C and 800°C (Glaser *et al*., 2002) and is generally considered to be relatively inert and resistant to chemical and biochemical processes (Forbes *et al*., 2006). The formation temperatures, duration of burn and source material are all factors that affect the chemical and physical properties of charcoal (Braadbaart, 2004; Braadbaart *et al*., 2004; Czimczik *et al*., 2002; Gundale and Deluca, 2006; McParland *et al*., 2007; Pyne *et al*., 1996; Scott, 2000; Turney *et al*., 2006). More details on the effects of temperature on the chemical and physical properties of charcoal are provided in sections 2.9 and 2.10.

Carbonisation, charcoalification or charring – the heating process by which plant material is converted to relatively stable carbon-rich blackened botanical remains under anoxic conditions at atmospheric pressure (Braadbaart, 2004; Braadbaart and Poole, 2008; Braadbaart *et al*., 2004; Braadbaart *et al.*, 2009; Sievers and Wadley, 2008). These are the terms that are widely used in the archaeological literature, but the process may also be defined as thermal decomposition, thermo-chemical degradation or pyrolysis (Braadbaart, 2004).

The majority of studies utilising charcoal outside of southern Africa have focused on sites in Europe, the Americas and northern Africa. There are a number of research programs in South America, particularly Brazil, as well as in Australia. The majority of these projects are multidisciplinary in nature, utilizing a range of evidence, such as pollen spectra, sediment analyses, phytolith studies, and charcoal analyses and, increasingly, the use of stable carbon isotopic analyses of soils and charcoals. The focus is on environmental reconstruction and tracking of past climatic change, as well as looking at the exploitation of woody species by humans.

Charcoal analysis along with palynological, biological and isotopic studies has been used to reconstruct past Holocene environments in Brazil and Amazonia (Behling and de Costa, 2001; Gouveia *et al*., 2002; Pessenda *et al*., 2004). Charcoal fragments and pollen samples were collected from a range of soil profiles and sediment cores. This

material was analysed to determine changes in the vegetation and climate during the late Pleistocene and Holocene. These studies revealed the complexity of the past vegetation in Brazil and indicated that fire had a definite effect on the various communities, resulting in shifts in the proportions of C_3 versus C_4 species. Fire effects were either a result of drier conditions creating a suitable environment for intense natural fires or due to human activities. A similar, multidisciplinary project has been carried out to provide a Holocene palaeoenvironmental record for the Whitsunday region in Queensland, Australia (Genever *et al*., 2003). Pollen and charcoal analyses provided data on the vegetation and fire histories of the area. Comparisons of charcoals from sediment cores and archaeological charcoals revealed that vegetation burning by humans was not a major cause of changing vegetation patterns, but the observed changes were due to climatic change.

A number of charcoal studies have been carried out in South Africa, predominately on Later Stone Age archaeological sites, dating from the late Pleistocene to the present (Cartwright and Parkington, 1997; Deacon, 1979, 1983a; Deacon *et al*., 1983; Dowson, 1988; Esterhuysen, 1992, 1996; Esterhuysen *et al.*, 1994; Esterhuysen and Smith, 2003; February, 1994a, 1994b; Prior, 1984; Prior and Price Williams, 1985; Tusenius, 1986, 1989; Schackleton and Prins, 1993; Wadley, 2000; Wadley *et al*., 1992). The number of successful projects listed attest to the suitability and usefulness of charcoal analysis as a means of examining past environments of the late Pleistocene and Holocene. One of the main reasons for the concentration on this period is the substantial amount of environmental proxy data available. This includes pollen sequences (Scott, 1982; Scott and Vogel, 1983; Scott and Thackeray, 1987; Scott *et al*., 1995, 2003) and faunal analysis (Avery, 1982, 1988; Klein, 1976, 1977, 1978, 1979, 1980, 1984). It is clear from the various charcoal-based palaeoenvironmental reconstructions that it is important to correlate the results with independent environmental proxy data. These proxy data can be obtained from a number of sources. Several summaries of climatic change in southern Africa during the Quaternary have been compiled, assimilating climatic data from a wide range of sources, such as pollen spectra, soil and sediment profiles, faunal analyses and oxygen isotope analyses (e.g. Abell and Plug, 2000; Deacon and Lancaster, 1988; Klein, 1984; deMenocal, 1995; Scott *et al.*, 1995; Tyson, 1986; Vogel, 1984; Werger, 1978). These summaries provide an understanding of the climatic and environmental trends

seen in southern Africa. They also are of the opinion that there is a need to apply a multi-proxy approach to reconstructing past environments. If such a range of proxy data exists, why should researchers perform charcoal analyses? The analysis of charcoal provides another independent measure of environmental conditions, but the validity of the proxy needs to be demonstrated. Once this has been demonstrated, there is an uniformitarian assumption that the proxies can be extrapolated.

2.2. Introduction to stable carbon isotopes

Stable isotopes have been used in a variety of archaeological (e.g. Lee-Thorp and van der Merwe, 1987, 1993; Sealy, 1986; Sponheimer *et al*., 2005), botanical (e.g. Codron *et al*., 2005c; Jahren, 2004; Loader and Hemming, 2004), palaeoenvironmental (e.g. Brink and Lee-Thorp, 1992; Cerling and Harris, 1999; Hedges *et al*., 2004; Schleser *et al*., 1999) and zoological (Codron *et al*., 2005a, 2005b, 2006, 2007; Sillen and Lee-Thorp, 1994; Sponheimer *et al*., 2003, Wallington *et al*., 2007) studies and the necessary methodology and equipment have been developed and refined over the last four decades. The stable isotopes of carbon, oxygen, hydrogen and more recently nitrogen and sulphur, have been used in the study of ecosystem dynamics to determine diets, habitat preferences and water balance (e.g. Ambrose, 1991; Codron *et al.* 2009; Gebrekirstos *et al.*, 2009; Martín-Benito *et al.*, 2010; Parker *et al.*, 2010; Tieszen, 1991).

The focus of this study is on stable carbon isotopes and the environmental information that can be derived from the isotopic analysis of trees, branches and charcoal, both modern and archaeological. Early carbon isotope research focused on the understanding of the various physiological and biochemical processes involved with the uptake and utilisation of carbon molecules by plants (e.g. Farquhar *et al*., 1988; Francey and Farquhar, 1982; Lajtha and Marshall, 1994; O'Leary, 1981, 1988, 1993; O'Leary *et al*., 1992). Traditional use of stable isotopes from archaeological botanical and faunal material has included the elucidation of dietary pathways (Brink and Lee-Thorp, 1992; Cerling and Harris, 1999, Sealy, 1986; Sponheimer *et al*., 2005) and the tracking of changes in the distribution of vegetation types (Codron *et al*., 2005c; Loader and Hemming, 2004).

Of particular relevance to this study are the carbon isotopic data that can be derived from the analysis of tree ring sequences. The primary value of tree ring isotopes is that they are sensitive bio-indicators of how carbon, oxygen and hydrogen have been modified by the tree in response to the environment. Stable isotope ratios are subject to climatic influences, such as temperature, rainfall, soil chemistry and changes in the local vegetation communities (Hedges *et al.*, 2004). In order to understand the isotopic signals from the modern tree rings and archaeological charcoals, it is necessary to understand some aspects of plant ecophysiology.

2.3. The effect of photosynthesis and fractionation on carbon isotopes

Carbon is taken up from air and converted into wood. It has two naturally occurring stable (non-radioactive) isotopes and each has similar chemical properties. Carbon-12 (^{12}C) is the most abundant, comprising approximately 98.9% of the total and carbon-13 (^{13}C) the remaining 1.1%. The difference in their mass allows physical, chemical and biological processes to discriminate against the ^{13}C isotope (McCarroll and Loader 2004). By convention where R= $^{13}C/^{12}C$ ratio, measurements on samples are expressed in delta notation ($\delta^{13}C$) as the deviation from the isotopic ratio of a reference standard known as Vienna Pee Dee Belemnite (VPDB) (Coplen, 1995) and using a per mille scale

$$\delta^{13}C\ (‰)_{PDB} = (R_{sample}/R_{standard} - 1) \times 1000$$

There are three photosynthetic pathways used by plants, C_3 (Calvin cycle), C_4 (Hatch-Slack cycle) and CAM (Crassulacean acid metabolism). Of these the C_3 and C_4 pathways are important to this study. These pathways involve interaction of atmospheric CO_2 with different enzymes and results in C_3 and C_4 plants having significantly different $^{13}C/^{12}C$ ratios (Ehleringer and Vogel, 1993; Lajtha and Marshall, 1994; Tieszen, 1991). It is these clear differences that allow one to distinguish between two of the plant types. C_3 plants have $\delta^{13}C$ values ranging from –34‰ to -23‰ and C_4 plants have $\delta^{13}C$ values from –9‰ to -17‰ (Ehleringer and Rundel, 1988). The isotopic variations seen between these two pathways are due to both environmental and genetic components (Ehleringer and Vogel, 1993; O'Leary, 1993; McCarroll and Loader, 2004). Most trees and woody plants utilise the C_3

photosynthetic pathway, in which atmospheric CO_2 is converted into the three-carbon molecule, ribulose-1, 5-biphosphate carboxylase oxygenase (RuBisCO). It should be noted that some tree species could utilise the C_4 photosynthetic pathway (e.g. Pearcy and Throughton, 1975). The carbon formed by this process is transported from the leaves to the stem, where it is stored in the plant tissues.

The use of stable carbon isotopes from tree sequences as an indicator of past climatic conditions is based on the discrimination against a particular carbon isotope during the photosynthetic process, also known as fractionation. There is a two-step fractionation of carbon (Fig. 2.1, Vol. II, pg. 3).

The first occurs during the uptake of CO_2 through the stomata of the leaves. As CO_2 diffuses through the stomata, there is a discrimination against $^{13}CO_2$ as the $^{12}CO_2$ isotope diffuses more rapidly into the leaves than the heavier $^{13}CO_2$ isotope. If the stomata are closed, such as in times of water stress, the tree has to make more use of the CO_2 trapped within the leaves, rather than from the atmosphere. This results in an increase in the use of ^{13}C for carbohydrate synthesis rather than the selective use of ^{12}C. Carbon isotope values will become less negative, reflecting some form of environmental water stress. More negative values are usually indicative of non-stressful (water) growing conditions. However, it is noted that increased levels of precipitation and excess soil moisture stress may result in reduced stomatal conductance reflected by ^{13}C enriched $\delta^{13}C$ values (Buhay *et al*., 2008). Most studies of trees from warm and drier climates relate the variations seen in $\delta^{13}C$ values to variations in moisture availability affecting stomatal conductance (Gagen *et al*., 2008). This is seen as a negative correlation between isotope values and relative humidity and precipitation (Tardif *et al*., 2008). In colder and wetter climates it appears that the $\delta^{13}C$ values are controlled by changes in photosynthetic rates rather than stomatal conductance (Gagen *et al*., 2008; Tardif *et al*., 2008). There, variations in carbon isotope values are related to variation in temperature and hours of sunshine (Gagen *et al*., 2008).

The second fractionation takes place during carbon fixation, when the ^{12}C isotope is more inclined to react with the photosynthetic enzyme (RuBisCO). These two key fractionations are expressed in the following equation (Farquhar *et al*., 1982):

$$\delta^{13}C_{plant} = \delta^{13}C_{atmosphere} - a - (b-a)\, c_i/c_a$$

The current value for $\delta^{13}C_{atmosphere}$ is considered to be -8‰, based on the use of VPDB standard (Coplen, 1995; McCarroll and Loader, 2004). The values for *a* and *b* represent the two fractionation events, where *a* (4‰) represents the discrimination against $^{13}CO_2$ during diffusion via the stomata and *b* (27‰) is the net discrimination due to carboxylation. The values for c_i and c_a represent the intercellular and ambient atmospheric CO_2 concentrations respectively. Since *a* and *b* are constant values the total fractionation is controlled by the relationship between c_i and c_a. If the intercellular CO_2 concentration (c_i) is higher than the ambient concentration (c_a), then there is an increase in the diffusion rate of CO_2 through the stomata. This results in a strong discrimination against ^{13}C during carboxylation, as more ^{12}C is available, causing higher $\delta^{13}C$ values. Tree ring $\delta^{13}C$ values are clearly depleted in ^{13}C relative to the ambient atmosphere. The magnitude of this discrimination seen in trees is a direct function of the ratio of intra-cellular CO_2 concentration in the leaves to the concentration of CO_2 in the atmosphere (Buhay *et al*., 2008). Although this ratio is affected by environmental conditions, it may be under some genetic control (Farquhar, 1991; Farquhar *et al.*1988). This allows the possibility to use measurements of discrimination to screen variation in water-use efficiency (WUE) The application of WUE studies using stable isotope analysis of trees has important ramifications, particularly for agriculture and forestry in regions where water resources are limited. These include forest ecological responses to changing climates (e.g. Battipaglia *et al.*, 2009), the selection of appropriate species for cultivation (e.g. Cui *et al.*, 2009;, Tanaka-Oda *et al.*, 2010) and plantation resource management (e.g. Forrester *et al.*, 2009; Jassal *et al.*, 2009; Kunert *et al.*, 2010; Martín-Benito *et al.*, 2010).

Local environmental factors such as irradiation, humidity, precipitation, air pollution, salinity, canopy cover and soil salinity may also have an influence on tree $\delta^{13}C$ values (Livingstone *et al*., 1999; West *et al*., 2001). The $\delta^{13}C$ values of a particular tree will vary according to a variety of factors that influence its stomatal conductance and photosynthetic rates (Fig 2.2, Vol. II, pg. 4)). The degree of influence of these factors will depend on conditions at the specific site where the tree grows. For example, if

water availability is the limiting factor, fractionation will largely be controlled by the rate of stomatal conductance and the $\delta^{13}C$ values may provide a record of variations in humidity, soil moisture and rainfall (Livingston and Spittlehouse, 1993). The $^{13}C/^{12}C$ ratios record the balance between stomatal conductance and the plant's rate of photosynthesis. In dry areas this balance is affected by relative humidity and soil water states, while in wetter areas, the amount of summer irradiance and temperature are responsible (McCarroll and Loader, 2004). The basis of this assumption is that during times of water stress, the stomata of the plant will close to limit loss of water via transpiration, reducing the uptake of CO_2 and resulting in more positive $\delta^{13}C$ values.

2.4. Corrections currently applied to $\delta^{13}C$ data

An important factor to note is the close link between the $\delta^{13}C$ values of the tree and the isotopic composition of the atmosphere (Arens *et al.*, 2001). There has been a global anthropogenically driven increase in the CO_2 concentration of the atmosphere. This has led to a decrease in the ^{13}C of atmospheric CO_2 by 1.5‰ since industrialization (~1850) (Epstein and Krishnamuthy, 1990; Freyer and Belacy, 1983. This trend is continually monitored and characterised (e.g. Carter, 2009; Gebrekirstos *et al.*, 2009; Gagen *et al.*, 2007; Loader *et al.*, 2007; Treydte *et al.*, 2001; 2009). These values then began to decrease rapidly from *c.a.* 1950 due to a more substantial contribution of isotopically depleted CO_2 in the atmosphere from amplified industrialisation (Loader *et al*., 2008; Rakowski *et al*., 2008). The anthropogenic alteration of atmospheric CO_2 as a result of burning fossil fuels (Buhay *et al*., 2008; February and Stock, 1999; Gagen *et al*., 2008; Leavitt and Lara 1994; Loader *et al*., 2008; Rakowski *et al*., 2008) is collectively known as the Suess effect. It was first identified in radiocarbon (^{14}C) concentrations from tree rings (Suess, 1955; Keeling, 1979). In terms of more recent carbon isotope time series from modern trees, the anthropogenic effect needs to be removed from isotope data (e.g. Buhay *et al*., 2008; Gagen *et al*., 2008; Kirdyanov *et al*., 2008; Loader *et al*., 2008; Liu *et al*., 2004; Tardif *et al*., 2008). This is accomplished by a mathematical adjustment of $\delta^{13}C_{plant}$ values for changes in $\delta^{13}C_{atmosphere}$ determined from ice cores and direct measurements (Loader *et al*., 2008; McCarroll and Loader, 2004, 2006).

In addition to the Suess effect, there is increasing evidence of a physiological response in trees to increased atmospheric CO_2 concentrations, which may affect fractionation. Unlike the Suess effect, this effect needs to be statistically detrended from time series. There are two ways that this may be achieved. The first method implicitly assumes that all trees will demonstrate the same uniform and linear response to changing CO_2 concentrations. Accounting for this response requires the addition of a fixed correction to $\delta^{13}C$ values per unit increase in CO_2. However, with an increase in the numbers of tree ring $\delta^{13}C$ series available, there is sufficient evidence to suggest that a tree's response to increased CO_2 is heterogeneous and nonlinear (McCarroll *et al.*, 2009). The second method is based on the conversion of tree ring $\delta^{13}C$ data into values for internal CO_2 concentration (c_i) and then on the estimation of the values of c_i that would have been obtained under pre-industrial conditions (Loader *et al.*, 2008; McCarroll *et al.*, 2009). This correction uses nonlinear regression, but is limited by two constraints based on the physiological response of the tree. First, a unit increase in the atmospheric CO_2 concentration (c_a) cannot result in more than an equivalent increase in c_i and, second an increase in water-use efficiency, due to an increase in c_a, is limited to the maintenance of a constant c_i/c_a ratio (McCarroll *et al.*, 2009).

As well as the above corrections, there is an increasing application of a further correction to minimise the isotope juvenile effect, an age related influence of carbon isotope values. During the first few decades of growth, young trees exhibit rapid branch elongation and height gain and consistently show depleted, but progressively less negative $\delta^{13}C$ values. These initially depleted $\delta^{13}C$ values are generally considered to be as a result of exposure to ^{13}C-depleted CO_2 respired from the forest floor and growth in a shaded environment (Buhay *et al.*, 2008; Gagen *et al.*, 2008). There is, however, evidence for an isotope juvenile effect in trees growing in more open, exposed areas and this is thought to be due to changes in hydraulic conductance as the tree matures affecting stomatal conductance (Gagen *et al.*, 2008). There are a variety of methods employed to remove this trend from time series. The simplest method is referred to as "juvenile cutting" and involves the exclusion of data from the first few decades of a time series taken from pith to bark. This method limits the length of a time series. An alternative method is to use statistical methods, such as Regional Curve Standardisation, to remove the juvenile effect (Gagen *et al.*, 2008).

2.5. Applicable international research

Over the last five decades a large archive of tree ring related data has been developed, primarily derived from studies carried out in the Northern Hemisphere. These include research into the improvement and testing of existing methodologies and experimentation to develop new methods (Dodd *et al.*, 2008; Li *et al.*, 2005; Loader *et al.*, 2003; Rinne *et al.*, 2005; Roden, 2008; Schleser *et al.*, 1999, Van Der Water, 2002). A substantial archive of research covering climatic records from modern tree ring time series provides ample evidence for the importance of such research (e.g. Anderson *et al*., 2005; Edwards *et al*., 2000; Guo *et al.*, 2005; Guo and Xie, 2006; Kirdyanov *et al*., 2008; Leavitt, 2002; Lipp *et al*., 1996; Liu *et al*., 2004; Loader *et al*., 2008; Poussart *et al*., 2004;Robertson *et al*., 2004; Stuiver & Braziunas, 1987; Tardif *et al*., 2008; Tarhule & Leavitt, 2004; Van Bergen & Poole, 2002; Weigl *et al*., 2008). Additional avenues of research include genetic and physiological responses to the environment, reflected in the isotopic composition (e.g. Brendel *et al.*, 2002; Comstock and Ehleringer, 1992; Dupouey *et al.*, 1993; Leavitt and Long, 1982; Pennington *et al.*, 1999) and the reconstruction of past environments (e.g. Ando *et al.*, 2003; Bechtel *et al.*, 2002; 2003; Danis *et al.* 2006; Feng and Epstein, 1995; Hunter *et al.*, 2006; Lockheart *et al.*, 1998; Mazany *et al.*, 1980; Nguyen Tu *et al.*, 2002; Van Der Water *et al.*, 1994). A summary of studies applicable to this project is provided in Table 2.1., Vol.II, pg. 38.

There is an increasing tendency for palaeoclimatic interpretations of modern, archaeological or fossilised material to be based on multiple strands of dendrological evidence from tree rings and other plant material (e.g. charcoal and branches). This has resulted in the combination of traditional dendrological data, such as ring widths, wood density and anatomy with stable isotopic data (carbon, oxygen and hydrogen) obtained from tree ring sequences. Dating methods, sampling techniques, sample processing and mass spectrometers have been greatly improved in recent times and it is possible to obtain large isotope data sets from larger sample sets. This has resulted in high resolution, well-dated times series that can be statistically correlated with a range of climatic and environmental variables. Non-climatic influences, the anthropogenic alteration of atmospheric CO_2 and the juvenile effect are now better

understood and most recent studies account for these prior to further analyses (Dongarra and Varrica, 2002; Dupouey *et al*., 1993; Gagen *et al*., 2008; Li *et al*., 2004 2005; Rakowski *et al*., 2008; Zhao *et al*., 2006). These improvements have allowed researchers to obtain detailed proxy records from tropical tree species that do not form clear annual growth rings (Poussart and Schrag, 2005; Poussart *et al*., 2004; Robertson *et al*., 2004).

It has been well established that $\delta^{13}C$ values record a plant's response to available moisture levels. This includes variation in precipitation, soil moisture, water-use efficiency and relative humidity, thereby allowing the reconstruction of past moisture conditions. In general $\delta^{13}C$ values become more negative during wetter periods and less negative during more arid times (e.g. Buhay *et al*., 2008; Comstock and Ehleringer, 1992; Dupouey *et al*., 1993; Feng and Epstein, 1995; Lipp *et al*., 1996; Liu *et al*., 2004). There are exceptions to this tendency for a negative correlation and they are due to additional environmental factors. Anderson *et al*. (2005) found a positive correlation between $\delta^{13}C$ and rainfall for a species that was salt wetland adapted. It has been demonstrated that the genetic adaptability of a species plays a role in the response to changing water availability (Brendel *et al*., 2002; Pennington *et al*., 1999). With more sophisticated sample strategies and mass spectrometers it is possible to create $\delta^{13}C$ time series indicating a record of intra-annual and seasonal response to shifts in moisture availability (e.g. Danis *et al*., 2006; Dodd *et al*., 2008; Garciá-G *et al*, 2004; Leavitt, 2002; Li *et al*., 2005; Tarhule and Leavitt, 2004). Although moisture availability is one of the dominant factors influencing the carbon isotope composition of tree rings, other environmental variables also have an effect. These include altitude (Garciá-G *et al*., 2004; Guo and Xie, 2006); latitude (Stuiver and Braziunas, 1987) and temperature, particularly during the summer growing season (Edwards *et al*., 2000; Leavitt and Long, 1982; Loader *et al*., 2008; Pearman *et al*., 1976; Tardif *et al*., 2008).

2.6. Climatic and environmental reconstruction using carbon isotopes

The majority of tree ring-based isotope research from the Northern Hemisphere has shown that $\delta^{13}C$ time series provide a proxy record of a number of climatic and environmental variables such as precipitation, temperature, humidity and changes in atmospheric CO_2. These data sets are a representative isotopic record of the plant's ecophysiological response to long-term, low frequency (decadal to centuries) and short-term, high frequency (seasonal, inter/intra-annual) variations in local and regional climate. Low frequency trends include changes in atmospheric CO_2 concentration, climatic conditions and isotopic composition (e.g. Dongarra and Varrica, 2002; Gagen *et al.*, 2007), while high frequency trends include seasonal shifts in moisture availability and temperature (e.g. Feng and Epstein, 1995; Leavitt, 2002).

2.7. Wood tissues: what to use?

Initially wholewood was used for stable isotope analyses of tree rings (Craig, 1954), but later research revealed that there are differences in the isotopic composition of the tissues forming wood (Wilson and Grinsted, 1977). Wood is composed of a range of material, including cellulose, lignin, hemicellulose, resins and tannins formed by a number of biochemical processes. These formation processes result in the components having differing isotopic compositions. Cellulose is typically ^{13}C enriched and lignin is ^{13}C depleted compared to wholewood (Benner *et al.*, 1987; Loader *et al.*, 2003; Van de Water, 2004; Wilson and Grinsted, 1977). The majority of recent studies use cellulose (e.g. Brendel *et al.*, 2002; Buhay *et al.*, 2008; Edwards *et al.*, 2000; Kirdyanov *et al.*, 2008; Leavitt, 2002; Lipp *et al.*, 1996; Liu *et al.*, 2004; Loader *et al.*, 2008; Mazany *et al.*, 1980; Roden *et al.*, 2008; Tardif *et al.*, 2008; Zhao *et al.*, 2006) as this is relatively easily extracted from wholewood, has a high level of homogeneity, and can be accurately linked to a specific growing period (McCarroll and Loader, 2004). With regard to trees with clear earlywood and latewood sections visible, the favoured portion to sample is the latewood (Weigl *et al.*, 2008) as this provides the best annual response. However, there is some evidence that earlywood tissue may be used (Helle and Schleser, 2004; Kress *et al.*, 2009; Robertson *et al.*,

2008). There is still no consensus amongst researchers as to which chemical constituent to use.

Although the majority of studies use cellulose, the use of wholewood is also valid, particularly given recent developments in on-line combustion methods and highly sensitive mass spectrometers (Leuenberger *et al*., 1998; McCarroll and Loader, 2004; Schleser *et al*., 1999). There is evidence from a number of species that a strong correlation exists between $\delta^{13}C$ values of cellulose and wholewood, both showing similar annual resolution (Cullen and Grierson, 2006, Loader *et al*., 2003, Verheyden *et al*., 2004, 2005). Cullen and Grierson (2006) suggest that wholewood be used when examining long term trends and cellulose is more suited when looking at high-frequency trends and responses to specific climatic events such as extreme drought and floods. Loader *et al*. (2003, 2004) have shown that wholewood may preserve the strongest climatic signal.

2.8. Stable carbon isotope proxy data from plant fossils and charcoal

The use of $^{13}C/^{12}C$ ratios from tree rings to trace past climatic conditions implies that wood tissue is isotopically invariable. While this may be the case for modern wood, it may not be so for fossil wood, leaves or charcoal that may have undergone a number of post-depositional changes (i.e. diagenesis). An artificial aging experiment in which four modern tree species (Schleser *et al.*, 1999) were aged in water kept at 180°C over various times showed that changes in the isotopic composition of the wood did occur. A two-stage isotopic change occurred during decomposition, an initial strong depletion of ^{13}C and then a gradual enrichment of ^{13}C, which slowly masks the initial depletion. A study of archaeological and fossil wood from Tertiary and Cretaceous sites (Van Bergen and Poole, 2002) showed similar changes in isotope composition due to molecular changes in wood structure that occurred over time. Such change in isotopic ratios of archaeological and fossil wood need to be understood clearly if such isotopic data are to be used in palaeoenvironmental reconstruction. Nevertheless the $\delta^{13}C$ analysis of various fossilised wood, leaves and charcoal extracted from soil organic matter dating to the Miocene and Pliocene has shown temporal variations in vegetation communities and environment (Ando *et al*., 2003; Bechtel *et al*., 2002, 2003; Lockheart *et al*, 1998; Lücke *et al*., 1999; Nguyen-Tu *et al*. 2002). Evidence for

shifting proportions of C_3 and C_4 plants due to changes in aridity and temperature during the Holocene in North and South America has been obtained from $\delta^{13}C$ data from charcoal, wood and leaf samples (Biedenbender *et al*., 2004; Gouveia *et al*., 2002; Hunter *et al*., 2006; Van de Water *et al*., 1994; Wooler *et al*., 2004).

The $\delta^{13}C$ values of archaeological charcoal have been used to quantify changes in past water availability (Aguilera *et al*., 2009; Ferrio *et al*., 2006). Ferrio *et al*. (2006) initially carried out a study of modern *Pinus halpensis* (Aleppo pine) cores from areas in the north-western Mediterranean Basin, with differing water availability, examining the effects of carbonisation on carbon isotope composition over a range of temperatures. It was established that, despite significant changes in $\delta^{13}C$ due to carbonisation, the original climatic signal of the wood was preserved. Identified archaeological charcoal (*P. halpensis*) from seven well-dated archaeological sites (Bronze Age to Modern Age) in the same region was then analysed and the $\delta^{13}C$ values indicated that water availability in the past was higher than present values (Ferrio *et al.*, 2006). Further research using the carbon isotope composition of identified oak charcoal (*Quercus ilex/coccifera*) from 11 contemporary archaeological sites in eastern Spain was used to create a map of autumn precipitation during the transition between the Bronze and Iron Ages (ca. 900-300 BCE) (Aguilera *et al.*, 2009). An empirical model of seasonal changes in precipitation recorded in the $\delta^{13}C$ values of extant *Q. ilex* was developed and applied to the $\delta^{13}C$ charcoal data from the archaeological sites. This was done to develop a spatial map of past precipitation in the region. The archaeological charcoal $\delta^{13}C$ values preserved an original environmental signal that indicated that precipitation in the region was higher in the past (Aguilera *et al.*, 2009). This suggests that a coherent environmental signal is preserved in ancient wood and charcoal, regardless of post-depositional changes and the effects of carbonisation.

2.9. The effect of combustion on stable isotope composition

A range of experimental work has been done to examine the effects of charring on wood and other plant material and how this may alter the isotopic composition of the source material and the products of combustion (e.g. Ballentine *et al*. 1998; Poole and Van Bergen, 2006; Czimczik *et al.*, 2002; Gundale and Deluca, 2006; Krull *et al*.,

2003; McParland *et al*., 2007; Poole *et al*., 2002; Turekian *et al*., 1998; Turney *et al*., 2006). In most cases it was found that the combustion temperature had the greatest effect on carbon isotope values of both source material and the products released during combustion (Braadbaart, 2004; Braadbaart *et al*., 2004; Gundale and Deluca, 2006; Pyne *et al*., 1996; Scott, 2000). The length of the combustion period may also affect isotopic composition, although researchers have produced conflicting results (Czimczik *et al*., 2002; Turney *et al*., 2006).

Czimczik *et al*. (2002) subjected modern hard and softwoods to charring under pyrolitic (oxygen free) conditions at various temperatures. The mass of the wood samples decreased from 7 to 84% depending on the temperature. Samples charred at temperatures below 150˚C had more positive $\delta^{13}C$ values, while those charred at temperatures above 150˚C were more negative. This was ascribed to the loss of isotopically heavier cellulose during the charring at higher temperatures. However, in the case of the research carried out by Czimczik *et al.* (2002), all the material was heavily treated prior to charring which was done under extremely controlled laboratory conditions and this is not necessarily representative of a natural fire.

Conflicting results were obtained from a laboratory study conducted by Turekian *et al.* (1998) where no isotopic differences were seen in C_3 plants over a range of combustion intensities, but C_4 plants showed a combustion-induced fractionation effect. Analysis of charred material collected in Australia from a variety of natural fires and controlled natural fires has produced different $\delta^{13}C$ values for charring from both C_4 (grasses) and C_3 (woody species) (Krull *et al.*, 2003). As opposed to charred material produced under pyrolitic conditions, the majority of material analysed was charred under natural conditions, in open flames with oxygen present. Charred material produced under pyrolitic conditions was also analysed to determine what differences in $^{13}C/^{12}C$ ratios may occur. The results showed that there were significant differences in the isotopic ratios of charred wood produced by the two methods. The C_4 charcoals from grasses had very ^{13}C-depleted $\delta^{13}C$ values, which was ascribed to the presence of highly ^{13}C-depleted organic carbon present in the phytoliths found in grasses. Das *et al*. (2010) recorded similarly ^{13}C-depleted $\delta^{13}C$ values for C_4 grasses combusted in laboratory experiments. The C_3 $\delta^{13}C$ revealed no significant isotopic change.

The results obtained by Krull *et al.* (2003) are encouraging and support the notion that significant isotopic data can be obtained from archaeological charcoals. From the range of identified species, it is clear that they are all woody C_3 species that were charred in natural oxygen rich fires. Therefore, $^{13}C/^{12}C$ ratios obtained from any archaeological charcoal samples should not theoretically have undergone any significant fractionation and the results would reflect climatic conditions, rather than the effects of fire temperature.

2.10. Pilot studies and experimental research in southern Africa

In southern Africa and Africa as a whole, there has been limited research into the use of tree rings and stable carbon isotopes as a climate proxy (Table 2.2, Vol. II, pg. 47). High seasonal temperature variations in the temperate regions of the Northern Hemisphere create distinct boundaries in the growth ring sequences of the various tree species used for dendroclimatological research. Earlywood tissue (lighter in colour) laid down at the start of the growing season is easily distinguished from the generally denser and darker latewood, produced towards the end of the growing season. This feature greatly eases the development of chronologies and hence the interpretation of data obtained from either an anatomical or isotopic analysis of the tree ring sequence. The situation in southern Africa is not as easily resolved because much of the region is tropical or subtropical. The formation of clearly visible and coherent growth ring sequences in tree species from tropical and subtropical regions is not a common occurrence and, in those species that do have a visible ring structure, interpretation is often difficult (Lilly, 1977). In more recent studies a number of climate-sensitive (particularly to rainfall) dendrochronologies have been established for several African tree taxa using growth ring width and density. Chronologies from two bushveld taxa, *Burkea africana* and *Pterocarpus angloensis*, showed a response to rainfall, relative humidity and temperature in Namibia (Fichtler *et al*., 2004). A 200-year rainfall response chronology was obtained for *P. angloensis* in Zimbabwe (Therrell *et al*., 2006). In addition, West African tree ring width of savanna taxa showed a significant correlation to annual rainfall patterns (Schöngart *et al*., 2006). Tree ring $\delta^{13}C$ values from several deciduous and evergreen species from semi-arid *Acacia* Woodland in Ethiopia showed a significant negative correlation with local precipitation patterns,

but not with temperature or relative humidity (Gebrekirtos *et al.*, 2009). A problem with southern African tree ring sequences is establishing an accurate chronology, particularly when given the lack of clearly defined growth rings. This can be overcome through high precision radiocarbon dating of selected rings (Vogel *et al*., 2001; Robertson *et al*., 2004; Norström *et al*., 2005).

Although some complications of interpretation exist, a number of successful tree ring studies have been carried out in southern Africa, utilizing both stable light isotopes and more traditional dendrological analyses such as ring widths and wood density. One of the earliest studies was the establishment of a 400-year long chronology reflecting rainfall patterns in the southern Cape using *Widdringtonia cedarbergensis* (Dunwiddie and LaMarche, 1980). Stable carbon isotopes in a 77-year long sequence of annual tree rings from *W. cedarbergensis* (February and Stock, 1999) showed a decreasing trend in $\delta^{13}C$ values due to depleted atmospheric carbon isotope composition, but no clear connection was demonstrated between the isotope data and climatic factors such as rainfall, suggesting that water availability is not the major limitation for growth of this taxon. This indicates that care should be taken in the selection of suitable species for isotopic analysis and the environmental requirements of such species needs to be understood.

The relationship between rainfall and $\delta^{13}C$ was tested by February (1997), who subjected two eucalyptus species to an artificial watering program over a 16-month period. Carbon isotope ratio data from leaf tissue, leaf cellulose and wood cellulose became more negative under wet conditions and more positive under drier conditions. The results suggest that $\delta^{13}C$ ratios may be a useful indicator of water consumption. An earlier study by Le Roux *et al*. (1996) produced similar results.

A study of $\delta^{13}C$ ratios for archaeological charcoal samples from three sites in the Elands Bay area, from the past 4000 years, indicated that there were periods of differing rainfall during that period (February, 1992; February and van der Merwe, 1992). The temporal variability in charcoal $\delta^{13}C$ values are considered to be climatically induced. To test the validity of these results, further experimental work was carried out, using archaeological charcoal data from archaeological sites located in the eastern part of South Africa (February, 1994a, b, 1997). Two species of wood

were identified as being the dominant species used for fuel at the sites, namely *Protea roupelliae* and *Combretum apiculatum*. Modern samples of these two species were collected along a rainfall gradient from the summer rainfall region of South Africa, close to areas with good rainfall records. Fresh wood and charred samples were subjected to isotopic analysis to determine whether there was any fractionation of carbon isotopes during pyrolysis. It was found that the $\delta^{13}C$ values for *P. roupelliae*, which can occupy a range of habitats, did not show any correlation with rainfall, but a significant correlation exists between xylem vessel size and frequency and rainfall (February, 1992). The results also indicate that stomatal control is not the only adaptive strategy, as in this case xylem vessel size can offset any stomatal response. *C. apiculatum* is more habitat-specific and $\delta^{13}C$ values show a significant link with rainfall (February, 1997). The differing results may likely be due to the habits of the two species. *C. apiculatum* is drought deciduous, losing its leaves in the dry season, whereas *P. roupelliae* tends to keep its leaves for a number of seasons. This may affect the isotopic composition, as the older leaves may retain ratios from previous years, which may be distributed to the plant at a later stage. This again indicates the necessity to understand the habits and environmental requirements of potential species for isotopic analyses.

The modern charcoal isotope data did not yield any significant results and February concluded that it would not be possible to use $\delta^{13}C$ values of archaeological charcoals as an indicator of past climatic conditions, however no archaeological charcoal was analysed (February, 1997). The results from the modern charcoal indicated that there was some degree of fractionation during the charring process which increased as temperature was raised (February, 1997). This may be cause for concern regarding the reliability of any isotopic data derived from charcoals, as the variation seen in $\delta^{13}C$ values may be reflecting the effects of fire temperatures rather than climate. This problem was addressed through a series of combustion experiments and the results are presented in Chapter 4.

$\delta^{13}C$ values from tree rings have also be used as a means to determine past regeneration environments in forested environments in South Africa (West *et al.*, 2001) and the values are based on differences in the isotope juvenile effect between individual trees. Using six species from two different forest types, a subtropical forest

in Kwazulu-Natal and a temperate forest in the Western Cape, the researchers demonstrated the potential to distinguish between shade-tolerant and shade-intolerant species. These differences allow one to determine whether individual trees originated in open or closed parts of a forest. The combined effects of shading, water-use efficiency and CO_2 recycling below the canopy, produces a more negative $\delta^{13}C$ signal (West *et al*., 2001). Thus a tree initially growing in a closed environment will have more negative $\delta^{13}C$ values as a juvenile and as the tree grows taller into and above the canopy, the $\delta^{13}C$ values will become more positive.

The water stress history of a dry adapted species, *Acacia erioloba* from southern Namibia was determined through stable carbon isotope analysis (Woodborne and Robertson, 2000, 2001; Woodborne *et al*., 2003). The $\delta^{13}C$ data from an *A. erioloba* specimen showed that values appeared to correlate well with rainfall patterns, rather than exclusively with groundwater levels as was originally thought (Woodborne and Robertson, 2000, 2001). Isotopic analysis of a number of *Pinus* species from two riparian zones (Woodborne *et al*., 2003) also showed that $\delta^{13}C$ time series from tree rings reflected the degree of water stress and response to variations in annual rainfall.

More recently, $\delta^{13}C$ time series and wood anatomy from two specimens of *Breonadia salicina* from the Limpopo Province were examined for their potential as a record of past water stress and rainfall response (Norström, 2008; Norström *et al*., 2005). The trees were dated using high precision radiocarbon dating techniques, rather than using ring counts as the authors claim that this species does not produce rings on an annual basis. The results revealed that climate changes seen in regional palaeoclimatic studies were reflected in the $\delta^{13}C$ time series and wood anatomy. A link to the annual rainfall patterns of the last century was also reflected in the isotopic and anatomical data. An additional indigenous species, *Adansonia digitata* (Baobab), sampled from the subtropical Mpumulanga Province produced $\delta^{13}C$ time series from annual rings showing a positive correlation with local rainfall patterns. These results suggest that *A. digitata* has the potential to produce long term proxy records of past climates with high temporal resolution (Robertson *et al*., 2006).

2.11. Conclusion

The above examples indicate that there is potential for African woody taxa to provide detailed palaeoclimatic proxy records. To improve the currently available archive of proxy evidence from such species, it is necessary to create well-resolved, high-resolution isotope and dendrological time series from more southern African species. The review of both local and international research has indicated that there are several aspects that need to be addressed in order to demonstrate the potential of archaeological charcoal as a source of proxy palaeoenvironmental evidence. These are:

- Selection of suitable species for isotopic analysis
- Creation of age models for selected species and determination of annual growth rings
- Selection of appropriate wood tissues for isotopic analysis (i.e. wholewood versus α-cellulose)
- Examining the effect of local environmental factors (e.g. precipitation, temperature, humidity) on the $\delta^{13}C$ values of tree rings and recording of environmental response
- Accounting and correcting for changes in the concentration of atmospheric CO_2, namely the Suess effect and the physiological response of trees to increased CO_2 concentrations
- Accounting for the isotope juvenile effect
- Examining the effects of combustion (temperature and length) on the isotopic composition of wood when converted into charcoal
- Examining the effects of burial on the isotopic composition of charcoal in an archaeological context
- Accurate identification of charcoal recovered from an archaeological site
- Comparison of the charcoal-derived palaeoenvironmental data with other palaeoenvironmental proxies

These aspects are addressed in the following chapters of this thesis. Chapter 4 deals with a number of key issues relating to the interpretation and understanding of

isotopic time series from modern tree species from southern Africa. In terms of the selection of suitable tree species for isotopic analysis, there are only a few species that have been well studied. The presence of annual growth rings and a simple age model was created for the species (*Mimusops caffra*) investigated in Chapter 4 through the use of conventional radiocarbon dating techniques. Through experimental work, it was demonstrated that wholewood was an appropriate tissue to use for further isotopic analyses and the isotope time series from *M. caffra* did provide a record of local environmental factors, particularly rainfall. The Suess and isotope juvenile effects, as well as any potential physiological responses to changing CO_2 concentrations were dealt with using Wavelet analysis.

Having addressed these issues, additional isotopic environmental time series were obtained from several *Podocarpus* specimens using wholewood. These results are presented in Chapter 5. This chapter also covers the effects of combustion on the isotopic composition of wood when converted into charcoal and demonstrates that both modern and archaeological charcoal from Sibudu Cave preserves a meaningful environmental signal. In Chapter 6 a sub-sample of the Sibudu charcoal samples used in Chapter 5 are re-analysed to determine the effects of post-depositional processes on the carbon isotope composition. The charcoal-derived isotope data are then compared with additional published environmental proxy data from Sibudu Cave showing that evidence of similar environmental change is recorded in the charcoal $\delta^{13}C$ data as well as by the proxies. In Chapter 7 a link between global climatic changes and a local manifestation of such change from the MSA layers of Sibudu Cave is demonstrated and potential causes of such change suggested.

CHAPTER 3. BACKGROUND TO SIBUDU CAVE

3.1. Sibudu Cave location, environment and geology

Sibudu Cave is located on the eastern face of a forested sandstone cliff in KwaZulu-Natal, approximately 40 kilometres north of the city of Durban (Fig. 3.1, Vol. II, pg. 5). The site opens in a south-westerly direction and overlooks the Tongati River, about 20m below the entrance. The modern coastline is 15km away. Although the site is known as Sibudu Cave, it is technically a large rock shelter. The shelter is 55m in length and approximately 18m wide, sloping abruptly from north to south (Fig. 3.2, Vol.II, pg. 6). The majority of the archaeological deposit is located in the upper, northern portion of the shelter.

The vegetation that currently surrounds Sibudu Cave is the remnant of a mixed forest community. The adjacent terrain is not suitable for sugarcane fields and has provided a refuge for indigenous vegetation, although there is the danger of encroachment by exotic plant species. The current vegetation community is also considered to be representative of a climax community, as indicated by the presence of *Celtis mildbraedii* (Natal Stinkwood), a species occurring in mature forests (Allott, 2004; Wadley, 2004). A climax plant community is a stable group of species that is self-perpetuating under the present environmental conditions (Smith, 1980). A rich diversity of taxa is to be found, representing a range of vegetation types, such as canopy species, woody lianas and climbers, forest margin species and representatives of woodland, bushveld and thicket vegetation (Acocks, 1988; Camp, 1999; Lubke and McKenzie, 1998; Mucina *et al.* 2006; Wadley, 2004, 2006). There are indications that in the past the Sibudu environment underwent a number of changes in the structure of local plant communities each culminating in a stable climax community as a result of environmental change.

The Sibudu region falls into the Indian Ocean Coastal Belt, an area of approximately 800km in length, from the Mozambique border to the Great Kei River mouth. Much of the original vegetation has been lost due to anthropogenic activities. The region was most recently affected by the agricultural activities of Iron Age inhabitants. What remains is to be found in isolated strips and remnant patches making classification

difficult (Mucina *et al*., 2006). The Indian Ocean Coastal Belt is now considered to be a biome based on vegetation structure and climatic characteristics that are markedly different from the Savanna biome. The current Indian Ocean Coastal Belt structure was established between after the Last Glacial between 15 000 and 7000 BP and is considered to be the youngest biome in South Africa (Mucina *et al.*, 2006). It has been divided into a series of vegetation units and the Sibudu region falls into the KwaZulu-Natal Coastal Belt (Mucina *et al.*, 2006). The area is also referred to by other researchers as Coastal Forest and Thornveld (Acocks, 1988), Moist Coastal Forest Thorn and Palm Veld (Camp, 1999) and Coastal Bushveld-Grassland (Low and Rebelo, 1998), in other words, a mix of coastal and interior ecozones. Coastal forest, described by a variety of researchers (Lubke and McKenzie, 1998; Moll and White, 1978), predominantly comprises evergreen species and it develops between 20m to 450m above mean sea-level.

The northern portion of the Indian Ocean Coastal Belt has marginally seasonal rainfall with the majority falling during the hot and humid summer months, but due to the influence of frontal systems from the southern regions, 30-40% of rainfall can be received during the mild, frost-free winter months. The mean annual precipitation for the area is 990mm with a range between 820-1270mm (Schulze, 1997). Mean annual temperatures range from 22-25˚C in summer and 17-20˚C in winter. The area experiences high relative humidity (60-70%) and high rates of evaporation (1500-1800mm per year). The moist summer heat assists in the maintenance of the temperature sensitive and moisture dependent tropical vegetation. The prevailing climatic conditions are influenced by the southward shift of the Intertropical Convergence Zone during the summer months and the ameliorating influence of the warm Agulhas Current flowing along the east coast (Mucina *et al.*, 2006). The summary climatic data for KwaZulu-Natal Coastal Belt are derived from instrumental records (1927 to 2005) held by the South African Weather Service and the Water Research Commission.

The cliff face hosting the shelter is identified as Natal Group Sandstone, about 490 Ma in age (Pickering, 2006). The thick layer of sediments forming the sandstones is derived from the erosion of predominantly granitic rocks of the Natal Metamorphic Province (Uken, 1999). The south-western section of the shelter is more exposed and

has been subject to more weathering than the south-eastern portion (Pickering, 2006). The shelter was created by the lowering of the Tongati River channel that down cut into the cliff face during a marine regression in Marine Isotope Stage (MIS) 6 (Maud, 2000; Pickering, 2006; Wadley and Jacobs, 2006). The upper northern floor of the shelter is composed of fine brown sediments and angular fragments of eroded sandstone. There are also numerous large blocks of sandstone in the north-eastern corner from the collapse of sections of the shelter roof. On the lower southern portion of the shelter, the sandstone floor is exposed as a series of ledges or steps (Pickering, 2006). An intrusive dolerite dyke is exposed in the cliff face cross cutting the sandstone approximately 15m upstream from the entrance to the site.

3.2. Excavation history, stratigraphy and cultural sequence

The site was first excavated by Dr. Aron Mazel in 1983 (unpublished notes in the Natal Museum; Wadley, 2001a). A small trial trench of approximately one meter deep was initially excavated. This area has subsequently been incorporated into the current excavation area (Wadley and Jacobs, 2006). The present excavations and research programs were started in 1998 (Wadley, 2001a, b) by Prof. Lyn Wadley and a team from the University of the Witwatersrand and are still ongoing.

More than 21m^2 has been excavated. The site is excavated by following the natural stratigraphy (Fig. 3.3, Vol.II, pg.7). Each square metre is assigned a location (e.g. A5, C6). The square is further divided into four 50cm quadrants (a-d) and material from each is curated separately. The a-quadrant is always the northern and eastern facing corner of each square. Initially the deposit was sieved through 2mm mesh, but from 2003 an additional 1mm mesh was employed to ensure the capture of micromammalian remains and small seeds (Wadley and Jacobs, 2004, 2006; Wadley 2006). The layers have been given specific names that are related to the character and colour of the predominant matrix (Wadley and Jacobs, 2006). The names have been given abbreviated letter designations (Table 3.1, Vol.II, pg.49). The rationale for using such unconventional nomenclature for the various layers is to assist excavators to recognise and remember them.

The designations for the specific layers sampled for this project are as follows:

a) Two layers, Grey Sand (GS) and Grey Rocky (GR), yielded samples from the Howiesons Poort.
b) A single layer, Spotty Camel (SPCA), yielded samples from the post-Howiesons Poort.
c) Two layers, Red Speckled (RSp) and Orange Mottled Deposit (OMOD), yielded samples from the late MSA.

The MSA layers from Sibudu have yielded a long sequence of time-related informally and formally named lithic assemblages (Fig. 3.4, Vol.II, pg.8). These include, from oldest to most recent, a pre-Still Bay MSA, Still Bay, Howiesons Poort, post-Howiesons Poort, a late MSA and a final MSA. The manufacturers of the different toolkits used a range of rocks including locally available dolerite, hornfels, quartz and quartzite. The pre-Still Bay assemblage (basal layer BS) is small and although not yet fully analysed, appears to differ significantly from the Still Bay material (Wadley and Jacobs, 2006). The Still Bay Industry (layers RGS and RGS2) is characterised by bifacial lanceolate points (Wadley, 2007). Residue analyses on the Still Bay points have shown that some were likely to have been butchery knives while others were utilised as spearheads (Lombard, 2006a). Above the Still Bay, layers PGS, GS2, GS, GR2 and GR produced a Howiesons Poort assemblage, characterised by backed segments. Some of these were probably arrow components (Wadley and Mohapi, 2008) and bone points (that were arrows) are also present (Backwell *et al*., 2008). The assemblage has been partly analysed (Delagnes *et al*., 2006; Wadley and Mohapi, 2008; Wadley, 2008). Use-wear and residue analyses confirm that segments were hafted as various parts of weapons (Lombard, 2006b; Pargeter, 2007). The post-Howiesons Poort, final and late MSA are informal assemblage names, based on the available ages and lithic characteristics and as such these names are not accorded formal industrial status (Wadley and Jacobs, 2006). These assemblages have been analysed and described by a number of researchers. The post-Howiesons Poort assemblage indicates no substantial change in typology or technology, but there is a shift in the type of rocks used (Cochrane, 2006; Villa and Lenoir, 2006). The late MSA layers have produced numerous unifacial points that, based on detailed analyses, are considered to have been utilised as spear points (Villa *et al.*, 2005; Villa and Lenoir, 2006). The final MSA assemblage has only been recovered from the eastern portion of the excavation and it contains examples of hollow-base points,

bifaces and wide segments (Wadley, 2005; Wadley and Jacobs, 2006). Microscopic analyses of residues from selected retouched and unretouched artefacts from the site have provided some understanding of the hafting and hunting technologies employed by the inhabitants of Sibudu Cave, for example, triangular points were used as spearheads (Lombard, 2004, 2005, 2006a, b; Villa *et al*., 2005; Williamson, 2004, 2005).

3.3. Dating of Sibudu Cave

Prior to the use of optically stimulated luminescence dating techniques a number of radiocarbon ages were obtained for Sibudu Cave. The Iron Age layer was dated to 960 ± 25 BP (Pta-8015) (Wadley and Jacobs, 2004). Two radiocarbon (^{14}C) dates on charcoal from the MSA layers were obtained by Mazel during his excavations at Sibudu. The first yielded an age of 26 000 ± 420 years BP (Pta-3765) and the second an age of 24 200 ± 290 years BP (Pta-3767). The samples that produced the ages have been considered as being out of context (Wadley and Jacobs, 2004). A further six radiocarbon dates (Table 3.2, Vol.II, pg.51) were obtained from the post-HP, late MSA and final MSA deposits from five samples of charcoal and one of bone. However these dates are considered as unreliable as these MSA deposits fall beyond or near to conventional and internationally accepted limits for radiocarbon dating (Reimer *et al*., 2004; Van der Plicht *et al*., 2004; Vogel and Beaumont, 1972).

Within the MSA deposits are numerous quartz grains that are suitable for optically stimulated luminescence (OSL) dating. A chronological sequence has been created for the MSA layers using optically stimulated luminescence techniques on single grain and single aliquot samples (Wadley and Jacobs, 2004, 2006; Jacobs, 2004; Jacobs *et al*., 2008a). Fourteen sediment samples were taken from deposits above the Howiesons Poort layers and these are presented in Table 3.2 (Vol.II, pg. 51). On the basis of these 14 dates, the post-Howiesons Poort MSA layers can be divided into three broad age clusters, ~58ka (post-Howiesons Poort), ~48ka (late MSA) and ~35ka (final MSA). Seven OSL dates (Table 3.2, Vol.II, pg. 51) for the Howiesons Poort, Still Bay and Pre-Still Bay industries were derived from additional sediment samples (Jacobs *et al*., 2008b; Jacobs and Roberts, 2008). Based on these results, the Howiesons Poort occupation falls between 65ka-62ka, the Still Bay dates to 70ka and

the Pre-Still Bay to between 75ka-72ka (Jacobs *et al*., 2008b; Jacobs and Roberts, 2008). On the basis of the consistency of the optically stimulated luminescence dates, the radiocarbon dates have been rejected as minimum ages.

3.4. Palaeoenvironmental research at Sibudu Cave

In addition to the detailed record of behavioural adaptations through time, Sibudu provides proxy environmental evidence (Fig. 3.4, Vol.II, pg.8). The sediments and faunal and botanical assemblages from Sibudu have been extensively analysed by a range of researchers to produce a detailed record of changes in the local environment. A brief introduction into the various sampling and analytical methods used at the site and summary of the environmental changes suggested by the different proxies is provided. As the focus of the project is on charcoal, a more detailed discussion on charcoal analysis was presented in Chapter 2.

3.4.1. Faunal data

The faunal assemblages from the MSA have been extensively analysed and the data serve as a proxy for the environment around the site, as well as a record of shifts in the procurement and utilisation of terrestrial, marine and freshwater resources (Cain, 2004, 2005, 2006; Clark and Plug, 2008; Plug, 2004, 2006; Plug and Clark, 2008; Reynolds, 2006; Wadley *et al*., 2008; Wells, 2006). A wide variety of species are represented. Changes in species composition and abundance between the MSA divisions >60ka to 48ka provide an indication of changes in the environment in the vicinity of the site through time. Preliminary taphonomic analyses have been carried out on both identifiable and unidentifiable material to determine a number of relevant characteristics. These include understanding prehistoric processing methods, the manner of acquisition, effects of non-human accumulating agents and indications of post-depositional alteration (Cain, 2005, 2006; Clark and Plug, 2008).

On the basis of these early results it appears that both before and after ~60ka, humans were the primary accumulating agent of the fauna (Cain, 2006; Clark and Plug, 2008; Plug, 2004; Wells, 2006). Micromammalian material was also identified (Glenny, 2006). Certain species are strongly habitat specific and the occurrence of different

species combinations can be used to identify different ecozones within an area. Fluctuations in the species composition from a site over time can be used to track changes in the composition of local vegetation communities. Eleven key species (rodents, shrews and bats) were identified in the Sibudu deposits (Glenny, 2006).

Faunal evidence also lends support to a heavily forested environment and a perennially flowing river during the Howiesons Poort occupation. Crocodile (*Crocodylus niloticus*), water mongoose (*Atilax paludinosus*), water monitor (*Varanus niloticus*), frog and fresh water mollusc species and a variety of waterfowl species attest to a permanent water source (Plug, 2006; Plug and Clark, 2008). The Howiesons Poort faunal assemblage is dominated (91.4%) by mammal species (such as blue duiker (*Cephalophus monticola*), bush pig (*Potamochoerus porcus*) and vervet monkey (*Chlorocebus aethiops*)) that favour closed or semi-closed habitats (Plug, 2004; Clark and Plug, 2008). A small proportion of the species identified, such as African buffalo (*Syncerus caffer*), blue wildebeest (*Connochaetes taurinus*) and roan antelope (*Hippotragus equinus*), from the Howiesons Poort layers are characteristic of predominantly open environments. This suggests that, in addition to exploiting forests, the inhabitants of Sibudu Cave were also accessing open savanna or woodland areas within the vicinity of the site. Further evidence for humid, moist forested environments is provided by micro-mammalian species, such as the Gambian giant rat (*Cricetomys gambianus*) and Geoffroy's horseshoe bat (*Rhinolephus clivosus*) (Glenny, 2006). Both species require humid conditions and the Gambian giant rat occurs exclusively in evergreen forests and woodlands (Skinner and Chimimba, 2005).

The faunal assemblage from the ~58ka layers shows marked differences in species composition from that of the Howiesons Poort layers. Small bovid species decrease and much larger species dominate the assemblage. These include equids, giraffe (*Giraffa cameleopardalis*) and other large to very large bovids, such as buffalo (*S. caffer*) and eland (*Tragelaphus oryx*) (Cain, 2006; Plug, 2004, Wadley *et al*. 2008). Duiker species (blue duiker (*C. monticola*) and red duiker (*Cephalopus natalensis*)) that tend to inhabit closed habitats such as thick forest/woodland persist in small frequencies and support the interpretation of the continual presence of a riverine forest/woodland community in the area. Grazers such as equids and alcelaphines

appear throughout the ~58ka sequence, but are present in higher frequencies in the upper layers indicating a shift to more open grassland environment (Cain, 2006; Clark and Plug, 2008; Plug, 2004). Such a marked increase in grazing species points to a major change in environmental conditions from closed forest/woodland communities to drier more open savanna/grassland communities (Clark and Plug, 2008; Wadley *et al*, 2008). Further evidence for a substantial shift in vegetation patterns is the presence of the Natal multimammate mouse (*Mastomys natalensis*) in layer MY (Glenny, 2006). This is a pioneer species that establishes itself in areas that have undergone a significant environmental change, and although the species has a wide habitat tolerance, it is not a forest dwelling species and has a degree of water independence, allowing it to occupy in dry areas (De Graaff, 1981; Meester *et al*., 1979; Skinner and Smithers, 1990).

The species composition of the ~48ka faunal assemblage provides evidence for a mosaic environment, consisting of a variety of vegetation communities. The RSp layer has produced a diverse species profile with representatives from a number of habitats. Savanna and grassland species such as zebra (*Equus quagga*), giraffe (*G. cameleopardalis*), warthog (*Phacochoerus africanus*), buffalo (*S. caffer*) and blue wildebeest (*C. taurinus*) occur, as well as a number of forest/woodland dwelling species such as bush pig (*P. porcus*), blue and red duiker and bushbuck (*Tragelaphus scriptus*) (Cain, 2006; Plug, 2004; Wells, 2006). At this time there is an increase in the numbers of small browsing species, a change that correlates with the increase the number of woody taxa represented by seeds (Wadley *et al.*, 2008). The Gambian giant rat (*Cricetomys gambianus*) is present again in RSp at ~48ka, indicating the presence of forested areas and rainfall over 800mm per annum. Another ecological succession event, prior to or during the formation of RSp, may be suggested by the occurrence of the Natal multimammate mouse (*Mastomys natalensis*), the pioneer micromammal species (De Graaff, 1981), previously seen in the ~58ka layers (Glenny, 2006).

3.4.2. Seed assemblage data

A range of carbonized seeds, nuts and fruit stones have been recovered from the MSA layers (Sievers, 2006; Wadley, 2004). The occurrence of these materials may be the

result of accidental carbonisation when hearths were built over already buried seeds, nuts and fruit stones (Sievers and Wadley, 2008). The analysis of these collections has allowed the identification of vegetation communities in the vicinity of the site and provided a proxy record of climatic changes indicated by variation in the community structure. Care needs to be taken to avoid interpretational bias due to the agents of accumulation, site formation processes, differential preservation, sample recovery and identification. Sixty-five taxa are represented, of which 35 were identified to family, genus or species level using modern comparative collections and a series of key attributes (Wadley, 2004). The remaining 31 taxa have been described as types until identification is possible (Sievers, 2006). The taxa include sedges, grasses, woody climbers, trees and shrubs. To identify temporal shifts in vegetation communities, Sievers (2006) examined the presence/absence of key marker species, changes in species abundance and variability in the co-occurrence of certain species.

Macrobotanical evidence (seeds) from the pre-60ka layers indicates a predominantly forested environment with patches of woodland. The environment is considered as woodland when woody species provide up to 75% canopy cover, whilst forests comprise predominantly of evergreen tree species with a continuous canopy, a variety of climbing species and epiphytes and low growing shrubs and ferns on the forest floor (Rutherford and Westfall, 1986). The occurrence of Cyperaceae (sedges) seeds in these layers suggests the presence of water throughout the year, because this family is generally heavily dependent on water. The dominant sedge, *Schoenoplectus,* occurring in the MSA layers, requires year-round standing water (Sievers, 2006). Further evidence for a permanent water source is given by skeletal material from hippopotamus (*Hippopotamus amphibious*) (Plug, 2006). A number of evergreen forest species have been identified in the charcoal assemblage from these pre-60ka layers. These include *Podocarpus* (Yellowwood), *Buxus* and *Sapium/Spirostachys* (Allott, 2005, 2006). *Podocarpus* appears to be the dominant forest species at this time. These then disappear from the record, but reappear in the uppermost MSA layers (Sievers, 2006).

Changes in the ratios of evergreen versus deciduous species were calculated for the MSA sequence (Sievers, 2006; Wadley, 2004). The majority of identified seeds from the woody species are evergreen, but there appears to be an increase in the occurrence

of deciduous species in the upper layers. The botanical evidence points to an increase in deciduous species after 60ka, accompanied by a greater variety of seed types (Sievers, 2006). This, however, may be the result of larger sample sizes due to more extensive excavations (Wadley, 2006). The frequencies of deciduous species increase noticeably through the ~48ka sequence even though evergreen species persist.

3.4.3. Charcoal data

There is abundant charcoal within the MSA layers of Sibudu Cave, indicative of extensive burning events. Charcoal has been located in discrete hearths, as well as scattered across layers. All the charcoal at the site seems to have been the result of deliberate burning of wood by the site inhabitants. Scattered charcoal outside of hearths is probably the result of hearth maintenance when ash and charcoal was scraped from disused hearths in antiquity (Cain, 2005). A comparison of the mineralogical and sedimentological characteristics and burnt bone from both discrete hearths and ashy deposits show that the ashy deposits are very similar to that within the hearths (Cain, 2005; Schiegl *et al.*, 2004).

Allott (2004, 2005, 2006) has identified temporal changes in vegetation and wood-use at the site from woody taxa identified in the charcoal assemblage. During excavation large, friable fragments of charcoal are collected by hand to prevent damage during sieving. The largest and best preserved fragments were recovered from the post-60ka MSA layers. Charcoal from the older layers is more friable, less abundant and smaller, likely as a result of post-depositional diagenesis (Allott, 2004). All other pieces were collected and curated from sieved material during sorting either on-site or at the laboratory. Only charcoal from the initial trial trench, composed of squares B5 and B6, was analysed. This was done as the trial trench provided the longest time sequence of the excavation at the time of analysis. To remove any additional bias to sampling, charcoal was exclusively taken from the sediment matrix and not from discrete hearths. The area of all seven layers (post-60ka) sampled by Allott was $2m^2$, with the exception of the Howiesons Poort layers, where only $1m^2$ had been excavated at the time of analysis. One-hundred and twenty fragments of charcoal were taken from each layer in five size categories. Identification of the charcoal was done using

internationally recognised methods involving a number of comparative wood/charcoal collections and a series of anatomical characteristics. Charcoal was identified as far as possible, usually to family or genus, by these methods. All charcoal samples were split into three, providing a cross-section, transverse section and a radial section. These were then viewed either through an incident light microscope or scanning electron microscope and key anatomical features recorded.

Woody taxa identified from the charcoal assemblage indicate a continuous presence of an evergreen riverine forest component. This riverine community is maintained throughout the MSA sequence due to the perennial Tongati River providing moisture in all the periods that were occupied by people (Allott, 2004, 2005, 2006). As previously indicated, there are a number of hiatuses that may represent dry periods between occupations (Wadley and Jacobs, 2006) and thus there are no records of what environmental conditions may have been like during these times. The charcoal assemblage from the middle ~58ka layers does not contain *Podocarpus*, but has *Acacia* spp., *Erica* spp., *Morella* spp. and *Ziziphus* spp. *Podocarpus* reappears in SPCA where *Acacia* species are absent, following the trend for the two genera to be mutually exclusive (Wadley, 2006). *Acacia* and *Erica* species reappear in the BSp layers, but *Podocarpus* does not. These changes in local woody species composition most likely represent changes in environmental conditions due to shifts between wetter and drier periods.

The ~48ka charcoal assemblage also reveals an increase in deciduous woody species. In layer RSp there is an unusual combination of tree species, with *Acacia* and *Podocarpus* occurring together. These two genera are normally mutually exclusive. This combination of species is not seen in any other MSA layer from Sibudu (Allott, 2006; Wadley, 2006). The overall species composition from the ~48ka charcoal assemblage suggests deciduous savanna woodland with taxa such as *Acacia* spp., *Albizia* spp. and *Celtis*. There is no evidence for the presence of *Podocarpus* in the remainder of the charcoal assemblage (Allott, 2006).

3.4.4. Pollen and phytolith data

Fifteen layers were assessed for their pollen content and composition. Pollen retrieved from eight of the layers was identified using microscopy and comparison with reference collections. The pollen data show changes in the local plant communities and the anthropogenic utilisation of plant resources (Renaut and Bamford, 2006).

Pollen and phytolith analyses from a number of ~58ka layers reveal the presence of grasses, sedges, fern spores and *Acacia* species (Renaut and Bamford, 2006; Schiegl and Conard, 2006; Schiegl *et al.*, 2004). The frequency of grass phytoliths increases in the upper layers. This may be due to more frequent use of grasses as tinder for fires, reflecting anthropogenic behaviour rather than environmental change (Schiegl and Conard, 2006; Wadley, 2006), although the fact that more grass was being brought to the site at this time may reflect an increase in the presence of grassland around Sibudu.

Grass pollen is present throughout the MSA sequence, but analysis of the phytolith composition of sediments shows an overall decrease in grass phytoliths and an increase in tree phytoliths at 50ka (Schiegl and Conard, 2006).

3.4.5. Sedimentological data

The Sibudu MSA deposits are finely stratified with complex layering, comprising numerous ash lenses and well-preserved hearths. Contacts between layers are relatively sharp and they undulate in certain areas (Pickering, 2006). Sedimentological analysis indicates that the sediment components, both anthropogenic and natural, show little or no indication of water transport and that the deposits are almost completely due to the presence and activities of the human occupants of the site (Goldberg *et al.*, 2009; Pickering, 2006). The lower layers are lightly bedded, lie flatter than the younger layers and are composed of light brownish-grey soft silts. The upper layers are more complex and are composed of various, coloured ashy layers. Archaeological cave/shelter sediments comprise a mix of biogenic, geological and anthropogenic components that link the history of a site to changes in the local environment (Schiegl and Conard, 2006). A detailed analysis of more than 525

sediment samples from most of the Sibudu stratigraphic layers and 20 samples from hearth/ash deposits (Schiegl *et al*., 2004; Schiegl and Conard, 2006) were subjected to Fourier Transform infrared spectroscopy, transmitted polarised light microscopy and scanning electron microscopy coupled with X-ray analysis. These techniques provide data on the mineralogical and phytolith composition of the deposits, which reflect changes in local environmental conditions.

Mineralogical evidence in the form of gypsum and calcite crystals within the deposits provides an indication of changing humidity levels through time (Pickering, 2006; Schiegl and Conard, 2006; Schiegl *et al*., 2004). Both materials require water to form, but calcite is less soluble than gypsum (Goldberg and McPhail, 2006; Schiegl and Conard, 2006) and will therefore persist during periods of higher moisture availability. During the >60ka period (Pre-Still Bay, Still Bay and Howiesons Poort), there is little or no gypsum present, but calcite crystals are prevalent suggesting higher than usual humidity during this time. In the ~58ka layers (post-Howiesons Poort) there are numerous gypsum nodules present in the deposits. Gypsum accumulations such as those in the ~58ka layers are considered as a useful indicator of past arid conditions (Goldberg and McPhail, 2006). The formation of such nodules would have required a brief phase of humidity, but due to their solubility in water, for the nodules to persist, conditions would have to have become more arid. There are both calcite and gypsum crystals present in the ~48ka layers (late MSA) and this suggests relatively dry conditions.

3.4.6. Magnetic susceptibility data

Further palaeoclimatic data were obtained from magnetic susceptibility at ~58ka, 48ka, 35ka and AD 1100 (Herries, 2006). Sediment samples were collected from the southern, northern and eastern stratigraphic faces. The magnetic susceptibility analyses of the Sibudu sediments provide a record of oscillating climatic conditions driven by changes in temperature and moisture levels (Herries, 2006). Since the deposits are largely due to anthropogenic activities within the site (Goldberg *et al*., 2009; Pickering, 2006); the magnetic susceptibility data should be interpreted with

care. It appears that the lower magnetic susceptibility readings correlate with periods of reduced human activity rather than a change in the environment.

The coldest period in the ~58ka layers occurred in the earliest layers, YA2 to G1, during the transition between MIS 4 and MIS 3. In more recent ~58ka layers, magnetic susceptibility results show alternating cooler and warmer conditions, although overall temperatures were lower than present (Herries, 2006).The upper layers (OMOD-MOD) of the ~48ka sequence still indicate a warming trend, but magnetic susceptibility readings do suggest an initial cooling period in OMOD (Herries, 2006).

3.5. Conclusions

Taken together, the available evidence from the Howiesons Poort layers shows that at the end of MIS 4, the environment around Sibudu Cave was cooler than at present, and conditions were also humid. A substantial evergreen forest was supported although areas of slightly more open woodland communities were also present. The post-Howiesons Poort (~58ka) layers provide evidence for oscillating climatic conditions from the coldest period at the end of MIS 4 into MIS 3. There is an overall warming trend with a number of cooler phases. These oscillations have resulted in a number of ecological successions in the Sibudu region, where substantial changes in the composition of the various plant and animal communities took place. The forested environment before 60ka was drastically reduced and open woodland and grassland communities developed in addition to forested areas. Due to the continual presence of water in the Tongati River, evergreen riverine forests may have persisted through the periods when MSA occupation occurred at Sibudu. There is no available environmental evidence for periods when the site was not occupied. This suggestion is supported by the almost continuous occurrence of Cyperaceae seeds and pollen, the persistence of a number of evergreen riverine species identified in the charcoal assemblage, as well as forest-dwelling species such as the blue duiker in the faunal material (Allott, 2005, 2006; Cain, 2006; Plug, 2004; Renaut and Bamford, 2006; Sievers, 2006; Wadley, 2006). At the end of MIS 4 and the start of MIS 3 colder than present conditions resulted in an increase in open grasslands, indicated by higher frequencies of grazing species and grass phytoliths. During the warmer phases of

MIS 3, the grasslands decreased in size and woodland savanna predominated. Based on the recent OSL dates there is a hiatus of 9.8 ± 1.3ka between the post-Howiesons Poort and late MSA occupations (Jacobs *et al*., 2008a). This hiatus coincides with a period of colluviation between 56ka-52ka, an indication of more arid conditions or transitional climates and reduced vegetation cover, recorded from a series of well-dated stratigraphic sequences from erosion gullies in KwaZulu-Natal (Botha, 1996; Botha and Partridge, 2000; Botha *et al*., 1992; Clarke *et al*., 2003; Wintle *et al*., 1995). A second hiatus of 12.6 ± 2.1ka occurs between the late and final MSA occupations (Jacobs *et al*., 2008a). This hiatus also coincides with a period of colluviation during the period 47ka-41ka, recorded in KwaZulu-Natal erosion gullies (Botha, 1996; Botha and Partridge, 2000; Botha *et al*., 1992; Clarke *et al*., 2003; Wintle *et al*., 1995). Environmental conditions were likely too harsh to permit the use of the shelter as a permanent dwelling. The proxy environmental data from the late MSA layers show an initial period of warming followed by a cooler period. The combined seed, phytolith and charcoal data from the YSp and RSp layers may indicate an overall increase in deciduous woodland within close proximity to the site. Further environmental data are available for the final MSA (~35ka) layers, but have not been considered for this study, as no suitable charcoal samples were available for isotopic analysis.

CHAPTER 4. RAINFALL CONTROL OF THE $\delta^{13}C$ RATIOS OF *MIMUSOPS CAFFRA* FROM KWAZULU-NATAL, SOUTH AFRICA

Paper 1: Rainfall control of the $\delta^{13}C$ ratios of *Mimusops caffra* from KwaZulu-Natal, South Africa

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Background to the paper

In this paper we present isotopic data obtained from two specimens of *Mimusops caffra*, a subtropical forest species that occurs specifically in Dune Forest on sandy soils along the northern coast of KwaZulu-Natal. In order to understand and interpret potential environmental responses from the archaeological charcoal it is necessary to first understand how and what ecophysiological signals are recorded in modern tree species from the region and develop a set of modern analogue data.

Growth ring sequences were obtained for each tree. Due to the manner by which rings are laid down in subtropical/tropical species it was necessary to determine if growth rings are laid down on an annual basis as they often have missing or additional growth rings. This was achieved by high precision radiocarbon dating techniques of three samples from the largest trunk section. The radiocarbon dates obtained showed that the growth rings were laid down on an annual basis and indicated the need for care when counting rings as growth rings are laid down in a lobate fashion and it may be possible to obtain incorrect ring counts.

The $\delta^{13}C$ values of wood samples pre-treated in two ways were compared to determine the most time-efficient method that yielded a meaningful climatic signal and this would be used for the duration of the project. Wholewood samples, pre-treated by Soxhlet distillation, were compared to wood samples from the same growth rings that were further processed to yield α-cellulose. The Soxhlet distillation was a two-step process, first utilising a 2:1 toluene-ethanol azeotropic mixture and then treated with 100% ethanol. The comparison of wholewood and α-cellulose $\delta^{13}C$

values showed a significant correlation ($r^2 = 0.75$, $P > 0.0001$, correlation = 0.85) and both time series preserved the same record of environmental responses, although wholewood values were depleted on average by 1.2‰ with respect to α-cellulose $\delta^{13}C$ values. On the basis of these results and data from recent literature, it was decided to use wholewood samples, a more time efficient method given the number of samples that were necessary for this study.

The $\delta^{13}C$ values for three specimens were compared with a local climate data set (rainfall, humidity and temperature) to determine which climatic factor had the most influence on $\delta^{13}C$ values creating a temporal record of the trees response to local environmental climatic conditions. This was carried out using signal processing techniques. The results show that *M. caffra* is adapted for an environment with high moisture availability. There is also evidence for an isotope juvenile effect during the early stages of growth. In the more recent growth rings, dating from 1950 to 2004, there is evidence for anthropogenic influences on the isotopic composition. Through the use of signal processing techniques (wavelets) it was possible to account for these influences and separate the inter-annual climatic response of the trees. The carbon isotope time series from *M. caffra* shows a strong inter-annual response to rainfall variability and allows the reconstruction of rainfall patterns beyond the instrumental record of the region.

Rainfall control of the $\delta^{13}C$ ratios of *Mimusops caffra* from KwaZulu-Natal, South Africa

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4.1. Abstract

High precision radiocarbon dating of individual growth rings demonstrates that *Mimusops caffra* produces annual growth rings. Direct comparison of the carbon isotope composition of the annual rings from two specimens from the northern coast of KwaZulu-Natal, South Africa, with the historic climatic record that exists from 1927, shows a response to the amount of rainfall recorded for each year. The $\delta^{13}C$ response is particularly sensitive to extreme events such as tropical cyclones and droughts. It is demonstrated that wholewood samples provide an accurate record of climatic conditions. Although the result allows long-term periodicity in rainfall to be determined over the 134 years during which the oldest tree grew, the last 20 years is overprinted with a $\delta^{13}C$ shift of anthropogenic origin. Signal processing approaches allow the inter-annual rainfall response to be separated from the long-term anthropogenic influences.

Keywords: Stable carbon isotopes, spectral analysis, wavelets, tropical cyclones, drought, environmental record

4.2. Introduction

Scenarios for climate change rely on circulation models that approximate the dynamics of the earth system. An important test of the accuracy of model forecasts is the skill with which the known trajectory of climate can be constructed. Unfortunately historic records of climate variability are too short in southern Africa to discern longer, multi-decadal, cycles of variability. Some highly resolved palaeo-records do exist (Holmgren *et al*., 2003; Scott, 1996; 1999; Scott and Woodborne, 2007a; 2007b;

Talma and Vogel, 1992) from southern Africa, but most are either too widely scattered to reconstruct spatial variability or too poorly resolved in the time domain to make synchronous inter-regional comparisons. Tree rings hold potential proxy evidence for geographical variations in past climates (McCarroll and Loader, 2004), but the dynamics of tropical and sub-tropical regions in southern Africa have not been as thoroughly studied as those of Northern Hemisphere temperate regions (e.g. Briffa *et al*., 2004, Chevillat *et al*., 2005; Ghosh and Brand, 2003; Kadonga *et al*., 1999; Leavitt, 2002; Martinelli, 2004; Van Bergen and Poole, 2000). One of the concerns is that accurate dating is required and low-latitude tree species do not have the distinctive growing season that leads to the formation of annual growth rings. However other seasonal factors, such as the occurrence of rainfall in southern Africa, provide a potential mechanism that might lead to the formation of annual rings. An initial assessment of 108 indigenous South African trees identified *Podocarpus* and *Widdringtonia* species as having the potential to provide climate proxies (Lilly, 1977). However, even for these species many factors will affect the formation of rings. Any study of environmental proxies from tree rings is challenged first to demonstrating that rings are annual, and second to demonstrate that the proxy measurement from the rings is environmentally meaningful.

Stable carbon isotope chronologies derived from tree ring sequences are an established means to reconstruct past environments. The fractionation of carbon isotopes during photosynthesis is influenced by environmental and climatic factors such as insolation, temperature, humidity and precipitation (Ehleringer *et al*., 1986; Farquhar *et al*., 1989; Francey and Farquhar, 1982; Leavitt and Long, 1991). The influence of these factors are recorded in the $\delta^{13}C$ values of tree rings and the variation seen in these values may serve as a climate proxy (e.g. Kadonga *et al*., 1999; Leavitt, 2002; Leuenberger *et al*., 1998; McCarroll and Loader, 2004). Where the environmental driver of the isotope values can be determined, the constraint is in the time dimension. The approach has only been applied to a few selected species that demonstrate annual growth patterns.

A 400-year dendrological sequence using *Widdringtonia cedarbergensis* from the southern Cape linked rainfall patterns to annual ring widths (Dunwiddie and LaMarche, 1980), and stable carbon isotopes in annual tree rings from a 100-year old

W. cedarbergensis (February and Stock, 1999) showed a decreasing trend in $\delta^{13}C$ values due to depleted atmospheric carbon isotope composition. These studies are at odds with one another as no clear connection was demonstrated between isotope data and climatic factors (e.g. rainfall). Several tree ring studies have been carried out in southern Africa. Stable carbon isotope ratios from fresh wood, cellulose and carbonised wood samples of *Combretum apiculatum* and *Protea roupelliae* from a climatic gradient from the summer rainfall region of South Africa were compared (February and Van der Merwe, 1992; February, 1997). *C. apiculatum* showed a significant correlation between $\delta^{13}C$ values and rainfall, whilst *P. roupelliae* did not. The water stress history of a drought- adapted *Acacia erioloba* trees from southern Namibia and the Northern Cape was elucidated through stable carbon isotope analysis (Robertson and Woodborne, 2001; Woodborne *et al*., 2003). The $\delta^{13}C$ data appeared to correlate with groundwater levels, and not exclusively with rainfall patterns (Robertson and Woodborne, 2001). Variable results were obtained from isotopic analyses of selected pine tree species. *Pinus patula* $\delta^{13}C$ variations correspond to local rainfall patterns but those of *Pinus elliotii* do not (Woodborne *et al*., 2003). The $\delta^{13}C$ composition and wood anatomy of two specimens of *Breonadia salicina* growing in the Limpopo Province reflected regional palaeoclimatic changes in the ring sequences over a 600-year period (Norström, 2005). Annual rainfall patterns of the last century were reflected in the isotopic and anatomical data of the most recent rings. Data from older rings showed that the trees were also responding to drier and wetter periods between the mid-1500s and the 1800s.

In this paper, we present $\delta^{13}C$ results from growth ring sequences of two specimens of *Mimusops caffra* from northern Kwazulu-Natal. The $\delta^{13}C$ data demonstrates the potential of *M. caffra* to provide evidence of climatic variability in KwaZulu-Natal.

4.3. Stable carbon isotope composition in trees

During carbon fixation lighter $^{12}CO_2$ molecules are more readily assimilated by the photosynthetic enzyme (RuBisCO) than heavier $^{13}CO_2$ molecules (Farquhar *et al.*, 1988). This creates both a CO_2 diffusion gradient within the stomata as well as an isotopic gradient. The $^{12}CO_2$ molecules diffuse faster through the intercellular space (stomates) than the heavier $^{13}CO_2$ molecules. This simplified mechanism underlies a

dynamic fractionation response to external environmental conditions. Trees maintain the water potential in their leaves by regulating the transpiration through the stomates (Farquhar *et al.*, 1982). When soil moisture levels are low the stomates close and photosynthetic pathways make more use of the CO_2 trapped within the leaves. This results in increased use of ^{13}C for carbohydrate synthesis during periods of water stress. Fractionation is relative to the available atmospheric CO_2 isotopic ratio, and can be expressed as:

$$\delta^{13}C_{plant} = \delta^{13}C_{atmosphere} - a - (b - a)\frac{ci}{ca} \qquad \text{(Farquhar } et\ al.\text{, 1982)}$$

The current value for $\delta^{13}C_{atmosphere}$ is considered to be -8‰, based on the use of VPDB standard (Coplen, 1995; McCarroll and Loader, 2004), and the value for *a* (4‰) represents the discrimination against $^{13}CO_2$ during diffusion via the stomata and *b* (27‰) is the net discrimination due to carboxylation. The values c_i and c_a represent the intercellular and ambient CO_2 concentrations respectively. Total fractionation is controlled by the relationship between c_i and c_a which varies in response to stomatal closure (Farquhar *et al.*, 1982).

It should be noted that stomatal response is not the only water regulatory mechanism of trees, and also a stomatal response may not always affect CO_2 exchange. Short term changes such as atmospheric relative humidity and soil moisture fluctuations may affect CO_2 exchange between the leaf and atmosphere. Longer term changes include anatomical changes (and hence the number of stomata) and rainfall variability may also result in complications during CO_2 exchange. The burning of fossil fuels has also led to a progressive decrease in the $\delta^{13}C$ values of the atmosphere and has consequently affected the $\delta^{13}C$ ratios of trees (Dongarra and Varrica, 2002; Leavitt and Lara, 1994; February and Stock, 1999). Localised combinations of anthropogenically altered and natural environmental factors such as air pollution, canopy cover and soil salinity have an influence on tree $\delta^{13}C$ composition (Farquhar *et al.*, 1989; Livingstone *et al.*, 1999; West *et al.*, 2001). It is necessary to establish if the $\delta^{13}C$ values of a tree species are controlled by environmental factors and not by the other complicating adaptive responses. “Calibration” against known climatic indicators is required, and while this may appear circular, the approach allows the

extension of the climatic reconstruction back beyond the time-frame of the calibration dataset to a time frame that is limited by the age of the tree species.

Stable carbon isotope research on woody tree material initially used wholewood samples (Craig, 1954; Libby *et al*., 1976), but subsequently it became clear that various components of wood differ isotopically (Wilson and Grinstead, 1977) and studies began to focus on cellulose (Barbour *et al*., 2001; Leuenberger *et al*., 1998). A number of methodological concerns were addressed by using cellulose. Cellulose cannot be mobilised by the tree and so it can be linked to a specific growth period. In addition, there are reduced problems of inter-annual variation and lignin ratios. Greater sample homogeneity can be obtained during cellulose purification.

The processing of α-cellulose and lignin is a lengthy process and the use of wholewood tissue over α-cellulose has been considered (Leuenberger *et al*., 1998; Schleser *et al*., 1999; Barbour *et al*., 2001; McCarroll and Loader, 2004). Research on the $\delta^{13}C$ values of lignin, cellulose and wholewood from oak tree rings showed that the $\delta^{13}C$ values for cellulose and lignin are offset by about 3‰ (Loader *et al*., 2003). There was no temporal offset shown in the $\delta^{13}C$ values, with all three materials displaying similar trends over time. When $\delta^{13}C$ values of the three tissue types were correlated with a number of climatic variables (temperature, precipitation and relative humidity), the strongest correlations detected were from wholewood data, followed by cellulose and lastly lignin. This suggests that wholewood has the potential to provide a suitable proxy of climate (Cullen and Grierson, 2006; Loader *et al*., 2003; McCarroll and Loader, 2004). Additional studies done comparing α-cellulose and wholewood isotope values have shown that both record the same climatic information and no climatic signals were lost when using wholewood over α-cellulose (Barbour *et al*., 2001; Warren *et al*., 2001). If information on specific climatic events, such as significant droughts or floods is required, it may be pertinent to utilise α-cellulose as this tissue type may record short term events more accurately (Cullen and Grierson, 2006). All these factors are important to consider especially when comparing data sets (Loader *et al*., 2003; McCarroll and Loader, 2004; Rinne *et al*., 2005). These effects are statistically more pronounced when sample sets are relatively small (West *et al*., 2001).

4.4. Materials and methods

Mimusops caffra (E. May ex A.DC) is a member of the Sapotaceae that occurs as a small to medium tree or shrub, depending on the location in which it grows. It is found in the tropics and subtropics at low altitudes and is restricted to the eastern coast of southern Africa from Mozambique, along the KwaZulu-Natal coastline, through the Transkei and into the Eastern Cape. *M. caffra* typically occurs in Dune Forest (Lubke *et al*., 1997; Lubke and McKenzie, 1998), growing above the high tide mark (Coates-Palgrave, 2003; Palmer and Pitman, 1972; Pooley, 2003; Van Wyk and Van Wyk, 1997).

A trunk disc was collected in February 2005 from a site in Umhlanga Rocks along the northern coast of KwaZulu-Natal (Fig. 4.1, Vol.II, pg.9). The disc (Umhlanga 1) was obtained from a large *M. caffra* felled as part of an urban development project and was located approximately 700 meters from the ocean. A second trunk disc (Admiralty Reserve 1) was collected from the Admiralty Marine Reserve approximately five kilometres south of Umhlanga Rocks (Fig. 4.1, Vo.II, pg.9). This tree was located about 50 meters from the ocean in the dune cordon. Although collected in 2005, both specimens had been felled some time in 2004.

The trunk samples were sanded with increasingly finer grades of sandpaper (grades P40µm to P1200µm) until growth ring sequences were clearly visible. The rings were traced around the circumference of each trunk using a binocular microscope and an incident light source. The total number of growth increments was established and each ring numbered from the exterior to the centre of the trunk. A total of 134 rings were counted for Umhlanga 1 and 56 rings for Admiralty Reserve 1.

4.4.1. High precision radiocarbon dating

Tropical and subtropical trees often have problematic ring sequences where rings may not form on an annual basis (Norström, 2005; Vogel *et al*., 2001; Woodborne *et al*., 2003; Robertson *et al*., 2004). It is therefore necessary to establish if observable rings are representative of annual growth cycles. Wood that was carefully extracted from three separate growth rings from Umhlanga 1, taking care to avoid contamination

from adjacent rings, was prepared for high precision radiocarbon dating. These were from ring 17 and ring 25 assuming they would correlate with the years AD 1987 and AD 1979 respectively. A third sample comprising a composite of the innermost part of the disk (rings 134-124) was also prepared. Samples of 20g to 25g from each of these samples were split into small fragments and treated with dilute acid (2% HCl) and alkali (2% NaOH) to remove any soluble organic materials. They were washed with distilled water, dried, combusted and the carbon dioxide gas collected and purified. The results were then calibrated using the high-resolution atmospheric radiocarbon record for Pretoria (Vogel *et al*., 2002).

4.4.2. Sample preparation and processing for isotopic analysis

The longest radial growth ring sequence was selected to ensure that a maximum number of samples for isotopic analysis could be taken. An offset transect of drillings (2mm diameter) was obtained from the intersection of the cambium and bark and the most recent growth ring to the centre of the trunk. The high resolution sampling strategy allows several samples to be taken from a single ring which highlights the within-ring variability, provided that the rings are greater than 2mm in width. If there was evidence that the tree grew continuously this might allow the testing of seasonal variability in the environmental response of the tree, and even though this seemed promising the dating resolution cannot answer this question. This sampling method has proven to be successful in a number of stable isotope studies of tree ring sequences (Norström, 2005; Woodborne *et al*., 2003).

Due to the large number of samples that were generated, a time efficient processing method of sample pre-treatment, developed by Green (1963) and adapted by Loader *et al*. (1997), was employed. Samples were subject to a toluene/ethanol Soxhlet distillation process to remove mobile constituents, such as resins, oils and waxes. These constituents may not be deposited during the formation of growth rings and the pre-treatment ensures they are not reflected in stable carbon isotope values (Loader *et al*., 1997). This method allows for the rapid batch processing of numerous small wood samples for stable carbon isotope analysis.

An initial test was carried out to determine whether the isotopic value of α-cellulose or wholewood samples would respond to an environmental variable. A comparison was carried out using the most recent 18 growth rings from the Umhlanga 1 trunk. One-hundred and forty-two samples from two adjacent sampling transects were run through a Soxhlet distillation process. Samples from one of the sample transects (n=71) were further processed to α-cellulose using a sodium chlorite/sodium hydroxide bleaching (Loader *et al*., 1997). The results show that α-cellulose produced the same inter-annual variability that is seen in the wholewood (Fig. 4.2, Vol.II, pg.10) with an offset of 1‰. Minor variations in $\delta^{13}C$ values are due to small differences in sample position across each growth ring. This is in line with results seen in studies previously mentioned (Cullen and Grierson, 2006, Loader *et al*., 1997; McCarroll and Loader, 2004). The carbon isotope values of wholewood and α-cellulose showed a significant correlation ($r^2 = 0.73$, correlation coefficient = 0.85, $P < 0.00001$). The rest of the results presented here are all derived from wholewood samples.

The carbon isotope analyses were made on a Thermoquest EA 1110 elemental analyser integrated with a VG Isogas SIRA 24 stable isotope ratio mass spectrometer, housed at the CSIR in Pretoria. Approximately 0.4mg aliquots of wholewood were run in duplicate. If the precision for each sample pair was unacceptable (0.30‰ variation), an additional aliquot was analysed. Two project specific standards, a C_4 grass (*Hyparrhenia hirta*) and a C_3 tree (*Shorea superba*) were used to correct for any sample size effects of the elemental analyser. These particular species were selected because they each represent average carbon isotope values for C_3 and C_4 plants. The average precision for the replicates was <0.12‰. Isotope results are expressed in delta (δ) notation on a per mille (‰) scale relative to the Vienna PeeDee Belemnite (VPDB) standard.

4.5. Results and Discussion

4.5.1. Radiocarbon dates

As the Umhlanga 1 tree was felled in AD 2004, ring 17 should have grown in AD 1987 or AD 1988, and ring 25 should have grown in AD 1979 or AD 1980 depending

on whether the 2004 ring had formed sufficiently at the time of sampling. The radiocarbon analysis of these samples (Table 4.1, Vol.II, pg.53) yielded "bomb carbon" that was calibrated against the recorded atmospheric ^{14}C record from Pretoria, South Africa (Vogel *et al*., 2002). The approach involves a direct comparison between the measured radiocarbon activity of the tree ring, and the record of atmospheric radiocarbon variability. Since the "bomb carbon" record shows an initial rise (as a result of above ground nuclear bomb resting) and a subsequent drop (the dilution of the atmospheric carbon as other reservoirs turn over after bomb testing went underground), there are typically two years in which the atmosphere matches any tree ring value. Only one of the intercepts can be correct, and by comparing the dates from several rings it is possible to determine which this is. Rings 17 and 25 calibrate to AD 1987 and AD 1979 respectively indicating that the rings are formed annually. The third sample comprising rings 124 to 134, taken from the centre of the tree, should have grown between AD1870- AD 1880. The most precise measurement of southern Hemisphere tree rings from this time is from McCormac *et al*. (2002), and their data suggest that the radiocarbon date should have been c. 150 BP or 98.2 percent modern carbon (pmc), but it is also noted that there are substantial inter-annual fluctuations that occur at this time. The result was 98.8 ± 0.95 pmc, which is within 1 sigma error of the anticipated value. The calibration is based on the INTERCAL98 dataset with a modelled 40-year southern hemisphere offset (Talma and Vogel, 1993) that corresponds very well with the INTERCAL04 dataset (Reimer *et al*., 2004)

On the basis of the three radiocarbon dates we conclude that *M. caffra* produces annual rings, Umhlanga 1 started growing in AD 1870, but only rings formed from AD 1927 onwards are used in the comparison with the climate variables because of the limits in the latter data. The isotope data set for Admiralty Reserve 1 spans the period AD 1960 to AD 2004.

4.5.2. $\delta^{13}C$ variability in response to environment

The growth rings from *M. caffra* are extremely variable in width and between five and seven isotope samples could be taken from the widest rings, while in narrower rings

only one or two samples could be removed. To avoid bias arising from the sampling, both the isotopic and climatic data were annualised. Climatic data (rainfall, temperature, humidity) were obtained from the South African Weather Service for three weather stations along the northern KwaZulu-Natal coast and were similarly annualised over the presumed growing season of trees in the southern Hemisphere (June-July). The available annual rainfall data (Fig. 4.3, Vol.II, pg.11) show a number of extremely wet periods as well as several dry periods during this time. The wettest years were 1951 and 1987, each with annual rainfall of over 1900mm. Other exceptionally wet years include 1950, 1975, 1983 and 1999. A drought occurred in 1958/9 and the driest conditions were recorded during 1992/3 where only 276mm of rain was recorded (Dube and Jury, 2003). The mean temperature record for the same time period (1927 to 2004) shows a gradual increase from approximately 1980 (Fig. 4.3, Vol.II, pg.11). The humidity record is limited, as consistent records were only available from 1981 to 2004. The 1992/3 drought is reflected as a notable decrease in humidity (Fig. 4.3, Vol.II, pg.11).

Figure 4.3 (Vol.II, pg.11) shows the variability in the $\delta^{13}C$ records for Umhlanga 1 and Admiralty Reserve 1. Both trees show evidence of a "juvenile effect" in the earliest years of growth. This manifested as more negative $\delta^{13}C$ values in the oldest rings and is due to the tree recruiting below the canopy where they are subject to lower light exposure that affects photosynthesis. Canopy thickness, leaf area and tree stand density will also have an influence on the "juvenile effect" (Norström, 2005; Vogel, 1978; West *et al*., 2001). In Umhlanga 1 and Admiralty Reserve 1, an overall downward trend in $\delta^{13}C$ values occurs after 1980. This may be a partial response to anthropogenic influences such as increased Greenhouse gasses, atmospheric pollution and an overall increase in mean temperature (February and Stock, 1999), but is almost certainly manifest in the isotopic value of the atmospheric CO_2. From the mid-1980s mean temperatures have gradually increased and the carbon isotope composition of the atmosphere has become more negative. The changes in the carbon isotope composition of the atmosphere can be seen in the record of $\delta^{13}C$ values of Pretoria air samples from 1968 to 2001 (Vogel *et al*., 2002) (Fig. 4.3, Vol.II, pg.11).

The climatic response manifests in the $\delta^{13}C$ record as a negative correlation with rainfall and temperature (Fig. 4.3, Vol.II, pg. 11). When rainfall deviates substantially

from normal (e.g. during tropical cyclones or droughts), this response is reflected in the $\delta^{13}C$ values. During periods of high rainfall $\delta^{13}C$ values become more negative and more positive in times of drier periods. This response is amplified during extreme years such as the extremely wet 1950/51 and 1992/93 drought years (Fig. 4.4, Vol.II, pg.13). A similar pattern is seen when $\delta^{13}C$ values are compared with temperature, although the correlation is weaker. Rainfall, however, is the main factor affecting the carbon isotope composition of trees, ameliorating the influence of temperature and anthropogenic factors. Due to this dominating influence the $\delta^{13}C$ values from *M. caffra* growth rings provide a record of past climatic conditions, particularly rainfall.

Based on the comparison of $\delta^{13}C$ values with rainfall it is possible to predict deviations in rainfall beyond the instrumental record (1927). The carbon isotope sequence for Umhlanga 1 extends to 1870 and the variation observed between 1870 and 1928 suggests that there is an irregular cycle (3 to 7 years) of wetter and drier than normal rainfall conditions. Historical records show that between 1870 and 1899 there was a rapid transition between wetter and drier than normal states (Nicholson, 2000). These fluctuations appear to be reflected in the $\delta^{13}C$ values of Umhlanga 1 from this period (Fig. 4.4, Vol.II, pg.13) Care should be taken when examining carbon isotope data from the earliest years of growth as the $\delta^{13}C$ values are affected by the "juvenile effect".

The superposition of the short-term climatic response (rainfall) and other long-term effects (juvenile and anthropogenic) in the isotope record complicates attempts to identify longer term rainfall forcing that might be incorporated into climate models. An approach that can be used is time series data analysis. The data sets were subject to Continuous Wavelet Transform (CWT) using a Morlet wavelet with a central frequency of 6 (Grossman and Morlet, 1984; Mallat, 1989). The data are decomposed into a frequency (period) domain which de-couples different underlying fundamental drivers. In respect of the isotopic data for Umhlanga 1 and Admiralty Reserve 1 the anthropogenic effect for example, decreases over the last two decades of tree growth while the juvenile effect affects approximately the first decade of growth. The result is a wave form with a frequency (period) in the order of 30-50 years (depending on the age of the tree) that dominates both time series. The presence of these effects is

manifest in the modulus of the CWT analysis (Figure 4.4A and 4.4B, Vol.II, pg.13) as red shading, while blue shading indicates frequencies that do not underlie the isotope data. A similar breakdown of the environmental variability is presented in figures 4.4C-E (Vol.II, pg. 13). It should be noted that the use of annualised data limits the minimum period in the analysis to approximately 2 years, while the upper limit of the period in the analysis is constrained to the cone of influence by length of the time series. As a result the inter-annual response to rainfall and the anthropogenic effect do not influence the analysis. This decoupling of drivers allows a comparison between the environmental forcing and the isotopic response of the tree without the interference of the overprinted signals.

The cross wavelet analysis (Hudgins *et al*., 1993) is a direct comparison between the underlying frequencies in two time series datasets (Figure 4.5AC_1-4.5AE_1, Vol.II, pg.14). The isotope data set from Umhlanga 1 (and Admiralty Reserve 1 but not shown) is compared with the environmental drivers, and where both have the same frequency of variation the modulus of the cross wavelet analysis is enhanced (depicted as red in Figure 4.5AC_1-AE_1, Vol.II, pg.14). Where a frequency is not present in either one or the other time series data set, there is no enhancement of the cross wavelet modulus (depicted as blue). The wavelet analysis and the cross of the isotopes with the environmental data highlight the 1950/51 wet period and the 1992/93 drought.

The relationship between the carbon isotope record preserved in *M. caffra* and rainfall is further enhanced by the cross wavelet phase analysis (Figure 4.5 AC_2-AE_2, Vol.II, pg.14). If rainfall variability from year to year is dominant in controlling the isotope values, then it should also consistently apply over multi-year time scales, but the superposition of the anthropogenic effect and the juvenile effect makes this difficult to asses. The phase relationship between the isotopes and the environmental variables is more coherent (the phase relationship is more or less constant across all periods of variability) for rainfall than any of the other variables. This is apparent from the dominance of near in-phase (yellow to pale blue) regions of the isotope/rainfall cross wavelet phase analysis (Figure 4.5AC_2, Vol.II, pg.14), which is in contrast to the high degree of phase variability (red to dark blue) of the isotopes/humidity or isotopes/humidity analyses (Figures 4.5AD_2 and 4.5AE_2, Vol.II, pg.14).

4.6. Conclusions

The isotopic variability inherent in wholewood samples of *M. caffra* provides a potential measure of past rainfall. An age model of this species was obtained using high precision radiocarbon dates and indicates that growth rings are produced on annual basis. During the period, 1927 to 2003, $\delta^{13}C$ values of *M. caffra* respond extremely well to a number of dramatic variations in the regional rainfall record, particularly during 1951 (wet), 1965 (dry), 1975 (wet), 1987 (wet), 1992 (dry) and 1997 (wet). The use of wavelets provides an optimal method that overcomes the seasonal and non-linear components of the data that restrict traditional inferences based on time series analysis. This approach is sensitive to extreme events such as tropical cyclones and droughts and it is noted that the periodicity is apparently random in contrast to other climate proxy data sets (Tyson *et al.*, 2002). The data imply that longer term climate forcing in the area is chaotic and a systematic seeding of climate change models with extreme events will only be probabilistically and not deterministically accurate.

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CHAPTER 5. STABLE CARBON ISOTOPE RATIOS FROM ARCHAEOLOGICAL CHARCOAL AS PALAEOENVIRONMENTAL INDICATORS

Title: Stable carbon isotope ratios from archaeological charcoal as palaeoenvironmental indicators

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Background to the paper

In this paper we demonstrate that an environmental proxy recorded in a tree ring sequence is conserved in the $\delta^{13}C$ values of the branch and charcoal from the same tree. The genus, *Podocarpus* was selected on the basis that it was the predominant species in certain Middle Stone Age (MSA) layers from Sibudu. The genus is considered as a dominant climax species in many of the forest types in South Africa.

Two trunk cores and corresponding branch samples, from Seaton Park (KwaZulu-Natal) were analysed and their $\delta^{13}C$ values compared. A trunk disc from a *P. falcatus* from the Baviaans Kloof (Eastern Cape) provided a $\delta^{13}C$ time series from trees growing in a more moisture-restricted environment, providing a contrasting analogue to the Seaton Park trees. The results for this specimen are presented in detail in Appendix B (section B.2), and show a response to annual rainfall variation. The $\delta^{13}C$ values of trunks and branches from Seaton Park both preserve the same environmental record, in this case correlating with humidity and temperature data from the region (Table 5.1., Vol.II, pg.54). Therefore it is possible to obtain a short record of past climatic conditions from the growth rings of branches. These results favourably suggest that an environmental signal may be preserved in branch charcoal.

Branch samples from five *P. latifolius* specimens were collected from Seaton Park. These branches were in the average size range of wood collected for fuel, currently utilised by firewood harvesting communities. The branches are likely to be representative of the size class of wood fuel collected by inhabitants of Sibudu Cave during the MSA. Branch discs were combusted under oxidising and reducing conditions and their respective $\delta^{13}C$ values compared with $\delta^{13}C$ values from their

corresponding un-burnt discs to determine if the same environmental record was preserved in burnt discs, as seen in the fresh wood discs. $\delta^{13}C$ values for the reduced and oxidised charcoal were depleted with respect to the source material but reflected the same response to prevailing climatic conditions as recorded in fresh wood. The branch sample sets provided evidence that it is possible to generate meaningful environmental signals from the $\delta^{13}C$ values of charcoal.

An additional experiment determined the range of isotopic variability in products released during various stages of combustion of a green branch of *P. falcatus*. This provided a further check that $\delta^{13}C$ values from charcoal are still representative of the original fresh wood signal. The results indicated that combustion temperature has a significant affect on the carbon isotope signature of the various products released during combustion, but the isotopic composition of the remaining wood tissue remains relatively constant. In wood-fuelled fires, the temperature range of 450 to 500°C depletes $\delta^{13}C$ values with respect to the source material, but the charcoal still preserves the seasonal and inter-annual isotopic trends. These modern analyses indicate that it is possible to obtain an isotopic record of past climatic conditions from archaeological charcoal. This was demonstrated through the comparison of archaeological charcoal $\delta^{13}C$ values from Sibudu and the modern analogue data. The archaeological charcoal $\delta^{13}C$ values from warm and moist periods were similar to those from Seaton Park and those from the cooler and drier periods fell within the range of $\delta^{13}C$ values from the Baviaans Kloof. The $\delta^{13}C$ values of archaeological charcoal therefore reflect environmental changes through time at Sibudu Cave. Additional proxy palaeoenvironmental data from the site have provided supporting evidence for these changes.

Stable carbon isotope ratios from archaeological charcoal as palaeoenvironmental indicators

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5.1. Abstract

The potential to provide environmental proxies using stable carbon isotopes from modern and archaeological charcoal is explored. Experiments on modern *Podocarpus* (Yellowwoods) show that $\delta^{13}C$ values of stems, branches and charcoal preserve proxy environmental conditions, including rainfall, humidity and temperature. An additional experiment showed that combustion temperature affects the carbon isotope signature of charcoal. Burning at 450°C to 500°C depletes $\delta^{13}C$ values with respect to the original wood, but the charcoal retains the seasonal and inter-annual isotopic trends recorded during the growth of the tree.

The $\delta^{13}C$ of *Podocarpus* charcoal from three levels from the Middle Stone Age site of Sibudu Cave, KwaZulu-Natal, South Africa, was compared with modern analogues from two different environments, Seaton Park (KwaZulu-Natal) and the Baviaans Kloof (Eastern Cape). Other environmental proxies from levels dated from >70ka and ~48ka, show that environmental conditions changed from warmer and wetter to colder and drier and finally becoming warmer and drier. The isotope data are consistent with this reconstruction. The results from this series of experiments indicate that it is possible to obtain meaningful palaeoenvironmental information from $\delta^{13}C$ values of archaeological charcoal

Keywords: Stable carbon isotopes, Archaeological charcoal, Palaeoenvironmental signals, Middle Stone Age

5.2. Introduction

The carbon isotope composition of wood varies in response to environmental parameters such as temperature, humidity and precipitation (e.g. Briffa *et al*., 2004; Edwards *et al*., 2000; Leavitt, 2002; McCarroll and Loader, 2004; Shu *et al*., 2005; Van Bergen and Poole, 2000). Charcoal and char are all forms of wood derived Black Carbon (BC) and it is possible that the isotopic environmental records will be preserved. Many climate and environmental studies have made use of stable isotope and other chemical data derived from burnt organic matter. These include examining changes in palaeovegetation (e.g. Behling and da Costa, 2001; Bendassoli *et al*., 2002; Biedenbender *et al*., 2004), carbon cycling and sequestration in ecosystems (e.g. Gleixner *et al*., 2001; Schmidt, M.W.I. *et al*.; 1999; 2002; Skjemstad *et al*., 1996) and changes in atmospheric carbon dioxide concentrations (e.g. Cope and Chaloner, 1980; Jones and Chaloner, 1991).

The processes by which charcoal is formed and preserved are well understood. Charcoal is defined as any blackened plant-derived material that has been significantly altered, chemically and structurally, through heating via fire (Forbes *et al*., 2006). It is considered to be biologically and chemically inert after formation and will undergo an extremely slow process of degradation (Cohen-Ofri *et al*., 2006; Forbes *et al*., 2006). Since charcoal has a high resistance to chemical oxidants (Bird and Gröcke, 1997; Skjemstad *et al*., 1996) it has a long residence time in deep soil profiles (Skjemstad et al., 2002) and geological records (Cope and Chaloner, 1980).

The goal of this paper is to demonstrate that it is possible to obtain meaningful palaeoenvironmental information from the carbon isotope ratios of archaeological charcoal. First we demonstrate that an environmental response to rainfall, temperature and humidity is recorded in modern samples of stems, branches and carbonised wood tissue of the selected genus, *Podocarpus.* We show that a degree of carbon fractionation occurs during the conversion of wood to charcoal. We present a continuous record of carbon isotope values for a wood sample combusted at a range of temperatures under oxidizing and reducing conditions to demonstrate the range of isotopic variability in the products of wood tissue combustion. The carbon isotope

ratios of archaeological *Podocarpus* charcoal (Allott, 2004, 2005, 2006) from selected layers representing three periods of the Middle Stone Age (MSA) sequence from the site of Sibudu Cave, KwaZulu-Natal, South Africa are then compared with the modern isotopic data. Additional archaeological isotopic data from *Celtis* charcoal are provided as further evidence that a palaeoenvironmental signal is preserved in archaeological charcoal.

5.3. Recent stable carbon isotope research on charcoal

In many environmental studies it is assumed that carbon isotope ratios derived from naturally occurring and anthropogenic charcoal are a direct representation of the isotopic values of the wood tissues from which they were formed, and hence a record of environmental and climatic signals (e.g. Behling and da Costa, 2001; Bendassoli *et al*., 2002; Biedenbender *et al*., 2004; Cope and Chaloner, 1980; Gleixner *et al*., 2001; Jones and Chaloner, 1991; Schmidt, M.W.I., *et al*., 1999; Skjemstad *et al*., 1996,). This assumption implies that there is an insignificant or constant fractionation of carbon isotopes during the burning process (Turney *et al*., 2006), but the changes that take place in the stable carbon isotope composition of plant material during carbonisation are still not well understood. Wood is composed of a range of macromolecules such as cellulose, lignin and various polysaccharides. These all have a characteristic carbon isotope composition and their relative abundances contribute to the overall isotopic composition of wood (Poole *et al*., 2002). During combustion these macromolecules are broken down into various specific pyrolysis products, mostly furans and pyrans, and released (Steinbeiss *et al*., 2006). The removal of these compounds is driven by their relative chemical stability and strength of molecular cross bonds. Differences between how these macromolecules break down during combustion allow cross-reactions between their products and produce a number of additional compounds prior to release (Steinbeiss *et al*., 2006).

Studies carried out to determine the effects of combustion on the carbon isotope composition of plant tissues and their products of combustion have produced varied and often contradicting results. A study comparing C_3 and C_4 vegetation (Turekian *et al*., 1998) showed that under laboratory conditions the C_3 vegetation yielded small differences in $\delta^{13}C$ values between the source material and aerosols, residues and

gasses produced during combustion. For the C_4 material there was a substantial fractionation during combustion. The $\delta^{13}C$ values for the C_3 fatty acids from the burnt samples showed enrichment relative to the un-burnt material (Ballentine *et al*., 1998). This is in contrast to the $\delta^{13}C$ results for the aerosols and particles collected by Turekian *et al*. (1998), where there was little or no difference in the $\delta^{13}C$ values of the burnt and un-burnt material. The opposite was seen in data collected from sediment-derived charcoal samples where no significant changes in $\delta^{13}C$ values were observed between burnt and un-burnt C_4 samples (Cachier *et al*., 1985; Krull *et al*., 2003). Bird and Gröcke (1997) found that charring wood and leaf samples resulted in more negative $\delta^{13}C$ values when compared with the un-burnt source material. An isotopic analysis of the combustion products of cellulose and lignin (Steinbeiss *et al*., 2006) indicated that there were no significant differences between the $\delta^{13}C$ values of the bulk cellulose and lignin and the mean $\delta^{13}C$ values of the volatile compounds. These results are similar to those presented by Turekian *et al*. (1998). However, it should be noted that the $\delta^{13}C$ values of some of the individual compounds were substantially different from the $\delta^{13}C$ values of the bulk material (Steinbeiss *et al*., 2006).

Significant variations in the overall chemistry of wood tissues were observed at combustion temperatures higher than 200°C (Baldock and Smernik , 2002), but despite the variability the overall chemical composition of the wood tissues was fixed within thirty minutes of combustion regardless of temperature (Turney *et al*., 2006). There are indications that the $\delta^{13}C$ values of the remaining source materials tend to be depleted, likely due to the selective removal of a ^{13}C enriched fraction and to an extent the degree of oxidation. The $\delta^{13}C$ depletion between the source material and the products of combustion in the various experiments ranges from as little as 0.1‰ to 0.4‰ (Krull *et al*., 2003; Turekian *et al*, 1998; Turney *et al*., 2006; Steinbeiss *et al*., 2006), whilst others show a more extensive change of between 1.0‰ to 7.0‰ (Leavitt *et al*., 1982; Turekian *et al*., 1998). There are however, exceptions to this. A set of propagules (peas) burnt at a range of temperatures showed enrichment in $\delta^{13}C$ values (Poole *et al*., 2002) and wood tissue subject to an artificial ageing treatment using heat and water (Schleser *et al*., 1999) produced enriched $\delta^{13}C$ values relative to the source material.

5.3.1. Charcoal stable carbon isotope research in southern Africa

In southern Africa research was carried out to determine the feasibility of obtaining climatic information from $\delta^{13}C$ analyses of archaeological charcoals by correlating modern data with rainfall records (February and Van der Merwe, 1992; February, 1997; 2000). An initial study showed that $\delta^{13}C$ values of wood charcoal varied through time and that such change was climatically induced (February and Van der Merwe, 1992; February, 1994b). Archaeological charcoal from three sites in the Elands Bay area, Western Cape Province, identified from the family Ebenaceae (genus *Diospyros/Euclea)* had significantly different $\delta^{13}C$ values from those of modern charcoal values from the same genera (February, 1992; February and Van der Merwe, 1992). The archaeological $\delta^{13}C$ values also showed variability through time with the oldest charcoal (4200 BP) having more negative values than the more recent material (460 BP). This result was attributed to a range of climate-influenced plant $\delta^{13}C$ values (February and Van der Merwe, 1992).

Further work was carried out to compare stable carbon isotopes in charcoal, wholewood and cellulose and to examine their isotopic relationship to rainfall (February, 1997). The $\delta^{13}C$ values from two woody species, *Combretum apiculatum* and *Protea roupelliae* were differently affected by rainfall. *C. apiculatum* $\delta^{13}C$ values for the wholewood and cellulose correlated significantly with rainfall values across a rainfall gradient. The $\delta^{13}C$ values from the *P. roupelliae* samples did not show any significant correlation with the same rainfall data. February (1997) attributed this difference to plant habits: *C. apiculatum* is drought deciduous with specific habitat requirements, whereas *P. roupelliae* is evergreen and grows in a wide range of habitats. None of the charcoal $\delta^{13}C$ values of either species showed a significant correlation to rainfall. This was thought to be as a result of the loss of hemicelluloses, cellulose and lignin during various stages of pyrolysis. The loss of tissue during combustion resulted in $\delta^{13}C$ values becoming depleted on average by 0.5‰ at 400°C and 1.0‰ at 500°C. This fractionation is not constant, with different pieces of wood reacting differently during pyrolysis.

It is obvious, based on the examples, that there is no singular explanation for the changes seen in carbon isotope ratios from burnt and un-burnt plant material. Whether

products of combustion are depleted or enriched in their respective $\delta^{13}C$ values is dependent on a number of factors. These include the type of plant material burnt (e.g. leaves, wood, seeds), size and surface area, available oxygen, photosynthetic pathway, cellulose/lignin ratio and amount and concentration of volatile compounds as well as the temperature at which the material is combusted (Baldock and Smernik, 2002; Ballentine *et al*., 1998; Bird and Gröcke, 1997; Cachier *et al*., 1985; Krull *et al*., 2003; Leavitt *et al*., 1982; Poole *et al*., 2002; Schleser *et al*., 1999; Steinbeiss *et al*., 2006; Turekian *et al*., 1998; Turney *et al*., 2006). In addition the heating temperature and length of burn will also influence $\delta^{13}C$ ratios. A further consideration is the various sample pre-preparation methods employed prior to combustion and the methods of combustion used before carbon isotope ratios are measured (Krull *et al*., 2003). Laboratory experiments may not produce results completely comparable to results derived from material produced in natural fires. Carbon isotope values of compounds released from flash pyrolysis experiments will differ from those produced under slow combustion conditions, such as natural burns or anthropogenic fires. This will be due to the production of secondary reaction compounds during a longer period of combustion. In this study we present $\delta^{13}C$ values from compounds and source material produced under combustion where the overall temperature increase is slow and the length of burn is considerably longer than flash pyrolysis.

Notwithstanding the complexity, there have been a number of studies that have shown that it is possible to obtain significant palaeoenvironmental information from $\delta^{13}C$ values from charcoal (e.g. Bendassoli *et al*., 2003; Bird and Gröcke, 1997; February and van der Merwe, 1992). It may be some time before an all-encompassing theoretical framework will predict the $\delta^{13}C$ of all trees from environmental conditions therefore we restrict ourselves to an empirical approach. We focus specifically on *Podocarpus.* The data presented in this paper show that it is possible to obtain meaningful environmental information from archaeological charcoal of this species since environmental causality is demonstrated in modern specimens.

5.4. Study area, material and methods

5.4.1. Background to Sibudu Cave

Sibudu Cave is located approximately 40 km north of Durban, about 15 km inland of the Indian Ocean, on a forested cliff overlooking the Tongati River (Fig. 5.1, Vol.II, pg.15). The site has a deep, well-preserved and well-dated MSA sequence that has been subject to archaeological excavation since 1998 (Wadley and Jacobs, 2004; 2006). Preservation of organic material, such as bone, charred botanical remains and charcoal from fireplaces is extremely good. The range of additional published environmental evidence currently available from the site includes charcoal and seed analyses (Allott, 2004; 2005; 2006; Sievers, 2006, Wadley, 2004), micro and macrofaunal analyses (Cain, 2006, Glenny, 2006; Plug, 2004; 2006; Wells 2006), sedimentological studies including magnetic susceptibility, palynological and phytolith analyses (Herries, 2006; Pickering, 2006; Renaut and Bamford, 2006; Schiegl *et al*., 2004; Schiegl and Conard, 2006) and tracking of temporal changes in communities through correspondence analysis (Reynolds, 2006).

The site was excavated by natural stratigraphy and layers are individually named using a nomenclature that refers to the colour of the predominant matrix (Wadley and Jacobs, 2006). The MSA stratigraphy is complex, but is generally clear with numerous thin, coloured interfingered layers and distinct well-preserved hearth structures (Fig. 5.2, Vol.II, pg.16). Sedimentological analyses of the deposit (Pickering, 2002; 2006) indicate that sediments mainly comprise anthropogenically derived material with very little evidence of water-borne transportation. These layers have been given abbreviated letter designations.

Fourteen optically stimulated luminescence (OSL) dates have been obtained and they suggest that the currently excavated MSA layers range in age from >70ka to ~35ka years ago (Jacobs *et al*., 2008a). The cultural sequence is broadly divided into undated layers older than 60ka and three younger age clusters (Jacobs *et al*., 2008a). The undated period includes the Howiesons Poort (HP) lithic assemblage, the Still Bay and several informal lithic industries. The three age clusters have been informally named on the basis of the lithic assemblages within them. The ~58ka

assemblages are referred to as the post-Howiesons Poort (post-HP), the ~48ka assemblages as the late MSA and the ~35ka assemblages as the final MSA. The archaeological charcoal analysed in this study was taken from the HP, the post-HP and the late MSA assemblages. No suitable charcoal samples were available from the final MSA.

A complex mosaic of habitats may always have existed near the shelter because of the influence of the Tongati River and because of the combination of shaded south-facing hill slopes and warm north-facing hill slopes. Based on recent analyses of seed (Wadley, 2004; Sievers, 2006), charcoal (Allott, 2004; 2005; 2006) and micro/macrofaunal assemblages (Cain, 2006; Glenny, 2006; Plug, 2004; 2006; Reynolds, 2006) from the site, the HP layers (GR and GS) represent a period of warm, moist conditions with a predominance of evergreen forest (dominated by *Podocarpus spp.*), probably on the southwest-facing hillside and some savanna species possibly on the north-facing slopes. During the post-HP period (SPCA) at ~58ka, conditions were cooler and drier than present with varied habitats that included some evergreen forest (perhaps restricted in its extent), riverine forest and open woodland. There appears to have been a shift to warmer (though still cooler than present) dry conditions with bushveld and limited forest communities during the late MSA (RSp and OMOD) at ~48ka.

5.4.2 Archaeological samples

The genus *Podocarpus* was selected for isotopic analyses because of its predominance in certain of the MSA layers at Sibudu Cave. *Celtis* charcoal was sampled from the same archaeological periods as the *Podocarpus* charcoal on the assumption that the different sensitivity of this genus to environmental factors might provide a more robust interpretative framework than the *Podocarpus* alone. The charcoal was identified using important wood anatomical features and a number of reference collections (Allott, 2004; 2005; 2006). The oldest layers analysed in this study, Grey Rocky (GR) and Grey Sand (GS) (Fig. 5.2, Vol.II, pg.16) are still undated, but are older than 60 ka and probably younger than 70ka (Wadley and Jacobs, 2006; Wadley, 2006). These layers are representative of the HP (>60ka) period. One hundred and

twenty two charcoal sub-samples (18 fragments) identified as *Podocarpus* were obtained from the GR and GS layers. A small set of 7 sub-samples (1 fragment) of *Celtis* charcoal was taken from GS. The next oldest layer sampled was Spotty Camel (SPCA) and has an age of ~58ka. This layer represents the post-HP period. Twenty sub-samples (2 fragments) of *Podocarpus* and 12 sub-samples (1 fragment) of *Celtis* were taken from SPCA. It should be noted that two species of *Podocarpus* were recorded for SPCA: *P. latifolius* and *P. falcatus* (Allott, 2004; 2005; 2006). The youngest samples were taken from two layers from the late MSA. Fifteen sub-samples (1 fragment) of *Podocarpus* were from Red Speckled (RSp) which has an OSL date of 46.6 ±1.9ka (Jacobs *et al*., 2008a). Thirty-six sub-samples (3 fragments) of *Celtis* were obtained from the Orange Mottled Deposit (OMOD) layer. OMOD has an OSL date of 48.3 ±1.8ka (Jacobs *et al*., 2008a).

These sample sets were selected from layers considered to be representative of the different climatic and environmental conditions that occurred in the Sibudu region during Oxygen Isotope Stages 3 and 4, as indicated by published palaeoenvironmental data. These environmental shifts may be reflected in the carbon isotope composition of the archaeological charcoal, if an environmental signal has been retained.

5.4.3. Modern Samples

Podocarpus is generally a dominant climax genus in many of the tropical and subtropical forest types in South Africa (Acocks, 1988; Killick, 1990; Palmer and Pitman, 1972; Pooley, 2003; Schmidt, E., *et al*., 2002) and their adaptability allow the genus to establish and maintain its presence. The genus is therefore potentially a good indicator of environmental change over time. The presence of *Podocarpus* implies a well-forested environment with high moisture availability. An absence or limited occurrence of the genus may imply a reduced forested environment and drier conditions and the genus may thus be restricted to sheltered and protected portions of the landscape. Archaeological charcoal from a second genus, *Celtis*, was subject to isotopic analysis. This genus has a larger distribution and the ability to adapt to a wider range of environments, particularly drier ones ((Acocks, 1988; Coates-Palgrave 2003; Pooley, 2003; Van Wyk and Van Wyk, 1997).

Five specimens of *Podocarpus latifolius* (Real Yellowwood) (Coates-Palgrave, 2003; Pooley, 2003; Van Wyk and Van Wyk, 1997) were sampled from a remnant portion of Coastal Forest (Low and Rebelo, 1998; Lubke *et al*., 1997; Lubke and McKenzie, 1998). This section of indigenous forest has been preserved as a small suburban park, called Seaton Park, located to the north of the city of Durban, about five kilometres from the coastline. There are about 100 species of indigenous trees in a fairly densely wooded environment (Nichols and Fairall, 1992). The park is located on the lower eastern slope of a steep-sided valley and is protected from winds originating from the coast. The *P. latifolius* trees sampled are located on a rough transect with samples from the upper region of the park down to the lower area that was at one stage a marshland (Nichols and Fairall, 1992).

Core samples (2, 3 and 4) were removed from the trunks of two *P. latifolius* specimens (B and C) for comparison with corresponding branch samples. Branch samples were cut from five selected *P. latifolius* specimens (B, C, L, M, N). The branches range in diameter from 30 to 60mm, which falls within the size ranges currently collected by firewood harvesting communities (Abbott and Lowmore., 1999; Sekhwela, 2003; Van Wyk and Gericke, 2000). In much of rural Africa, branches collected for firewood would be predominantly derived from deadwood that had either fallen to the ground or that was easily accessible. Modern ethnobotanical studies have shown that harvesting firewood is predominantly done on naturally produced, dry (dead), but not rotten, wood (Archer, 1990; Abbott and Lowmore, 1999; Shackleton, 1998; Sekhwela, 2003; Van Wyk and Gericke, 2000). Some dead wood is cut or chopped from trees or shrubs from easy-to-reach branches and thin trunks.

An additional trunk sample of *Podocarpus falcatus* from the Baviaans Kloof area, Eastern Cape Province (Fig. 5.1, Vol.II, pg.15) formed part of the modern sample set. This tree was analysed to provide a stable carbon isotope record from trees growing in a moisture restricted environment as a contrast to the Seaton Park trees, which grow in an environment with much higher moisture availability.

A single green branch sample of a specimen of *P. latifolius* was collected from the grounds surrounding the laboratory in Pretoria and was subjected to isotopic analysis to determine the range of variability in the carbon isotope values of the products released from wood tissues during the stages of combustion

5.4.4. Wood and charcoal preparation and analysis

Three discs, 15mm thick, were cut in sequence from each branch from Seaton Park. These discs, core samples and the Baviaans Kloof sample were sanded with increasingly finer grades of sandpaper (grades P40µm to P1200µm) until their growth ring sequences were clearly visible. The total number of growth increments was established for each disc/core using a binocular microscope and an incident light source.

One disc from each sample set was left un-burnt; the second set was tightly double wrapped in heavy duty aluminium foil to exclude oxygen and was placed in a pre-heated muffle furnace at 450°C for two hours. This produced charcoal under a reducing (O_2-limited) environment. The third set of discs was placed on a foil-covered grid over an open wood fire to simulate natural burning conditions and left until completely charred. This produced charcoal under oxidizing (O_2-rich) conditions. This process took less than 15 minutes for the largest disc (60mm in diameter) and about five minutes for the smallest (30mm in diameter).

Material for isotope analysis was drilled along two adjacent offset transects using a 2mm drill bit, from the intersection of the cambium and bark with the last visible growth ring to the centre of the disc or end of the core. This method of sampling growth ring sequences has proven to be successful in a number of stable isotope studies (McCarroll and Loader, 2004; Norström, 2005; Woodborne and Robertson, 2000; 2001; Woodborne *et al*., 2003). Each sample was allocated to a portion of a corresponding ring or rings to allow comparison of the $\delta^{13}C$ values. The growth rings from *P. latifolius* are extremely variable in width. Between three and five samples could be taken from the widest rings, but in narrower rings only one or two samples could be removed. This precludes examining the isotopic variability on a sub-annual

basis in this species, and so the $\delta^{13}C$ ratios for each core/branch were annualised by averaging the results for each ring.

The method used to pre-treat the wood samples was developed by Green (1963) and adapted by Loader *et al*. (1997). This method allows for the rapid batch processing of small wood samples for stable carbon isotope analysis. Samples were run through an ethanol/toluene Soxhlet distillation process in order to remove mobile constituents such as resins, oils and waxes. The Soxhlet process removes those materials that may not coincide with the formation of the growth rings and ensures that they are not reflected in the stable carbon isotope values obtained (Loader *et al*., 1997). The un-burnt wood samples were run as wholewood. Some recent isotope studies of tree ring sequences have produced stronger climatic signals from wholewood samples than those derived from either α-cellulose or lignin (Loader *et al*., 2003; McCarroll and Loader, 2004).

The charcoal samples from the two burning methods were placed in Pyrex test-tubes and covered with a 1% hydrochloric acid solution. These were placed in an oven at 70°C overnight, washed with distilled water until pH neutral and dried. The archaeological charcoal samples were pre-treated with 1% HCl overnight and washed with distilled water until pH neutral.

Aliquots of approximately 0.2mg were combusted on-line in a Thermo Flash Elemental Analyser (1112 series) integrated via a Thermo Finnigan Con-flo III system with a Thermo Delta V Plus Isotope Ratio Mass Spectrometer, housed at the Ecosystem Processes and Dynamics (EPD) laboratory, CSIR, Pretoria. Each sample was run in duplicate. If the precision for each sample pair was unacceptable (0.10‰ variation), an additional sub-sample was run. The average precision for the modern replicates was <0.06‰ and for archaeological samples <0.08‰.

An additional experiment was carried out using a green sample of *P. latifolius*. A 10mm long piece of the branch was placed directly into the furnace of the elemental analyser, with no pre-treatment. The temperature of the furnace was manually increased in steps by between 5 and 15°C and the carbon isotope composition of the released products were measured using the mass spectrometer at each temperature

increment. This was done under a reducing environment until a temperature of 1000°C was reached. Once the furnace reached 1000°C, oxygen was introduced and carbon isotope composition of the gasses released were measured at regular intervals until the sample was completely combusted.

Two project specific standards, a C_4 grass (*Hyparrhenia hirta*) and C_3 tree (*Shorea superba*) were used. These species were selected as they each represented average carbon isotope values for C_3 and C_4 plants. All stable carbon isotope results were expressed in delta (δ) notation on a per mille (‰) scale. The stable carbon isotope ratios were measured as deviations from international standard reference materials and are reported relative to the Vienna PeeDee Belemnite (VPDB) standard (Coplen, 1995) using the following equation (Francey and Farquhar, 1982):

$$\delta_{sample} = \left(\frac{\left(R_{sample} - R_{s\tan dard} \right)}{R_{s\tan dard}} \right) \times 1000$$

$$R = {}^{13}C/{}^{12}C$$

5.5. Results and discussion

5.5.1. Comparison of $\delta^{13}C$ values from modern trunk cores and branches

The annualised carbon isotope variability for three core samples (Core 2, 3 and 4) as well as the branch data (Branch B and Branch C) were compared with annual rainfall, temperature and humidity records for the region (Fig. 5.3, Vol.II, pg.17). A striking feature of the data is that Core 2 and Core 3, despite being from the same tree, show quite substantial differences. The implication is profound. At any point in time the tree appears to have the ability to partition its resources in such a way that the isotopic signal of the wood formed in one part of the tree differs from that in another. This vital fractionation appears to remain relatively consistent through the growth of the trees, as all three cores appear to have common features that might be anticipated on the basis of the theoretical discourse on the control of carbon isotope values in trees. During three periods of multi-year below-average rainfall (1963-1968, 1978-1980, and 1992-1993) the isotope values in the trees become progressively more positive. During intervening years the results tend in a negative direction.

There is a degree of redundancy between the environmental datasets (although the comparison is limited to last 23 years in the case of humidity) and so a statistical regression approach was adopted to determine which of the environmental variables best correlates with the isotope data. A summary is presented in Table 5.1(Vol.II, pg.55). Core 2 correlates with both temperature and humidity at the >99 % and >95% confidence levels, but core 3, from the same tree, only correlates with humidity at the >99% confidence level. This suggests that temperature plays a role in the partitioning of resources in the tree and will likely link to the aspect of the area sampled relative to insolation. Core 4 is also correlated with temperature (>99% confidence) but not humidity nor rainfall. Both of the branch samples have weak correlations with rainfall and humidity, and none with temperature.

The correlation between carbon isotopes and humidity appears to be common to all the samples with the exception of Core 4. This may be a result of the position of this tree within a riparian zone. The persistence of water availability at the roots of this tree may mean that it is not water limited and that the control of photosynthesis in then by temperature (possibly as a proxy for sunlight).

5.5.2. Charcoal experiments

Two factors that produce variation in charcoal isotope signatures are the formation temperature and the isotopic value of the original wood. During combustion certain moieties will volatilize and others will concentrate. The rate at which the combustion temperature increases will affect the amount of secondary reaction products formed (Gundale and DeLuca, 2006). Secondary products may be more thermally stable and persist for a longer period within the source material. In respect of the source material different plant tissues contain different proportions of chemicals (e.g. lignin, cellulose) and have different physical properties. Therefore different woody species will produce different qualities of charcoal based on their physical and chemical properties. In the case of archaeological charcoal, it will be derived from woody species that burn slowly and produce a large amount of long-lasting charcoal. These species would have been selected by the original inhabitants of an archaeological site. Since fuel wood is generally collected in the form of branches rather than tree trunks (Archer, 1990; Abbott and Lowmore, 1999; Shackleton, 1998; Sekhwela, 2003; Van

Wyk and Gericke, 2000), the charcoal from archaeological sites will likely be derived from branches burnt as fuel. It remains to be demonstrated that branch $\delta^{13}C$ values preserve the environmental signal during the process of charcoal formation.

The $\delta^{13}C$ values for the charcoal experiment in which disks from each of 5 branches were subject to different charcoal formation protocols are presented in Figure 5.4 (Vol.II, pg.18). Under both reducing and oxidizing conditions the overall isotopic trends are similar to that seen in the wholewood $\delta^{13}C$ values. In all five branch samples the $\delta^{13}C$ values for charcoals produced under both a reducing and oxidizing environment show depleted isotopic values relative to the wholewood (Fig. 5.4, Vol.II, pg.18). These results are consistent with the majority of findings from previous combustion experiments (See section 5.3) where the products of combustion (gasses, aerosols and residues) had $\delta^{13}C$ values that were more negative with respect to the C_3 source material (Ballentine *et al*., 1998; Bird and Gröcke, 1997; February, 1997; Czimczik *et al*., 2002; Steinbeiss *et al*., 2006; Turekian *et al*., 1998; Turney *et al*., 2006). There were occasional exceptions to this trend. In branch C, $\delta^{13}C$ values from three rings showed enrichment relative to the wholewood values and one ring (AD 1994) from branch M also produced an enriched $\delta^{13}C$ value. This occasional enrichment seen in these two branches may be as a result of the presence of different chemical compounds in those particular growth rings. Overall the depletion ranges between -0.02‰ and -3.00‰. Branch L produced $\delta^{13}C$ values with the greatest depletions relative to its corresponding wholewood values. This branch was collected in a live state and had a higher concentration of ^{13}C enriched volatile compounds or simple saccharides that were released during combustion resulting in more ^{13}C depleted charcoal. The other branches were collected in a dry (dead) state and would have lost the more easily released volatile compounds. The $\delta^{13}C$ values of the reduced charcoals (mean = -1.4‰) are slightly more negative than the oxidized charcoals (mean = -1.2‰).

The apparent differences in $\delta^{13}C$ between wholewood and the oxidised and reduced BC products imply that a selective pyrolitic distillation takes place during charcoal formation. The elemental analyser combustion experiment provides an indication of the sequential loss of different macromolecules and associated secondary products from the wood on the basis of their strength and degree of cross-linking. The degree

of isotopic depletion or enrichment of the remaining charcoal will depend on the extent to which fractions are driven off during combustion, the compound-specific isotope value and the absolute abundance of these fractions. Ideally each fraction could be identified and the $\delta^{13}C$ related back to the physiological process involved in its formation. However our approach is limited to an empirical quantification of the sequential process of carbon isotope fractionation in charcoal formation in *Podocarpus*. Further research is required to identify the precise chemical makeup of the pyrolitic distillation products.

The initial measurements during the elemental analyser combustion experiment were done without the addition of oxygen by the instrument. The evolution of CO_2 in the oxidation column of the elemental analyser implies that volatile carbon containing molecules are driven from the wood. Under reducing conditions it is clear that the combustion temperature has a significant effect on the carbon isotope fractions driven off. Initially $\delta^{13}C$ values are more positive, probably due to the loss of simple saccharides and ^{13}C enriched volatiles that are not thermally stable. After 200°C they are generally more negative. A second stage of more positive fractions is released at about 350°C. Between 100°C and 475°C, the average temperature range for a normal wood-fuelled fire, $\delta^{13}C$ values of released compounds are more negative with respect to the source material. The $\delta^{13}C$ values between 475°C and 1000°C (Fig. 5.5, Vol.II, pg.19) show a large range of variation, with extremely positive values (-17.0‰) and very negative values (-31.0‰). This extreme variability may be as a result of secondary products breaking down at higher temperatures.

When the incremental increases in temperature failed to produce further CO_2, the elemental analyser was reprogrammed to inject oxygen in order to oxidise any remaining (black) carbon. At this point the range of variability in $\delta^{13}C$ values was greatly reduced.

The result of the controlled charcoal experiment demonstrate that the potential for fractionation of the carbon isotope signal in wood during charcoal formation is high, but that this will offset the former data set relative to the latter. The elemental analyser experiment suggests that this mechanism involves the release of particular compounds, and that the consistency of the offset will likely be linked to the

temperature of the fire in which the charcoal was formed. However further experimentation is necessary to better understand the processes. Provided the offset during charcoal formation is consistent, the results suggest that archaeological charcoal may be a source of palaeo-environmental proxy data.

5.5.3. Comparison of modern and archaeological $\delta^{13}C$ values

Grouped modern and archaeological *Podocarpus* charcoal $\delta^{13}C$ values are presented in the form of box and whisker plots (Fig. 5.6, Vol.II, pg.21). The HP (> 60ka) data corresponds well with the modern *P.latifolius* charcoal data from Seaton Park and it is likely that the Coastal Forest found in Seaton Park provides a good analogue for environmental conditions during the HP period. Botanical and faunal assemblages from the HP levels indicate that the environment around Sibudu was predominantly evergreen forest with cool, moist and humid conditions (Allott, 2004; 2005; 2006; Cain, 2006; Glenny, 2006; Herries, 2006; Pickering, 2006; Plug, 2004; 2006; Renaut and Bamford, 2006; Reynolds, 2006; Schiegl *et al*., 2004; Schiegl and Conard, 2006; Sievers, 2006; Wadley, 2004; 2006).

The post-HP (SPCA) stands out clearly within the archaeological sample set because of the mean isotopic value and also because it has the smallest standard deviation, sample variance and range. Furthermore the isotopic values are derived from two different species of *Podocarpus*, identified by Allott (2005, 2006) (Table 5.2, Vol.II, pg.55). This cannot be explained by the analogous modern isotopic data from Seaton Park. The SPCA charcoals had to have been produced in the same way as the other archaeological charcoals and the material did not appear to be any less burnt than that from the late MSA and HP. Therefore the $\delta^{13}C$ values for the SPCA charcoals must represent an isotopic record of markedly different environmental conditions compared to those of the HP period. The current proxy environmental data from Sibudu Cave (See section 5.4.1) indicate that conditions were cold and dry with restricted forested areas and an increase in open grass and woodland faunal and floral communities. Forest communities would have been limited to sheltered areas along the riverside where moisture levels were sufficiently high for *Podocarpus* to grow (Allott, 2006; Sievers, 2006; Wadley, 2006), but overall conditions were dry resulting in more

positive $\delta^{13}C$ values. Positive excursions in the isotope values of the modern *Podocarpus* specimens are correlated with reduced rainfall and humidity, or with an increase in temperature. Since the independent proxies suggest that temperatures were colder than at present we conclude that the isotope evidence points towards a drier environment. Here the Baviaans Kloof may present an analogous environment. Although there is no data for burnt material from the Baviaans Kloof, based on our burning experiments, estimated $\delta^{13}C$ values for carbonised material from Baviaans Kloof would be depleted by -2‰ to -3‰. The $\delta^{13}C$ values for SPCA would fall into the range of variation seen in the Baviaans Kloof carbon isotope values, but comparing the range of variability it may imply an even drier environment.

Further research will elucidate the significance of the low variance in the SPCA carbon isotope values for *Podocarpus*, but it is possible that this reflects the degree to which the full adaptive response of the genus to variance in rainfall/humidity was possible. Such a scenario may apply if conditions were sufficiently dry that the forest could only perpetuate in refugia such as river valleys (similar to the Baviaans Kloof). The environmental constraint on growth would imply highly reduced interannual variability in the rainfall/humidity response of the trees because of the persistence of riparian water, but with a possibly over-riding dry response.

The isotopic evidence from the late MSA (RSp) layer is consistent with a warm (cooler than present) and dry environment. Proxy environmental data (See section 5.4.1) from RSp suggests that a major environmental shift occurred. This resulted in a warmer mosaic environment of grassland, woodland and savanna communities with riverine forest along the Tongati River providing a number of potential locations for *Podocarpus* to establish (Allott, 2006; Glenny, 2006; Sievers, 2006; Wadley, 2006).

A similar trend of isotopic variation is seen in the mean $\delta^{13}C$ values of *Celtis* charcoal from the same periods (Fig. 5.7, Vol.II, pg.22). The *Celtis* $\delta^{13}C$ values from the post-HP (SPCA) period are also markedly more positive than those from the HP. This suggests that this genus was responding in a similar manner as *Podocarpus* to drier conditions during the post-HP. As two very different genera are showing similar trends in their isotopic composition over time, it is very likely that an environmental signal of local conditions is preserved in their charcoal. It is also interesting to note

that the variability in the *Celtis* results from the Grey Sand (GS) level is very small (based on a small sample of 7) and that the adaptive strategy of this genus might have been limited at this time in much the same way that *Podocarpus* was during the deposition of layer SPCA. The evidence for a moist, evergreen forest environment suggests that *Celtis* was at the wet limit of its adaptive capability.

5.6. Conclusions

A two-step process by which the carbon isotopic composition changes during combustion manifests at temperatures below 150°C to 200°C, when isotopic values of the products of combustion can either become more enriched (Czimczik *et al*., 2002) or more depleted (Schleser *et al*., 1999), while above 150°C to 200°C, the tendency is for $\delta^{13}C$ values to become more negative. This has been linked to a pyrolitic distillation of chemical compounds within the wood, but the chemical changes that take place in a branch during drying must also be considered. When a branch dies certain volatile and labile compounds are easily removed as the branch dries out. Charcoal formation takes place under reducing combustion, during which less volatile components and labile compounds are removed. During oxidizing combustion, oxygen from outside the wood will eventually oxidize and remove the majority of material, resulting in the formation of ash, provided the temperature remains high enough. In the case of an anthropogenic fire, charcoal will only form in the centre regions of the fire where there is no outside oxygen available (reducing environment).

The main factor affecting the isotope composition of charcoal is therefore the combustion temperature. Normal wood-fuelled fires burn at an average temperature of 450°C to 500°C and although overall $\delta^{13}C$ values are depleted, this temperature range does not obscure the environmental signals originally recorded in the wood tissue. Accordingly it is possible to obtain environmentally meaningful carbon isotope values from archaeological charcoal, but it is necessary to establish modern equivalents from a number of different environments to determine the full range of possible adaptive strategies available to that particular species. The comparison of modern wood and charcoal, and archaeological charcoal carbon isotope values has shown that branches and carbonised wood of *Podocarpus* and *Celtis* preserve a record

of local environmental conditions and it is therefore possible to use $\delta^{13}C$ data from archaeological charcoal as a palaeoenvironmental proxy.

The environmental changes reflected in the isotopic data from the archaeological charcoal from Sibudu Cave are supported by additional proxy environmental evidence indicating that conditions changed from warm and moist in the Howiesons Poort to cooler and drier in the ~58ka (SPCA) period and then slightly warmer again during the ~48ka late MSA (RSp) (Allott, 2004; 2005; 2006; Cain, 2006 Glenny, 2006; Herries, 2006; Pickering, 2006; Plug, 2004; 2006; Renaut and Bamford, 2006; Reynolds, 2006; Schiegl *et al*., 2004; Schiegl and Conard, 2006; Sievers, 2006; Wadley, 2004; 2006). These changes in environmental conditions affected the adaptive responses of *Podocarpus* species and these responses are shown in the carbon isotope ratios of the identified archaeological charcoal. The *Podocarpus* and *Celtis* species growing during the Howiesons Poort and late MSA periods had a wider range of adaptive responses than those growing around Sibudu Cave during the post-HP period. This limited adaptive response around 58ka was due to the expansion of an open grassland environment and the restriction of evergreen forests to the river valley during drier conditions.

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CHAPTER 6. PAST ENVIRONMENTAL PROXIES FROM THE MIDDLE STONE AGE AT SIBUDU CAVE

Title: Past environmental proxies from the Middle Stone Age at Sibudu Cave, KwaZulu-Natal, South Africa

Manuscript in preparation.

Background to the paper

In Chapter 5 it was demonstrated that it is possible to obtain an environmental signal from $\delta^{13}C$ values of charcoal. In this chapter the method is applied to archaeological charcoal from selected MSA levels at Sibudu Cave. Charcoal samples from the Howiesons Poort (62ka-65ka), post-Howiesons Poort (~58ka) and the late MSA (~48ka), identified as either *Podocarpus* or *Celtis* were subjected to stable carbon isotope analysis. In addition a subset of *Podocarpus* and *Celtis* charcoal samples were further pre-treated using the acid-alkali-acid (AAA) method and their isotopic values obtained. This was done to exclude the potential effects of post-depositional contamination on the isotopic signature of the samples. The archaeological $\delta^{13}C$ values were compared with $\delta^{13}C$ values from modern *Podocarpus* specimens from two different environments, Seaton Park and the Baviaans Kloof. These two areas provided modern analogues for what past environments may have been like around Sibudu Cave during the Howiesons Poort and post-Howiesons Poort periods.

The isotopic variability seen in the archaeological $\delta^{13}C$ values of the two genera provides a reflection of their past environments. This variability is not a product of the combustion process or as a result of post-depositional contamination. This allowed a reconstruction of the local Sibudu environment during the Howiesons Poort, post-Howiesons Poort and late MSA occupations based on the isotopic data. The Howiesons Poort period was moist and humid and the region was covered in evergreen forest with patches of woodland. During the post-Howiesons Poort occupation there was a shift to drier and cooler conditions than those of the Howiesons Poort and vegetation communities changed from closed forest/woodlands to more open savanna grasslands. The environment underwent another shift during

the late MSA to a mosaic of different vegetation communities, with savanna, grassland and forested woodlands represented. The isotope data from the archaeological charcoal provides evidence for these environmental shifts and are supported by the additional proxy environmental evidence from Sibudu Cave.

Past environmental proxies from the Middle Stone Age at Sibudu Cave, KwaZulu-Natal, South Africa

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6.1. Abstract

Stable carbon isotope ratios of archaeological *Podocarpus* (Yellowwood) and *Celtis* (Stinkwood) charcoal, from the Howiesons Poort (62ka-65ka), post-Howiesons Poort (~58ka) and late Middle Stone Age (~48ka) layers of Sibudu Cave, KwaZulu-Natal, South Africa, indicate environmental shifts. During the Howiesons Poort, the local environment is thickly forested and moist and more humid than present. The environment changes during the post-Howiesons Poort occupation into the late MSA occupation, becoming drier and colder than present with vegetation shifting to open savanna grassland or woodlands. It is demonstrated that the isotopic observations reflect past environmental conditions and are not an artefact of the charcoal formation process or post-depositional contamination. Evidence for a changing environment during these occupations is supported by environmental proxy data derived from sediments, botanical collections and faunal assemblages from Sibudu. The results suggest that both the absolute value and the variance of the charcoal isotope values contain important palaeoenvironmental information.

Key words: Sibudu Cave, Middle Stone Age, Howiesons Poort, stable carbon isotopes, charcoal, environmental proxies.

6.2. Introduction

The use of fire by people who lived in the past has ensured regular accumulations of charcoal in archaeological sites. Good organic preservation and well dated sequences make such sites important archives of palaeoenvironmental data. Stable isotopic data ($\delta^{13}C$) obtained from charred organic matter have been used to track changes in

palaeovegetation (e.g. Bechtel *et al*., 2002, 2003; Behling and da Costa, 2001; Biedenbender *et al*., 2004), for carbon cycling (e.g. Gleixner *et al*., 2001; Schmidt, M.W.I., *et al*., 2002) and to determine changes in atmospheric oxygen and CO_2 concentrations (Cope and Chaloner, 1980; Jones and Chaloner, 1991; Scott and Glasspool, 2006). Most of the aforementioned studies have concentrated on palaeoenvironmental changes in deep geological time utilising carbon isotopic data derived from fossil plants and coal. To date there have been relatively few studies focusing on wood charcoal. The majority of these studies have linked variation in the $\delta^{13}C$ values of archaeological charcoal with changes in precipitation (Aguilera *et al*., 2009; February, 1992, 1994a, b; February and Van der Merwe, 1992; Ferrio *et al*., 2006; Fiorentino *et al*., 2008), with varying degrees of success.

It is assumed that the relative changes in $\delta^{13}C$ values from natural and anthropogenic charcoal directly represent the $\delta^{13}C$ values of the source material (e.g. Briffa *et al*., 2004; February and van der Merwe, 1992; Leavitt, 2002; McCarroll and Loader, 2004; Van Bergen and Poole, 2000). This implies that an insignificant or constant fractionation of carbon isotopes occurs during combustion (Turney *et al*., 2006). However, potentially significant changes in carbon isotope composition can occur during combustion resulting from the plant organ combusted, its chemical composition, tissue ratios, surface area, the availability of oxygen and the combustion temperature (Baldock and Smernik, 2002; Ballentine *et al*., 1998; Bird and Gröcke, 1997; Cahier *et al*., 1985; Hall *et al*., 2008; Krull *et al*., 2003; Leavitt *et al*., 1982; Poole *et al*., 2002; Schleser *et al*., 1999; Steinbeiss *et al*., 2006; Turekian *et al*., 1998; Turney *et al*., 2006). This can result in either isotopically depleted or enriched $\delta^{13}C$ values relative to the pre-combusted value. These changes in stable carbon isotope composition during carbonization are still not fully understood and recent studies have yielded conflicting results (Krull *et al*., 2003; Leavitt *et al*., 1982; Steinbeiss *et al*., 2006; Turekian *et al*., 1998). This may be due to sample pre-preparation and combustion methods employed and laboratory experiments that may not produce results completely comparable to results derived from material produced in natural fires. Furthermore, although charcoal is considered to be biologically and chemically inert after formation and undergoes an extremely slow process of degradation (Bird and Gröcke, 1997; Cohen-Ofri *et al*., 2006; Forbes *et al*., 2006; Skjemstad *et al*.,

1996), the effects of post-depositional contamination (e.g. humic and fluvic contaminants) on the isotopic composition of charcoal needs to be considered.

Charcoal isotopic data derived from the archaeological site of Sibudu Cave, KwaZulu-Natal, South Africa show interesting trends through time (Hall *et al*., 2008). Preliminary interpretation of this data suggested a palaeoenvironmental driver, and did not account for the possibility that the carbonization processes or post-depositional effects might be the underlying cause. Here, we explore the extent to which post-depositional effects may have affected the isotopic composition of the charcoal and examine how the combined ancient environmental proxies from Sibudu support each other and compare with modern environmental indicators.

6.3. Current environmental research at Sibudu Cave

6.3.1. Background to the site

Sibudu Cave is an important Middle Stone Age (MSA) archaeological site in southern Africa (Fig. 6.1, Vol.II, pg.23) because of the excellent preservation and the long depositional sequence. The site has a long sequence of time-related informally and formally named lithic assemblages (Wadley and Jacobs, 2004, 2006). From oldest to most recent, the assemblages represent a Pre-Still Bay (layers BS to LBG), Still Bay (layers RGS, RGS2), Howiesons Poort (layers PGS to DRG), post-Howiesons Poort (layers Br under YA2 to BSp), late MSA (layers YSp to PB) and a final MSA (layers Mou to Co) (Fig. 6.2, Vol.II, pg.24). The post-Howiesons Poort, late MSA and final MSA are informal assemblages based on available ages and lithic characteristics (Wadley and Jacobs, 2006). The chronological sequence was created by using Optically Stimulated Luminescence (OSL) techniques on single quartz grains, yielding 21 dates for the site (Jacobs, 2004; Jacobs *et al*., 2008a, b; Jacobs and Roberts, 2008; Wadley and Jacobs, 2004, 2006). Fourteen OSL dates were obtained for the post-Howiesons Poort, late MSA and final MSA layers, with weighted mean ages of 57.5 ± 1.4ka, 47.6 ± 1.2ka and 35.1 ± 1.4ka respectively. Seven OSL dates for the Howiesons Poort, Still Bay and Pre-Still Bay industries were derived from additional sediment samples. Based on these results, the Howiesons Poort occupation falls between 65-62ka, the Still Bay dates to 70ka and the Pre-Still Bay to between

75-72ka (Jacobs *et al*., 2008b; Jacobs and Roberts, 2008). Hiatuses of 9.8 ± 1.3ka and 12.6 ± 2.1ka occur between the three more recent age clusters, so the site was not continually occupied (Jacobs *et al*., 2008a; Wadley and Jacobs 2006). Due to the hiatuses it is not possible to provide a complete sequence of environmental change, but detailed environmental data are available for the occupational pulses. There are no geological hiatuses indicated and all sediment build-up within the site is derived from anthropological debris (Goldberg *et al.*, 2009; Pickering, 2006).

Multi-disciplinary research has enabled reconstruction of palaeoenvironmental conditions through the Howiesons Poort and post-Howiesons Poort periods (Table 6.1, Vol.II, pg.56). Studies include: geomagnetic susceptibility (Herries, 2006), mineralogy (Schiegl *et al*., 2004; Schiegl and Conard, 2006), geology (Pickering, 2006), charcoal (Allott, 2004, 2005, 2006), carbonised seeds (Sievers, 2006; Sievers and Wadley, 2008; Wadley, 2004, 2006), pollen (Renaut and Bamford, 2006) and macro- and micro-faunal remains (Cain, 2004, 2005, 2006; Clark and Plug, 2008; Glenny, 2006; Plug, 2004, 2006; Wadley *et al.*, 2008; Wells, 2006).

6.3.2. Proxy evidence for environmental change at Sibudu Cave

Sibudu Cave is located in a mosaic environment that has probably persisted throughout the MSA, and care is needed when drawing conclusions about past climatic and environmental conditions. Recent micromorphological analysis of sediments from the site indicates that they are predominantly anthropogenic in origin (Goldberg *et al.*, 2009). This highly anthropogenic nature of the deposits may make the interpretation of proxy environmental evidence, such as geomagnetic susceptibility, more complex than the authors have recognised in their publications. The available non-isotopic proxy environmental evidence from the site is described briefly, and a summary is presented in Table 6.1 (Vol.II, pg.56). Two key factors, aspect and the Tongati River (Fig. 6.1, Vol.II, pg.23), affect the range of faunal and botanical diversity seen throughout the MSA occupations. The ever-present river (at least during the periods that Sibudu was occupied) would have allowed for the localised occurrence of certain species that would not have otherwise have survived in drier areas surrounding the site. The following vegetation types - forest, savanna,

woodland and grassland - require definition because they are repeatedly used in this study and in other publications pertaining to Sibudu Cave. Forests comprise predominantly of evergreen tree species, typically 8 to 30m in height, a variety of climbing species and epiphytes and low growing shrubs and ferns on the forest floor, and canopy cover is continuous (Rutherford and Westfall, 1986). Savanna is a mixed tropical and subtropical vegetation type comprising a predominantly deciduous woody species with a grass component (Scholes, 1997); it can be divided into sub-types, dependent on the distribution of woody species. When woody species provide up to 75% canopy cover, the form of savanna may be referred to as woodland (Rutherford and Westfall, 1986). Near Sibudu, the north-eastern slopes and the plateau are more exposed to sunlight than the southern slopes and are hence warmer and, even today, these areas support deciduous woodland, savanna and grassland communities (Fig. 6.1, Vol.II, pg.24). The cooler, sheltered south-western cliffs, where the shelter is situated, support evergreen forest species and would have provided refugia for forest species during drier-than-present periods (Fig. 6.1, Vol.II, pg.24). Nevertheless some of the past palaeoenvironmental conditions that have been observed (for example at ~58ka) are sufficiently extreme that they could not have arisen under current conditions. Reconstructions of local vegetation communities for the Howiesons Poort, post-Howiesons Poort and late MSA periods, based on the available botanical evidence (seeds, pollen and charcoal), are presented in Figures 6.3a-d (Vol.II, pgs. 25-28).

6.3.3. Howiesons Poort layers (65ka-62ka)

The occurrence of *Schoenoplectus* spp. (sedge) seeds in all the MSA layers suggests the presence of perennial water in the Tongati River as this sedge requires year-round standing water (Sievers, 2006). Aquatic reptiles and mammals, fresh water mollusc species and a variety of waterfowl species also attest to a permanent water source close to the shelter (Plug, 2006). A number of evergreen forest taxa have been identified in the charcoal assemblage, including *Podocarpus* which is the dominant genus (Allott, 2005, 2006). Other taxa such as *Kirkia* spp. (not present in the area today), which is a dry-adapted genus, were also identified, indicating the presence in the vicinity of Sibudu of plant communities requiring drier-than-present conditions. In

summary, the macrobotanical evidence from the Howiesons Poort layers indicates a predominantly forested environment, but with neighbouring patches of woodland/savanna (Fig. 6.3a, Vol.II, pg.25) of a kind for which there is no modern analogue. This interpretation is supported by evidence from the faunal assemblage.

Forest-dwelling mammal species such as *Cephalophus natalensis* (red duiker), *Cephalophus monticola* (blue duiker), *Potamochoerus porcus* (bushpig), *Tragelaphus scriptus* (bushbuck) and *Chlorocebus aethiops* (vervet monkey) are present in the faunal assemblage (Clark and Plug, 2008). These species are characteristic of modern *Podocarpus* spp. forest (Cooper, 1985), although they may occur in other types of wooded environments. The faunal assemblage at this time mostly comprises (91.4 %) species inhabiting closed or semi-closed habitats (Clark and Plug, 2008). Only 8.6% of species that favour open environments, such as *Syncercus caffer* (African buffalo), *Connochaetes taurinus* (blue wildebeest) and *Hippotragus equinus* (roan antelope) are present in the assemblage. Nonetheless, they indicate that there must have been patches of open woodland/savanna communities in the general area and this supports the earlier suggestion that a mosaic of vegetation types probably existed throughout the site occupations. Further evidence for humid, moist, forested environments in the vicinity of the site is provided by the presence of micro-mammalian species, such as the Gambian giant rat (*Cricetomys gambianus*) and Geoffroy's horseshoe bat (*Rhinolephus clivosus*) (Glenny, 2006; Skinner and Chimimba, 2005). The Gambian giant rat also implies cooler than present conditions because it cannot tolerate mean summer temperatures above 34°C. Numerous calcite crystals within the deposits indicate more humid conditions during this time than later in the sequence where gypsum predominates (Pickering, 2006; Schiegl and Conard, 2006; Wadley, 2006). Both calcite and gypsum require some moisture to form, but gypsum is more soluble in water than calcite and is therefore a better indicator than calcite of locally arid conditions.

The evidence from the Howiesons Port layers shows that the environment around Sibudu Cave was cooler than present, which possibly allowed for more effective moisture retention that supported a substantial evergreen forest, probably on the south-western slopes, with patches of open woodland/savanna within exploitation range of the inhabitants of Sibudu (Fig. 6.3a, Vol.II, pg.25).

6.3.4. Post-Howiesons Poort layers (~58ka)

The changing composition of plant and animal communities in the post-Howiesons Poort layers provides evidence for oscillating climatic conditions from the coldest period at the end of MIS 4 into MIS 3. Correspondence analysis of the seed and faunal data (Reynolds, 2006) from this period suggests that these changes were time-related fluctuations in response to a changing environment. Magnetic susceptibility readings imply that the coldest conditions in the Sibudu sequence occurred in the earliest of the post-Howiesons Poort layers at the transition between MIS 4 and MIS 3. In more recent layers, magnetic susceptibility results show alternating cooler and warmer conditions with an overall warming trend, although temperatures remained lower than present (Herries, 2006). The deposits are largely due to anthropogenic activities within the site (Pickering, 2006); therefore the magnetic susceptibility data should be interpreted with care. The lower magnetic susceptibility readings may correlate with periods of reduced human activity rather than a change in the environment.

The post-Howiesons Poort seed assemblage comprises a mixture of sedges and woody tree and shrub species (Fig. 6.3b, Vol.II, pg.26). Cyperaceous nutlets (including *Schoenoplectus* spp.) occur in the lower layers indicating the presence of open or semi-permanent water (Sievers, 2006). The majority of identified seeds originate from woody evergreen species, but deciduous species increase in the upper, more recent layers that coincide with the warming trend. Pollen and phytoliths include representatives of grasses, sedges and fern spores (Renaut and Bamford, 2006; Schiegl and Conard, 2006; Schiegl *et al*., 2004). The frequency of grass phytoliths increases in the upper layers and this may reflect an increase in grasslands around Sibudu, but this interpretation must be made cautiously because grass seems to have been used for tinder in some of the fireplaces (Schiegl *et al*., 2004). Woody species from the charcoal assemblage indicate a continuous presence of an evergreen riverine forest component (Allott, 2004, 2005, 2006). *Podocarpus* reappears in the post-Howiesons Poort layer called SPCA where *Acacia* species are absent, following the general trend for the two genera to be mutually exclusive (Wadley, 2006). *Leucosidea sericea*, which can be a pioneer, was identified in the charcoal assemblage of two post-Howiesons Poort (~58ka) layers (Allott, 2006). This species does not occur near

to the coast under modern environmental conditions; it occupies high altitudes and is often in areas with cold winters with frost (Killick, 1990, Pooley, 2003).

The faunal assemblage of the youngest post-Howiesons Poort layers is markedly different in species composition from the earlier Howiesons Poort layers. Small bovid species decrease through the oldest post-Howiesons Poort layers, until they are almost absent in the youngest post-Howiesons Poort layers where large species dominate the assemblage, including equids and large to very large bovids (Cain, 2006; Plug, 2004). Grazers appear throughout, but they are present in higher frequencies in the upper layers (Cain, 2006; Clark and Plug, 2008; Plug, 2004); suggesting a shift from exploiting fauna in closed forest/woodland communities to those in drier, more open savanna/grassland communities. The Natal multimammate mouse (*Mastomys natalensis*), a pioneer species, is found in layer My (Glenny, 2006). Although the species has a wide habitat tolerance, it is not a forest dwelling species and has a degree of water independence, allowing it to occur in drier areas (De Graaff, 1981; Meester *et al*., 1979; Skinner and Smithers, 1990).

After an initial bitterly cold phase during the post-Howiesons Poort, a warming trend is observed in the combined proxy evidence. In contrast to the afforestation that existed in the Howiesons Poort period, open woodland and grassland communities seem more common by ~58ka. There is an intriguing possibility that the initial post-Howiesons Poort occupations may have taken place during an early phase of succession; this is implied by the presence of the pioneer species *Leucosidea sericea* (shrub) and *Mastomys natalensis* (rodent).

6.3.5. Late MSA layers (~48ka)

Proxy environmental evidence from the late MSA layers suggests an initial period of warming followed by a cooler period. Cyperaceae nutlets (Sievers, 2006) and the presence of hippopotamus (*Hippopotamus amphibious*) (Plug, 2006) attest to the presence of a permanent water source. Seeds from evergreen species persist, but the frequencies of deciduous species increase throughout the late MSA sequence (Sievers, 2006). The charcoal assemblage also reveals an increase in deciduous woody species

(Allott, 2006) and the vegetation profile can be interpreted as deciduous savanna woodland with common taxa including *Acacia* spp., *Albizia* spp. and *Celtis* spp. (Fig. 6.3c, d, Vol.II, pgs.27-28). In layer RSp (Fig. 6.3c, Vol.II, pg.27) there is an unusual combination of *Acacia* and *Podocarpus*. This species combination is not seen in any other MSA layer at Sibudu (Allott, 2006; Wadley, 2006). RSp is the youngest Sibudu layer to contain *Podocarpus*; there is no evidence for its presence in more recent layers (Allott, 2006). The phytolith composition at this time shows an overall decrease in grass and an increase in trees (Schiegl *et al*., 2004) and the general impression is that grassland patches were reduced at the expense of woodland.

The faunal species composition of the late MSA layers suggests a mixed environment. The RSp layer has produced a diverse faunal species profile with savanna, grassland and forest/woodland dwelling species present (Cain, 2006; Plug, 2004; Wells, 2006). The Gambian giant rat (*C. gambianus*) is represented again in layer RSp, indicating the presence of forested areas and rainfall over 800mm per annum (Glenny, 2006; Skinner and Chimimba, 2005). A substantial environmental change prior to or during the formation of RSp at 46.6ka, is suggested by the re-occurrence of the Natal multimammate mouse (*M. natalensis*) (Glenny, 2006). Indirect evidence for a dramatic climate change between the post-Howiesons Poort and late MSA is also suggested by a hiatus of 9.8 ± 1.3ka between these two occupational phases (Jacobs *et al*., 2008a). Environmental conditions were likely unsuitable for the use of the shelter as a permanent dwelling during the hiatus, perhaps because of a particularly arid phase (Jacobs *et al*., 2008a). Magnetic susceptibility readings suggest an initial cooling period in OMOD (Herries, 2006), followed in MOD by a further warming trend.

6.4. Stable carbon isotope analysis of archaeological charcoal from Sibudu

6.4.1. Background

Stable carbon isotope values from wood are sensitive bio-indicators and can elucidate a tree's response to changes in external environmental conditions such as temperature, rainfall, humidity, soil chemistry and changes in local vegetation (Hedges *et al*., 2004). An important question is whether the environmental response is preserved in

charcoal? In keeping with other archaeological research based on charcoal and other burnt plant material, we define charcoal as a charred or carbonised product of wood burning in an oxygen-poor atmosphere (Braadbaart, 2004; Braadbaart *et al*., 2004; Cohen-Ofri *et al*., 2006; Sievers and Wadley, 2008).

Hall *et al*. (2008) showed that the $\delta^{13}C$ values of modern *Podocarpus latifolius* (Real Yellowwood) from a remnant patch of Coastal Forest in Seaton Park, KwaZulu-Natal preserved the equivalent overall response to environmental conditions under different combustion conditions (reducing and oxidising). Annualised $\delta^{13}C$ values of fresh wood and charcoal from trunks and cores from the same specimen were compared with annual rainfall, temperature and humidity records for the region. During periods of below normal mean annual rainfall isotope values became progressively isotopically enriched, whereas they became depleted in periods of higher rainfall. A statistical regression showed that there is a strong negative correlation of isotope values with humidity and temperature and a weaker negative correlation with rainfall (Hall *et al*., 2008). These results indicate that an environmental signal *is* preserved in charcoal under constant combustion conditions (Hall *et al*., 2008), and that, in the case of *Podocarpus*, the signal associated with humidity changes is particularly reliable.

A detailed analysis of the Sibudu charcoal assemblage has been carried out by Allott (2004, 2005, 2006). All charcoal examined by Allott was derived from ashy deposits rather than discrete hearths to avoid sample bias. Charcoal from a discrete hearth may only represent a limited range of species. Through the analysis of sediments (Schiegl *et al*., 2004; Goldberg *et al*., 2009) and burnt bone (Cain, 2005) from the ash deposits, it has been demonstrated that they are clearly hearth-related deposits, possibly representing multiple hearth cleaning episodes. The archaeological charcoal was identified using a range of documented anatomical features visible under an incident light microscope. These key features were compared with identified charcoal samples from modern reference collections to allow identification to family, genus, species or type (Allott, 2006).

6.4.2. Material and methods

One-hundred and twenty-two charcoal samples identified as *Podocarpus* were obtained from the Grey Rocky (GR) and Grey Sand (GS) layers, both of which occur within the Howiesons Poort (62ka-65ka) suite of layers. Seven samples of *Celtis* charcoal were taken from GS. Twenty samples of *Podocarpus* and 12 samples of *Celtis* were taken from Spotty Camel (SPCA, ~58ka), which is within the post-Howiesons Poort suite of layers. It should be noted that two species of *Podocarpus* were tentatively recorded in SPCA, *P. latifolius* and *P. falcatus* (Allott, 2004, 2005, 2006). Fifteen samples of *Podocarpus* were from Red Speckled (RSp) (46.6 ± 1.9ka). Thirty-six samples of *Celtis* were obtained from the Orange Mottled Deposit (OMOD) (48.3 ± 2.0ka). These samples are representative of the late MSA suite of layers.

The archaeological charcoal samples were pre-treated with 1% HCl overnight and washed with distilled water until pH neutral. Aliquots of approximately 0.2mg were combusted on-line in a Thermo Flash Elemental Analyser (1112 series) integrated via a Thermo Finnigan Con-flo III system with a Thermo Delta V Plus Isotope Ratio Mass Spectrometer, housed at the Ecosystem Processes and Dynamics (EPD) laboratory, CSIR, Pretoria. Each sample was run in duplicate. If the precision for each sample pair was unacceptable (0.10‰ variation), an additional sub-sample was run. The average precision for the replicates of the archaeological samples was <0.08‰ (Hall *et al*., 2008).

In order to account for the potential effects of post-depositional contaminants, such as humic substances and/or ground water carbonates, subsets of *Podocarpus* and *Celtis* charcoal from each period were further pre-treated using the acid-alkali-acid (AAA) method. The charcoals were treated with a 0.5% sodium hydroxide (NaOH) solution overnight in a refrigerator, washed with distilled water until pH neutral and then treated again with a 1% HCl solution, washed and dried. These samples were combusted in precisely the same manner as the original samples. The average precision for replicates of the subsets was <0.09‰.

6.4.3. Results

The mean $\delta^{13}C$ values for the archaeological charcoal analyses are presented in Figure 6.4 (Vol.II, pg.29) and summarised in Table 6.2 (Vol.II, pg.58). The data are composed of the original $\delta^{13}C$ values presented in Hall *et al*. (2008) and new $\delta^{13}C$ values for the archaeological charcoal samples pre-treated by the AAA method.

When the $\delta^{13}C$ values of the original charcoal samples are compared with their equivalent AAA $\delta^{13}C$ values, they indicate that there is relatively little post-depositional contamination of the Sibudu charcoal. On average there is 0.2‰ difference between the *Podocarpus* $\delta^{13}C$ AAA and original values and a 0.5‰ difference between the *Celtis* $\delta^{13}C$ AAA and original values. The AAA isotopic values for both *Podocarpus* and *Celtis* are slightly more negative than the original values, with the exception of samples from the late MSA layer RSp (Fig. 6.4, Vol.II, pg.29). The standard deviation, range and variance values from the AAA samples are very similar to those of the original samples (Table 6.2, Vol.II, pg.58). On the basis of these results, the observed isotopic values of the archaeological charcoal from Sibudu can be considered to reflect an environmental signal, rather than post-depositional contamination. It is important to note that the degree of post-depositional contamination must be assessed on a site-by-site basis.

In layer GR (Howiesons Poort) *Podocarpus* $\delta^{13}C$ values have a range of 5.03‰ and a variance of 1.23‰. Samples from Howiesons Poort layer GS have a range of 2.17‰ and variance of 0.35‰. The *Podocarpus* isotope values are dependent on environmental variables. Thus the variability seen in the $\delta^{13}C$ values of *Podocarpus* is due to variations in environmental conditions at the time. The physiological response of the tree is, however, adaptive and the tree cannot respond to all states of the environment. When environmental conditions, such as too little or too much moisture availability can no longer be met by the adaptive response of the tree becomes limited. *Podocarpus* populations in the region were not restricted in their adaptive response by prevailing environmental conditions during the Howiesons Poort. In the post-Howiesons Poort SPCA layer *Podocarpus* $\delta^{13}C$ values are less negative than the Howiesons Poort values, with a very narrow range of 0.5‰ and the variance is very low (0.02‰). This suggests that the local distribution of *Podocarpus* was highly

restricted during the post-Howiesons Poort and that it existed at the limit of its adaptive responses to the local environment. In the late MSA (layer RSp); *Podocarpus* $\delta^{13}C$ values are again more negative than the post-Howiesons Poort values, with a range of 1.3‰ and variance of 0.18‰. This suggests that a *Podocarpus* population utilising a wider range of adaptive responses to local environmental conditions was again established, but that the range of responses was not as varied as that seen during the Howiesons Poort. The charcoal $\delta^{13}C$ values from the Howiesons Poort and late MSA layers fall within the range of modern *Podocarpus* charcoal values (-28.07‰ to –23.42‰) for trees from Seaton Park, KwaZulu-Natal. In contrast the $\delta^{13}C$ values for the post-Howiesons Poort charcoals from layer SPCA fall within the range of modern values (-25.32‰ to -20.07‰) of a *Podocarpus* specimen from a refuge forest community from the Baviaans Kloof, Eastern Cape where *Podocarpus* is restricted to river margins due to lower moisture availability.

The mean absolute $\delta^{13}C$ values for *Celtis* charcoal from contemporary MSA layers show a similar isotopic distribution pattern to that of *Podocarpus* (Fig. 6.4, Vol.II. pg.29). *Celtis* $\delta^{13}C$ values are more negative during the Howiesons Poort and late MSA and more positive during the post-Howiesons Poort. However when the range and variance of the *Celtis* data are considered, these show very low values during the Howiesons Port compared to those of the *Podocarpus* samples, suggesting a limited adaptive response to the local environmental conditions. During the post-Howiesons Poort and late MSA the *Celtis* data have higher range and variance than *Podocarpus*. This suggests that both genera could have been affected in different ways by a change in the environment. In dry areas/conditions carbon isotope compositions are affected by relative humidity and soil water status. Under conditions of high moisture availability, sunlight and temperature are responsible for changes in isotopic composition (McCarroll and Loader, 2004). The more positive $\delta^{13}C$ values for charcoal samples therefore suggest dry, warm conditions during the post-Howiesons Poort to which *Celtis* is better adapted than *Podocarpus*. The more negative values seen in the Howiesons Poort are more indicative of higher moisture availability and cooler conditions favourable for *Podocarpus*.

6.5. Discussion

Podocarpus is considered to be a dominant climax genus in many of the tropical and subtropical forest types in South Africa (Acocks, 1988; Killick, 1990; Palmer and Pitman, 1972; Pooley, 2003; Schmidt, E., *et al*., 2002) and adaptability to different environmental conditions (which may affect the $\delta^{13}C$ signature) allows the genus to establish and maintain its presence. The genus requires high moisture availability and it grows from sea-level to 2150m. The presence of a *Podocarpus* community implies a forested environment with effective precipitation, while an absence or limited occurrence of the genus implies reduced forest and drier conditions. *Celtis* species are more widespread and are adaptable to a larger range of environments than *Podocarpus*, particularly drier extremes (Acocks, 1998; Coates-Palgrave, 2003; Pooley, 2003; Van Wyk and Van Wyk, 1997).

The $\delta^{13}C$ values of a particular tree or woody shrub vary according to local site specific factors, such as aspect, soil type and proximity to water sources. The isotopic values of *Podocarpus* reflect a physiological response to the environmental conditions: when water stress increases isotopic values become less negative. Where the isotopic values oscillate between more and less negative values it implies that the tree has a physiological response to the implicated dry and moist environmental conditions. This indicates that both the absolute value of the isotopes and their variance have environmental implications. A low variance suggests that the trees are using a limited range of their physiological adaptive responses to cope with prevailing environmental conditions. This implies that environmental conditions changed sufficiently that the trees could only exist at the outer limit of their range.

Proxy environmental data from Sibudu Cave, summarised earlier in this paper, imply that during the Howiesons Poort (62ka-65ka), environmental conditions were cool and humid favouring the presence of evergreen forests (with *Podocarpus*) on the southern slopes surrounding the site (Fig 6.3a, Vol.II, pg.25). Here, the mean $\delta^{13}C$ values for *Podocarpus* are the most negative (GR = -24.62‰ and GS = -24.47‰), suggesting that there was high moisture availability at this time. The $\delta^{13}C$ values have a high sample variance and wide range indicating that the genus was growing in a variety of habitats. Although *Celtis* is also present, its $\delta^{13}C$ values have a low sample

variance and range, suggesting a limited adaptive response to the open, dry north-eastern hillside opposite the shelter.

The $\delta^{13}C$ values for *Podocarpus* charcoal from the post-Howiesons Poort (~58ka) have very low sample variance and range, and they are less negative values than those of the Howiesons Poort sample. The low variance suggests that *Podocarpus* was exceedingly constrained by the prevailing environmental conditions and was limited in its adaptive responses. The less negative absolute values suggest that the genus was responding to more arid conditions. *Celtis* $\delta^{13}C$ values from the same period have a higher variance and range, suggesting a wider distribution and adaptive response favouring the more arid conditions at this time. Other proxy data from the post-Howiesons Poort imply that forested areas were greatly reduced and conditions were drier and with less moisture availability than the Howiesons Poort.

During the late MSA (~48ka) occupation, changes in environmental conditions resulted in a mosaic vegetation around Sibudu Cave, comprising a mixture of grassland, savanna, woodland, evergreen and riverine forest communities (Figs. 6.3c, d, Vol.II, pgs.27-28). The sheltered south-western areas would have favoured evergreen forest species, such as *Podocarpus*. Stable carbon isotope values of *Podocarpus* charcoal from the late MSA (RSp layer) show a higher variance and range than those from the post-Howiesons Poort, but much lower than those of the Howiesons Poort charcoal. This suggests that during the late MSA only microhabitats were suitable for *Podocarpus*. Mean $\delta^{13}C$ values for *Podocarpus* and *Celtis* are more negative again in the late MSA, suggesting that available moisture levels were higher than those during the post-Howiesons Poort. No *Celtis* charcoal was available from RSp, but $\delta^{13}C$ values for *Celtis* charcoal from OMOD (also with an age of ~48ka) suggest that the genus was able to establish itself in a similar fashion as seen at ~58ka (Fig. 6.3d, Vol.II, pg.28). The sample variance and range for the OMOD data are similar to those of the SPCA data suggesting that environmental conditions during both periods were suitable for *Celtis*.

6.6. Conclusion

Stable carbon isotope values from archaeological charcoal samples of two genera, *Podocarpus* and *Celtis*, from MSA layers at Sibudu Cave, have provided proxy environmental data that support those from other archaeological sources. The consistencies in the environmental interpretation of the $\delta^{13}C$ charcoal data and the other palaeoenvironmental proxies from Sibudu Cave through time, implies that the patterns in the isotopic data are not an artefact of charcoal formation at different burning temperatures. Nor are they an artefact of post-depositional contamination.

Carbon isotope values were more negative during the Howiesons Poort than the post-Howiesons Poort, when drier conditions are implied. Isotope values became more negative again during the late MSA occupation suggesting greater moisture availability once more. Stable carbon isotope values from *Podocarpus* charcoals suggest that the genus was widespread during the Howiesons Poort when evergreen forests were more prominent than later, at least in the immediate vicinity of Sibudu, but during the post-Howiesons Poort these forests were reduced to sheltered refuge areas and the genus showed a very limited adaptive response. The *Celtis* isotopic data show a similar trend with $\delta^{13}C$ values in the forested Howiesons Poort period indicating a limited adaptive response. During the post-Howiesons Poort and late MSA, the genus shows a wider range of adaptive strategies suited for woodland and savanna environments.

Stable carbon isotope data from archaeological charcoal have the potential to provide a useful source of proxy evidence for localised environmental changes. It is, however, necessary to account for potential sources, such post-depositional contamination of charcoal, which may adversely affect observed isotopic signals. This needs to be carried out on a site by site basis. In the case of Sibudu Cave, a suite of environmental proxy evidence from isotopic, botanical, faunal and sedimentological data provides a detailed picture of how the local environment changed between the Howiesons Poort, post-Howiesons Poort and late MSA occupations.

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CHAPTER 7. ECOSYSTEM CHANGE DURING MIS 4 AND EARLY MIS 3: EVIDENCE FROM MIDDLE STONE AGE SITES IN SOUTH AFRICA

Paper 4: Ecosystem change during MIS 4 and early MIS 3: Evidence from Middle Stone Age sites in South Africa

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Background to the paper

In this paper we provide a link between global climatic changes and a local manifestation of such change from the MSA layers of Sibudu Cave. Climatic records from a number of sources, both in the Northern and Southern Hemispheres indicate that around 60ka there was a significant change in global climate patterns, with conditions becoming cooler and drier than present. This change had an effect on the local climate around the site of Sibudu. Evidence for this global change is derived from a number of sources, including the Vostok Ice Core, the Greenland Ice Sheet Project (GISP, GISP2) and from Hulu Cave in China. At around 60 ka there was a significant Heinrich Event (HE6), a period of extensive ice-rafting, which affected climatic patterns. This event appears to have been locally manifested at Sibudu Cave where the environment around the site underwent a significant change. Evidence for this is seen in changes in the local fauna and flora recorded from MSA layers at Sibudu. Stable carbon isotope data from archaeological charcoal from this time period indicates a retreat of the forest community and an increase in more open savanna grassland around the site.

Ecosystem change during MIS 4 and early MIS 3: Evidence from Middle Stone Age sites in South Africa

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7.1. Abstract

Several Middle Stone Age (MSA) site in southern Africa present evidence of environmental changes during Marine Isotope Stages (MIS) 4 and 3 between 70ka and 50ka. Of these, Sibudu Cave, KwaZulu-Natal, has yielded a detailed record of how global-scale climate change events manifest locally. During MIS 4 (70ka to 60ka) conditions were similar to those during the Last Glacial Maximum. During the transition between MIS 4 and MIS 3 at around 60ka the Sibudu environment changed from a predominantly forested community to more open grass/woodland mosaic. Other MSA sites from across South Africa provide complementary palaeoenvironmental proxy data but imprecise dating presents a cross-correlation challenge. Archaeological sites on the western portion of South Africa appear to have been abandoned earlier and for longer than sites in the East, most likely as a result of adverse climatic conditions. Regional scale climate events in southern Africa are driven by ocean/atmosphere interactions, and at this time weakening of the palaeo-Agulhas Current and an eastward shift of the Agulhas Retroflection resulted in lower sea surface temperatures and a corresponding decrease in humidity and rainfall.

Key words: Middle Stone Age, palaeoenvironmental proxy data, MIS 4, environmental change

7.2. Introduction

The planet is currently warming (Trenberth *et al*., 2007) and climate change modelling is the means by which the future impact of this warming is determined. The high level of complexity in ecosystems reduces the skill of models to forecast the future impact of these changes. An alternative approach is to examine past records of

global climates where the associated ecosystem responses are documented. Evidence for past global warming change is derived from isotope analysis of marine cores and polar ice cores. The most recent of these global-scale events took place at the end of the last glaciation commencing approximately 20ka ago and is known as Marine Isotope Stage 2 (MIS 2). The penultimate warming was between MIS 4 (70ka to 60ka) and early MIS 3 (60ka to 50ka), and is the subject of this analysis. While the warming is known to have taken place, little evidence has been presented for the associated terrestrial ecosystem response to this change.

Through multi-disciplinary analysis of cultural, biological and geological material from middle and lower latitude terrestrial sites it is possible to reconstruct local environmental conditions through time. Eight South African Middle Stone Age (MSA) archaeological sites (Fig. 7.1, Vol.II, pg.30) provide environmental evidence for MIS 4/early MIS 3. These are Sibudu Cave, Border Cave, Rose Cottage Cave, Klasies River Mouth, Boomplaas Cave, Blombos Cave, Diepkloof Rockshelter and Wonderwerk Cave. Between ~75ka-55ka there was a fundamental change in technological and cultural features compared with earlier and later MSA assemblages (Mellars, 2006; Mitchell, 2002, pp.71-106). The stone tool industries of this time include the distinctive Still Bay and Howiesons Poort assemblages. Although the Still Bay has ages of ~75ka-70ka (Jacobs *et al.*, 2008a, b; Tribolo *et al*., 2005) and the Howiesons Poort dates to between 65ka-60ka (Jacobs *et al.*, 2008a, b; Tribolo *et al.*, 2005) depending on the site and dating methods, the Still Bay is the older of the assemblages and always underlies the Howiesons Poort. There is associated evidence for modern human behaviour such as the use of composite stone tools, sophisticated hunting techniques and symbolic expressions (Mellars, 2006; Mitchell, 2002). The appearance of the Still Bay and Howiesons Poort in various MSA sites across South Africa is thought to be a response to sharply oscillating climatic conditions during MIS 4 and early MIS 3 (Mellars, 2006, Mitchell, 2002).

The ecosystem responses that took place at the archaeological sites in southern Africa are indicated by several lines of evidence including the macro-faunal, micro-faunal, botanical and sedimentological evidence that are preserved. It is possible that the environmental proxies themselves may be biased. Anthropogenic influences (e.g. choice of firewood), excavation, sampling methods and the differential preservation

of material may complicate the situation (Allott, 2005). Nevertheless where distinctive environmental shifts are shown to occur, they should have regional manifestations, and they should be recorded in the other sites.

In this analysis the local and regional records are compared with palaeoclimatic data from a number of sites around the world to obtain a global perspective of climate change at this time. Lake sediments, speleothems, aeolian deposits and archaeological sites have become a source of palaeoenvironmental proxies at a local terrestrial scale. This paper compares records of climate change for the period 70ka to 50ka (MIS 4 and early MIS 3) from a number of ocean cores as well as archaeological, geological and other types of research sites from southern Africa.

7.3. Sibudu Cave

Sibudu Cave, located in KwaZulu-Natal, was the most recent of the MSA sites to be excavated in southern Africa, and has the advantages of being well-dated and the subject of multi-disciplinary studies. It has an MSA cultural sequence that contains pre-Still Bay, Still Bay, Howiesons Poort, post-Howiesons Poort, late and final MSA stone tool assemblages (Cochrane, 2006; Delagnes *et al.*, 2006; Villa *et al.*, 2005; Villa and Lenoir, 2006; Wadley, 2005, 2007, 2008). Optically Stimulated Luminescence (OSL) ages for 14 sediment samples (Table 7.1, Vol.II, pg.60) from the three youngest lithic phases are available (Jacobs, 2004; Jacobs *et al.*, 2008a, b; Wadley and Jacobs, 2004, 2006); these have weighted mean ages of 57.5 ± 1.4ka (post-Howiesons Poort), 47.6 ± 1.2ka (late MSA) and 35.1 ± 1.4ka (final MSA) (Jacobs *et al*., 2008a). Ages for the Pre-Still Bay, Still Bay and Howiesons Poort layers are based on seven OSL dates derived from additional sediment samples (Jacobs *et al*., 2008a, b; Jacobs and Roberts, 2008). Based on these results, the Howiesons Poort occupation falls between 65ka-62ka, the Still Bay dates to 70ka and the Pre-Still Bay to between 75ka-72ka (Jacobs *et al*., 2008a, b; Jacobs and Roberts, 2008). The three broad age clusters, ~58ka, ~48ka and ~35ka and the Howiesons Poort are distinguished by differences in lithic assemblages, environmental characteristics and long hiatuses of 9.8 ± 1.3ka and 12.6 ± 2.1ka (Jacobs *et al*., 2008a, b; Wadley and Jacobs 2006). A third hiatus occurred after ~35ka occupation, lasting until about 1000 BP.

7.3.1. Sibudu environment during MIS 4

Sedimentological and mineralogical analyses of the Still Bay and Howiesons Poort layers show relatively high percentages of calcite and this suggests higher humidity (Pickering, 2006; Schiegl and Conard, 2006; Wadley, 2006). Magnetic susceptibility data from layers deposited at the end of MIS 4 (~58ka) imply a cold, glacial climate (Herries, 2006). The cold ~58ka layers contain gypsum growth within the sediments. However, the magnetic susceptibility data should be viewed with caution, as sedimentological analysis of the deposits indicate that the majority of the sediment is anthropogenic in origin and this may complicate interpretation (Goldberg *et al.*, 2009; Pickering, 2006).

Tree species richness is well correlated with evapotranspiration across a wide range of ecosystems. Changes in evapotranspiration will thus have an influence on tree species composition and distribution on a localised site level, as well as on a broad community level (Stephenson, 1998). Local levels of moisture availability at a site are dependent on effective evapotranspiration which is not only affected by precipitation, but a number of factors including aspect, slope, temperature, humidity, wind speed and direction, soil moisture content, depth and type and the presence of rivers (McDowell *et al.*, 2008; Verstraeten *et al.* 2008). The carbonised seed assemblage prior to 58ka from Sibudu Cave is predominantly composed of evergreen taxa, implying the presence of closed forested environments (Sievers, 2006; Wadley, 2004). This interpretation is supported by the composition of woody taxa identified in the charcoal assemblage. Taxa such as *Podocarpus*, *Buxus* and *Curtisia*, evergreen forest species are noted. The presence of these species suggests that available moisture was high during this period, but not necessarily higher than present (Allot, 2006). Although the assemblage at this time is dominated by *Podocarpus* species (Allott, 2004, 2005) and the area appears to have been predominantly a forested one, there is evidence for a woodland/savanna community in the vicinity (Allott, 2006). Throughout the MSA occupations at Sibudu there is evidence that a mosaic environment existed around the site, partly due to the location of the site and the continual presence of the Tongati River (Wadley, 2006). Carbonised Cyperaceae

(sedges) are present throughout the MSA sequence. Sedges grow in moist conditions and the occurrence of *Schoenoplectus* spp. seeds indicates open water, demonstrating that the Tongati River, which flows in front of the site, was perennial during site occupations. Carbon isotope analyses of *Podocarpus* and *Celtis* charcoal from Howiesons Poort layers (65ka-62ka) indicate conditions of elevated levels of water availability and humidity (Hall *et al*., 2006, 2008).

The extensive faunal assemblage provides further evidence of environmental change through time in the Sibudu area. The Howiesons Poort faunal assemblage is dominated (91.4%) by small species preferring semi-closed or closed habitats, such as blue duiker, bushbuck, bush pig and vervet monkey (Clark and Plug, 2008). This supports the botanical evidence for the presence of an evergreen forested environment. In addition, a small suite of species (8.6%), including buffalo and blue wildebeest show the occurrence of open savanna/woodland near the site (Clark and Plug, 2008), supporting the charcoal data (Allott, 2006). This provides further evidence for a mosaic of vegetation types in the area. A large variety of aquatic species including mammals, reptiles, water birds, fish, amphibians and molluscs have been identified (Plug, 2004, 2006) and these, together with the presence of *Schoenoplectus* spp. seeds, demonstrate that the Tongati River was perennial, even in the past.

The micromammal species composition provides further evidence for a cooler, humid forested environment. Two key species, *Cricetomys gambianus* (Giant rat) and *Rhinolophus clivosus* (Geoffroy's horseshoe bat) both require humid conditions. In addition *C. gambianus* cannot tolerate high overall temperatures (Glenny, 2006).

7.3.2. Sibudu environment during MIS 3

Palaeoenvironmental evidence from the post-Howiesons Poort layers (58.5 ± 1.4ka) indicates a general trend of oscillating warm/cool phases and drier conditions than seen during the Howiesons Poort. Magnetic susceptibility data suggest an initial (~58ka) very cold environment which became progressively warmer through MIS 4, alternating with brief cool phases (Herries, 2006). Sedimentological and

mineralogical analyses reveal a high proportion of gypsum nodules in many of the layers (Pickering, 2006; Schiegl and Conard, 2006; Wadley, 2006). Such gypsum accumulations may be considered an indicator of arid conditions (Goldberg and MacPhail, 2006).

The vegetation patterns show a reduction of forested areas and an increase in more open woodland and grassland communities, reflecting the previous trend of a mosaic environment around the site. Pollen and phytolith data, although limited, reveal the presence of a grass-dominated community and the presence of savanna taxa such as *Acacia* (Renaut and Bamford, 2006; Schiegl and Conard, 2006; Schiegl *et al*., 2004). The seed assemblage is still dominated by evergreen forest taxa, but it also reveals an increase in the number of deciduous savanna/woodland taxa (Sievers, 2006; Wadley, 2004). The composition of the charcoal assemblage shows a similar trend with the presence of dry-adapted genera such as *Acacia*, *Celtis* and *Ziziphus* and cooler climate indicators such as *Erica* spp. (Allott, 2005, 2006). The presence of riverine forest taxa attests to the continued presence of a mosaic of vegetation communities around the site. A substantial change in the local environment is suggested by the occurrence of a pioneer shrub species, *Leucosidia sericea* (which at present does not occur near the coast) (Allott, 2006).

Micromammal evidence for a significant environmental shift at the same time is derived from the identification of another habitat specific pioneer species, *Mastomys natalensis* (Natal multimammate mouse) which does not inhabit forested areas (Glenny, 2006). Carbon isotope values from *Podocarpus* and *Celtis* charcoal from ~58ka layers are less negative than those from the earlier Howiesons Poort layers, suggesting that both species were responding to more arid conditions at ~58ka (Hall *et al*., 2006, 2008).

The faunal species composition of the ~58ka layers (post-Howiesons Poort) shows a dramatic shift in response to the changing environment. At this time the highest proportion of large grazing species is recorded indicating an open environment with increased grass cover (Wadley *et al*., 2008). Small bovid species are less frequent and larger savanna/woodland species such as giraffe, zebra, blue wildebeest and red hartebeest dominate the assemblage (Cain, 2005, 2006; Clark and Plug, 2008; Plug,

2004; Wadley *et al*., 2008; Wells, 2006). A recent analysis of the fauna (Clark and Plug, 2008) shows that during the youngest post-Howiesons Poort layers there was predominantly open savanna/woodland with large grazers. Prior to this, there was still a riverine forest community, along with the savanna/woodlands. The occurrence of a riverine forest faunal community during the early phase of the post-Howiesons Poort suggests that the transition between forest and grassland during MIS 4 and MIS 3 was gradual, rather than abrupt.

7.4. Archaeological evidence of environmental change from other MSA sites in South Africa

Improvements in OSL, electron spin resonance (ESR) and uranium-series (U-Th) dating should allow correlations between available environmental proxies from Sibudu and other MSA sites. However there are complications. When comparing the chronologies for the distinctive stone tool technical complexes between sites, it is clear that there are some discrepancies. An underlying assumption is that these stone tool assemblages would be ubiquitous and autochthonous, and that the timing of their rise and demise should be well matched. The transition from Still Bay to Howiesons Poort is not synchronous across MSA sites, and the assemblage that precedes the Howiesons Poort is not always designated Still Bay. This may be because the Still Bay is of very short duration and does not always occur in all sequences. However, it is likely that the problem lies with the precision and accuracy of the dating techniques. This is as a result of MSA sites being dated through a variety of methods at a number of dating laboratories, creating an inconsistent array of ages, particularly for the Howiesons Poort and Still Bay Industries (Jacobs and Roberts, 2008; Jacobs *et al.*, 2008b). The dating conundrum complicates attempts to fine-tune palaeoenvironmental evidence across space or through time. In order to resolve this conundrum, Jacobs *et al.* (2008b) dated nine MSA sites using a single laboratory, a single operator and maintaining constant experimental parameters and a common set of procedures using the single quartz grain OSL technique. For consistency in this study, the most recently published luminescence ages are used (where available), but earlier published dates based on other methods have been noted (Table 7.2, Vol.II, pg.61).

A summary of palaeoenvironmental evidence from Sibudu Cave and these sites is presented in Figure 7.2 (Vol.II, pg.31) and Table 7.2 (Vol.II, pg.61). The majority of these sites are located on or near to the coast, particularly in the southern Cape. Exceptions are Rose Cottage Cave, Border Cave and Wonderwerk Cave which are located in the South African interior.

7.4.1. Palaeoenvironmental evidence from archaeological sites during MIS 4

Evidence from Border Cave, northern KwaZulu-Natal, suggests fluctuating environmental conditions between 80ka and 60ka. Proxy data from cave sediments (Butzer *et al*., 1978), microfauna (Avery, 1982, 1992) and macrofauna (Deacon and Lancaster, 1988; Klein, 1977) indicate that conditions were cooler and moister than present. Local vegetation communities comprised extensive *Podocarpus* dominated forest and thick bush towards the end of MIS 4. As with Sibudu Cave there is evidence for a hiatus from about 58ka. Evidence of spring activity and rock spalling from the Howiesons Poort sediments of Rose Cottage Cave, eastern Free State Province, suggest that conditions in this area were also colder and moister than present (Deacon and Lancaster, 1988). Charcoal and pollen analyses indicate that there were complex changes in the vegetation. Prior to the Howiesons Poort occupation the local vegetation comprised riverine and other well-watered communities. Vegetation diversity decreased during the Howiesons Poort suggesting a drying trend (Wadley *et al*., 1992). Wonderwerk Cave, in the Northern Cape Province, has yielded non-archaeological evidence from sedimentary (Beaumont and Vogel, 2006; Butzer, 1984a, b; Butzer *et al*., 1979) and faunal (Avery, 2006; Beaumont, 1990) analyses of the cave deposits. The site was not occupied from 70ka until 12.5ka, but sediment layers were formed by natural processes. These non-archaeological data suggest that prior to 30ka the local environment was drier and colder than present, with grazers present throughout the sequence. This is thought to be due to very low rainfall conditions in the interior of South Africa between MIS 4 and MIS 2, when it has been estimated that rainfall was about 60% lower than present values (Johnson *et al*., 1997).

Klasies River Mouth, a complex of caves and overhangs on the southern Cape coast has provided faunal, botanical and geological evidence from the MSA II, Howiesons Poort and post-Howiesons Poort levels, suggesting a shift from cooler in MSA II to more moderate conditions in the post-Howiesons Poort. Environmental interpretations from this site are complicated by rising and falling sea levels under interstadial and stadial conditions, respectively, and there are some inconsistencies amongst the ages and cultural designations of the various levels, so interpretations should be made with care. The MSA II faunal assemblage is dominated by browsers (86%) indicating a bushy/wooded terrestrial environment. The presence of Antarctic/sub-Antarctic marine mammals suggests a colder marine environment than seen during the Holocene (Deacon and Lancaster, 1988). During the Howiesons Poort there is an increase in grazing species suggesting the presence of grasslands (Deacon, 1989, 1995; Deacon and Lancaster, 1988; Singer and Wymer, 1982) and cooler, possibly drier conditions than recorded for the MSA II levels (Avery, 1992, Klein, 1976, 1983; Thackeray, 1992, Thackeray and Avery, 1990). The $\delta^{18}O$ values of shell samples from MSA I through the Howiesons Poort and MSA III deposits also suggest a cooling trend (Deacon and Lancaster, 1988) through MIS 4. Palaeoenvironmental reconstructions from the Howiesons Poort levels of Boomplaas Cave, southern Cape, are based on faunal assemblages (Avery, 1982; Deacon *et al*., 1984) and charcoal and pollen analyses (Scholtz, 1986). During MIS 4 environmental conditions were extremely harsh, being much colder and drier than present. The site of Blombos Cave on the southern Cape coast has no Howiesons Poort occupation, only evidence of the earlier Still Bay and MSA II phases. The site was sealed by dune sands from ~70ka until 2000BP (Jacobs *et al*., 2006). Proxy data from marine fauna and shellfish assemblages and geological analyses indicate a transition between warm conditions during MIS 5a to colder conditions during MIS 4 (Henshilwood *et al.*, 2001). Faunal (Parkington *et al*., 2005), charcoal (Cartwright and Parkington, 1997) and sedimentological (Butzer, 1979) analyses from Diepkloof rock shelter in the north-western Cape indicate that during the Howiesons Poort occupations the climate was cooler and moister than present. Charcoal assemblages from this period contain afromontagne species such as *Podocarpus* and *Kiggelaria* which require year-round moisture (Cartwright and Parkington, 1997).

7.4.2. Palaeoenvironmental evidence from archaeological sites during early MIS 3

Evidence from sites in the interior of South Africa is presented first again, followed by evidence from the coastal sites. Based on proxy data from Border Cave, the local vegetation shifted from dense woodland communities to more open woodland savanna with variations in the amount of grass versus bush at the beginning of MIS 3 (Avery, 1982; Butzer *et al.*, 1978; Klein, 1977). These shifts are consistent with the Sibudu environmental records. At Rose Cottage Cave the environment appears to have become colder than in MIS4 and there is evidence of more mesic conditions (Wadley *et al.*, 1992). There is a hiatus after the post-Howiesons Poort occupation indicated by a layer of almost culturally sterile orange sand that lasted from ~48ka-~35ka (Harper, 1997). Although no archaeological evidence is available from Wonderwerk Cave during this time, sedimentary evidence (Beaumont and Vogel, 2006; Butzer, 1984a, b) indicates that the environment was generally dry.

At Klasies River Mouth, faunal evidence from post-Howiesons Poort levels shows a further increase in grazing species compared with the Howiesons Poort levels. This indicates a continuation in the shift to more open grasslands and cooler conditions in early MIS 3. After 50ka the sites were sealed by dune sands (Butzer, 1978; Deacon and Lancaster, 1988; Singer and Wymer, 1982). Oxygen isotope data from shell samples from this period confirm the cooling trend (Deacon and Lancaster, 1988). At Boomplaas Cave charcoal studies indicate that during early MIS 3 (60ka-50ka) a cold, very dry harsh climate prevailed, based on the range of woody species reflected (Scholtz, 1986). From ~55ka to 40ka conditions began to ameliorate (Deacon and Lancaster, 1988). In general, evidence from Boomplaas suggests that MIS 3 was cooler and moister than the subsequent Last Glacial Maximum (Deacon *et al*., 1984). A series of occupational hiatuses occurred after the Howiesons Poort (Deacon, 1979). No environmental evidence is available from Blombos Cave because the site was sealed by dune sands during this time (Henshilwood *et al*., 2001). Charcoal data from Diepkloof indicates a change in the selection of firewood species composition at this time suggesting a shift to drier conditions (Cartwright and Parkington, 1997; Parkington *et al*., 2005). This shift to drier conditions is supported by the dominance of larger grazing species in the faunal assemblage (Parkington *et al*., 2005).

During MIS 4 and early MIS 3, climatic conditions were extremely variable and resulted in the majority of the sites being abandoned for prolonged periods when conditions were unsuitable for human occupation (Fig. 7.2, Vol.II, pg.31). Assuming that the landscape was abandoned by people due to environmental conditions, it is interesting that occupation at sites in the eastern summer-rainfall region of South Africa (Sibudu Cave, Border Cave and Rose Cottage) persisted after the western (including the winter-rainfall zone) sites were abandoned. Environmental evidence from the various sites suggests that the eastern regions experienced greater precipitation than the more arid western regions. Towards the end of MIS 3 and through MIS 2 the environment over much of central South Africa was extremely harsh and sites in this area were unoccupied for periods of up to 60 000 years. During MIS 2 western South Africa appears to have been wetter than present conditions (Chase and Meadows, 2007). Climatic conditions in the eastern regions were more favourable for longer periods, but during MIS2, most sites show intermittent occupations or an absence of occupation.

7.5. Other terrestrial records of palaeo-climate change

In South Africa terrestrial palaeoclimatic records from numerous caves, lacustrine, spring, fluvial and coastal systems provide proxy data from different climate zones across the country. Butzer (1984a, b) compiled a series of proxy records based on sedimentological and lithostratigraphic analyses from a number of such sites. During MIS 4 and early MIS 3, the southern Cape region experienced humid to sub-humid conditions, whilst the south-western Cape was semi-arid. The south-central and eastern regions of South Africa were initially cold and humid during MIS 4, becoming warmer and humid during MIS 3. The northern portions of the country were cold and arid during this time. Palynological and sedimentological evidence from Agulhas Plain lunette dune accretion from two pans in the southern Cape region (Fig. 7.1, Vol.II, pg.30) suggest that from the end of MIS 4 and early MIS 3 conditions became slightly drier or similar to present conditions (Carr *et al.*, 2006). Indications of arid and cold conditions in the northern regions are supported by a range of pollen data from the sites of Florisbad, Wonderwerk Cave and Kathu Pan, located in the northern and north-central parts of South Africa (Fig. 7.1, Vol.II, pg.30). The pollen data

suggests that between ~75ka-64ka this area was extremely arid and cold (Van Zinderen Bakker, 1995).

The Pretoria Saltpan (Tswaing Crater), a meteorite impact north-east of Tshwane in the Gauteng Province, provides a rainfall record for the last 200ka (Partridge, 1999; Partridge *et al*., 1997; 1999). The rainfall record was created through a comparison of the crater sediment texture with soil profiles taken from sixteen sites across a rainfall gradient in eastern South Africa. This comparison revealed a strong correlation between soil texture and regional precipitation (r = 0.88) (Partridge *et al.*, 1997). The Tswaing lithological data and the soil-precipitation relationship were used to produce a late Pleistocene record of rainfall. The rainfall data show a decrease between 70 and 60ka associated with decreasing insolation (Fig. 7.3, Vol.II, pg.32), however the interpretations should be made cautiously as relative dating methods were used at the site. The data suggest that the eastern summer rainfall area of South Africa was becoming drier at this time. This would have affected the vegetation communities around Sibudu Cave and Border Cave. The forested environments would have been reduced to river/stream margins and an expansion of woodland and grassland savanna communities would have occurred.

Oxygen isotope time series from U-Th dated speleothems from two sites in South America, Bahia State (NE Brazil) and Botuvera Cave (SE Brazil) provide a record of oscillating temperatures and aridity (Fig. 7.4, Vol.II, pg.33). During MIS 4, conditions were cool and wet followed by warmer and drier environments during early MIS 3 (Cruz *et al*., 2005; Wang *et al*., 2004). The Botuvera speleothem $\delta^{18}O$ record shows a notable spike at ~70ka indicating a very wet period and then an abrupt shift to an arid period, suggesting a shift to glacial conditions in MIS 4. This is in contrast to results from the $\delta^{18}O$ records (Fig. 7.4, Vol.II, pg.33) from the northern hemisphere Hulu Cave speleothems, which show that between 70ka and 55ka conditions were warm and dry before abruptly becoming cool and wet (Wang *et al*. 2001).

The long term variability of East African climates has been reconstructed using a series of drill cores from Lake Malawi and Lake Tanganyika (Cohen *et al*. 2007, Scholz *et al*., 2007). Between 135ka-70ka these regions experienced episodic periods of extremely arid conditions. After 70ka the climate seems less variable and overall

conditions became more humid and general moisture availability increased. This is thought to be due to diminished precessional scale variability (Cohen *et al*., 2007). These post 70ka conditions are similar to those indicated by the Hulu speleothems and suggest that the East African climate was more likely influenced by changes occurring in the northern hemisphere.

7.6. Palaeoenvironmental evidence from southern African deep sea cores

Deep sea cores along the western, southern and eastern coasts of southern Africa provide evidence of the local manifestation of global climatic changes during MIS 4 and early MIS 3. Cores from the Walvis Ridge and Namibian continental slope along the western coast of southern Africa (Fig. 7.1, Vol.II, pg.30) provide a record of climatic variability regulated by shifting climatic fronts during glacial and interglacial periods (Little *et al*., 1997; Pichevin *et al*., 2005; Stuut *et al*., 2002). The $\delta^{18}O$ records of *Globorotalia inflata*, a pelagic foram (Fig. 7.5A, Vol.II, pg.34) and the proportion of aeolian dust (Fig. 7.5B, Vol.II, pg.34) from MD962094 indicate intensified south-east trade winds and enhanced winter rainfall during MIS 4 and relatively arid conditions during MIS 3 (Stuut *et al*., 2002, 2004). Several cores, MD962094, GeoB1706, 1711, and MD962086/87 provide records of variation in upwelling events of the cold Benguela Current that flows northwards along the western coast of southern Africa (Fig. 7.6a, Vol.II, pg.35). Upwelling is controlled by the relative position of the Subtropical Convergence Zone, which affects the heat flux into the southern Atlantic Ocean from rings of warm water spawned from the Agulhas Current Retroflection (Little *et al*., 1997). Geochemical, micropalaeontological and isotope records from GeoB1706 and 1711 show that during MIS 4-3 upwelling of cold nutrient-rich water increased (Little *et al.*, 1997). Weaker trade winds during MIS 3 resulted in warmer water from the Agulhas Current moving into the colder Benguela region (Pichevin *et al*., 2005). South-east trade winds show increased intensity during glacial periods resulting in increased upwelling (Pichevin *et al*., 2005; Stuut *et al*., 2002). Dust grain-size (Fig. 7.5C) data from MD962087 and alkenone-based sea surface temperatures from MD962086/87 (Fig. 7.5D, Vol.II, pg.34) indicate a similar pattern. Evidence for humid conditions during glacial periods and drier conditions during interglacials is derived from OSL dated cores (WC03-1, 2, 5, 10, 11 and 18) taken from aeolian dune sands along the west coast of South Africa (Fig. 7.1, Vol.II,

pg.30) (Chase and Thomas, 2006, 2007). Changes in the sediments were related to variations in moisture, wind strength and sediment supply. There were periods of increased activity/deposition of aeolian sands during MIS 4 associated with increased humidity (Chase and Thomas, 2006, 2007). The study area is within the winter rainfall zone of South Africa. Rainfall in this area is influenced by westerly temperate frontal systems and these are thought to be more vigorous during glacial periods, resulting in wetter conditions (Barrable *et al*., 2002).

Cores PS2487-6 from the Agulhas Retroflection and MD962080 from the Western Agulhas Bank (Fig. 7.1, Vol.II, pg.30) provide records of variation in the frequency and intensity of Agulhas warm water leakages, responses to shifts in the STCZ and global changes (Flores *et al*., 1999, Rau *et al*., 2002). $\delta^{18}O$ and $\delta^{13}C$ records, foraminifera species assemblages and sediment composition and texture show that during glacial periods (MIS 2, 3, 4) there was a northwards displacement of the STCZ and an eastward movement of the Agulhas Retroflection (Fig. 7.6b, Vol.II, pg.36) (Flores *et al*., 1999; Rau *et al*., 2002). Further evidence for changes in ocean current circulation patterns comes from core MD02-2589 on the southern Agulhas Plateau, where isotopic and grain-size data indicate a northward shift of the Antarctic Circumpolar Current (Molyneux *et al*., 2007) which would have had an impact on the Agulhas Current. An eastward shift of the Agulhas Retroflection (Fig. 7.6b, Vol.II, pg.36) would have an impact on environmental conditions along the eastern coast of southern Africa and may be a factor determining the environmental changes seen in the local Sibudu environment at around 60ka. It has been demonstrated that the Agulhas Current has a significant influence on the summer rainfall patterns of the eastern coast at a variety of timescales (Cook *et al*., 2004).

Core RC17-69, off the eastern coast of KwaZulu-Natal (Fig. 7.1, Vol.II, pg.30) was influenced by the warm Agulhas Current. Foraminifera assemblages from this core suggest that during glacials, the Agulhas Current was weakly developed in summer months and may have been replaced by cooler subtropical waters during winter months (Hutson, 1980). During the LGM the current was seasonably variable and heat transport from tropical latitudes was reduced (Prell and Hutson, 1979; Prell *et al*., 1980a, b). The cooling or reduction of the Agulhas Current during MIS 4 (~60ka), coupled with glacial conditions, would have had a significant impact on the

environment of the east coast of southern Africa (Reason and Mulenga, 1999). Oxygen isotope records from core MD 73-025, south of Madagascar, and RC17-69 do indicate a cooler period during MIS 4 (Prell and Hutson, 1979; Shackleton, 1977; Tyson, 1991).

7.7. Discussion and conclusion

Oxygen and deuterium isotope sequences from Greenland (GRIP, N-GRIP, GISP2) and Antarctic (Vostok, EPICA, Byrd) ice cores (Fig. 7.7, Vol.II, pg.37) show that MIS4 was a period during which the earth emerged from near glacial conditions (the ice core data for 65ka are almost analogous to those of 22ka, which is considered to be the height of the last glacial). The problem of mid-latitude ecosystem responses is complicated by differences between the northern and southern hemisphere records, particularly regarding the timing of major events (e.g. Blunier *et al.*, 1998; Blunier and Brook, 2001; Jansen *et al*., 2007; Leuschner and Sirocko, 2000; Petit *et al*., 1999; Schmittner *et al*., 2003). This makes it difficult to determine whether ecosystem changes are responding to Northern or Southern Hemisphere forcing, or whether low- to mid-latitude forcing of climate change took place. Earlier studies (e.g. Blunier *et al*., 1998; Blunier and Brook, 2001, Leuschner and Sirocko, 2000, Petit *et al*., 1999) suggested that the timing of large southern hemisphere climate events leads the northern hemisphere by 1500-3000 years. More recent research suggests that the south leads the north by approximately 400-500 years (Schmittner *et al*., 2003).

Where global climatic changes are synchronous in the northern and southern high latitude records, they should manifest in low latitude regional and local palaeoenvironmental records. In both Greenland and Antarctic ice cores, rapid increases in air temperatures of 5-10°C (Landais *et al*., 2007; Rahmstorf, 2002) are followed by a rapid return to cold (stadial) conditions. Notable cold phases called Dansgaard/Oeschger (DO) events (Dansgaard *et al.*, 1984; Oeschger *et al.*, 1984) occur with 1000, 1450 and 3000 year cyclicities (Leuschner and Sirocko, 2000). Twenty-two DO events have been identified in the Greenland ice cores and nine corresponding DO events have been identified in the Antarctic cores (Bender *et al*., 1994). Antarctic DO events are characterised by slower warming and cooling than Greenland events (Bender *et al*., 1994). Significant DO events are followed by

massive episodic discharges of icebergs from the Laurentide and Scandinavian ice-sheets and are called Heinrich events (Bond *et al*., 1993; Heinrich, 1988; Leuschner and Sirocko, 2000; Rahmstorf, 2002). Heinrich events always occur during cold stadials and are followed by an abrupt shift to warmer climatic conditions (Bond *et al*., 1993; Rahmstorf, 2002). Between ~70ka and 50ka (MIS 4 and 3) Greenland oxygen and deuterium isotope records (Fig. 7.7A and 7.7B, Vol.II, pg.37) , Vol.II, pg.36from the GRIP (Blunier and Brook, 2001; Grootes *et al*., 1993), GISP2 (Blunier and Brook, 2001) and N-GRIP (Landais *et al*., 2007; Jouzel *et al*., 2005, 2007) ice cores, and Antarctic records from the Byrd (Blunier and Brook, 2001), EPICA (Jouzel *et al*., 2005) and Vostok (Petit *et al*., 1999) ice cores (Fig. 7.7C, 7.7D and 7.7E, Vol.II, pg.37) show that DO events 20-17 and H6 are documented in both the Northern Hemisphere and Southern Hemisphere. The isotopic excursions are not as great in Antarctica and the changes are not as abrupt as those in Greenland (Bender *et al*., 1994). This can be clearly seen during H6, between DO 18 and DO 17 in Antarctica where $\delta^{18}O$ and δD values gradually become less negative indicating a slower warming trend than that seen in the Greenland records. The changes should therefore have an environmental impact at low latitudes.

Linking the terrestrial ecosystem and environmental proxies to climate change proxies requires consideration of the Earth System, and in particular the role of ocean currents in heat distribution. The difference in the rate of change in the Northern and Southern Hemispheres is possibly due to changes in the global thermo-haline circulation (Schmittner *et al*., 2003). A change in the northward circulation of warmer water from the southern oceans (reduction of the warm Agulhas Current eddies) into the Atlantic Ocean would result in a cooling of the northern latitudes and warming in the southern latitudes (Blunier *et al*., 1998; Blunier and Brook, 2001; Rahmstorf, 2002; Schmittner *et al*., 2003; Stocker, 2000, 2002). The Agulhas Current plays a significant role in determining the weather patterns over southern Africa, and hence DO events should be recognisable in the palaeoenvironmental records of the region. However the correlation may not be as simple as a teleconnection. Palaeoclimatic data from core MD97-2120, east of New Zealand, suggests that southern hemisphere mid and low latitude climates were more variable than can be inferred from the Antarctic ice core data (Pahnke *et al*., 2003; Pahnke and Zahn, 2005). This highlights the need to

examine a range of proxy data sets derived from both marine and terrestrial sites to improve understanding of regional and local climatic variability through time.

Focussing on the Southern Hemisphere, an important observation in the Antarctica data sets is the similarity between the climatic conditions during MIS 4 and MIS 2, the Last Glacial Maximum (LGM). This is not limited to the $\delta^{18}O$ and δD records. The dust, Fe, Ca and other chemical flux records from the Vostok and EPICA ice cores (Fig. 7.8, Vol.II, pg.38) are proxies for sea ice extent (sodium (Na) flux); marine biological productivity (sulphate (SO_4) flux); aridity of surrounding continents (Iron (Fe), calcium (Ca), dust, methane (CH_4)); aerosol fluxes of marine, volcanic, terrestrial, cosmogenic and anthropogenic origin and direct records of changes in atmospheric gas (CO_2) composition (Petit *et al.*, 1999; Wolff *et al*., 2006). The data all suggest that during MIS 4 conditions were as severe as those during the cold and dry MIS 2, but not as prolonged. By gathering more evidence for the environmental manifestations this may contribute to a better understanding of why a shift to interglacial conditions occurred at the end of MIS 2, but did not occur at the end of MIS 4 despite the apparent similarity in precursive conditions.

The local manifestation of the global-scale climate events has been reviewed in this paper. Sibudu Cave has yielded the most comprehensive record for the region. Faunal and botanical assemblages, cave sediments, magnetic susceptibility of sediments, geology and carbon isotope analysis of charcoal show that at the end of MIS 4 the environment around Sibudu Cave was humid and cooler than present, supporting a substantial evergreen forest with patches of drier, open woodland/savanna (Table 7.2, Vol.II, pg.61). The shifts seen in the plant and animal communities preserved in the ~58ka layers provide evidence for oscillating climatic conditions into MIS 3. Evidence from more recent layers implies alternating cooler and warmer conditions with an overall warming trend, although temperatures remained lower than present (Herries, 2006). The forested area that existed in the pre-60ka period may have been reduced by ~58ka, allowing more open woodland and grassland communities to develop during the cooler and drier phase. During the warmer phases of MIS 3, grasslands decreased and woodland savanna predominated. Indirect evidence for a dramatic climate change between the ~58ka and ~48 ka occupations is suggested by a hiatus of 9.8 ± 1.3ka between these two occupational phases (Jacobs *et al*., 2008a, b;

Jacobs and Roberts, 2008). This hiatus coincides with a period of colluviation between 56ka-52 ka, an indication of arid conditions or transitional climates with reduced vegetation cover, recorded from a series of well-dated stratigraphic sequences from erosion gullies in KwaZulu-Natal (Botha, 1996, Botha and Partridge, 2000, Botha *et al*., 1992, Clarke *et al*., 2003, Wintle *et al*., 1995). Environmental conditions were likely unsuitable for the use of the shelter as a permanent dwelling during hiatus periods, perhaps because of a particularly arid phase (Jacobs *et al*., 2008a, b).

The predominant forest type in KwaZulu-Natal is classified as part of the Indian Ocean coastal belt biome (Mucina *et al*., 2006) and it requires high moisture levels (rainfall, humidity). Forested communities are also constrained by the local substrate and therefore migration over time to more suitable areas is not a viable option (Eeley *et al*., 1999). During colder and drier periods such as during early MIS 3 the forested areas would have been reduced.

Evidence for glacial conditions in the southern hemisphere during MIS 4 and a shift to an ameliorating climate in MIS 3 has been recovered from Antarctic ice cores, deep sea cores and speleothems. These are similar to the harsh cold conditions seen in the LGM (MIS 2). The proxy environmental data from deep sea cores and other sites from the eastern region of South Africa indicate that between 70ka and 60ka the prevailing climatic conditions were colder and wetter than present. However, during MIS 3 temperatures began to slowly rise. Rainfall data from the Tswaing Crater suggests that rainfall began to decrease during this time, in response to decreasing insolation. On the western portion, proxy data from deep sea cores and aeolian dune sand deposits indicate colder and humid conditions with increasing wind strengths associated with lower Sea Surface Temperatures (SST) and increased cold water upwelling along the western coast during MIS 4 and MIS 3. Prior to 70ka, the south-western proxy data suggests that relatively arid conditions persisted. Records of local and regional climate change from southern Africa show that during the period 70ka-50ka, conditions were overall colder and drier in the eastern regions and colder and wetter in the western regions. Western and south-western MSA sites were abandoned earlier and for longer than MSA sites on the eastern region of South Africa as the local environments were less suitable for human occupation than in the east. Studies utilising a range of palaeoenvironmental proxies (e.g. pollen sequences and

speleothems) and modern meteorological records combined with various Global Circulation Models have indicated that the eastern portion of South Africa responds differently to climate change from that of the western regions (Barrable *et al*., 2002; Cook *et al*., 2004; Scott *et al*., 2008) . The eastern portions are influenced by moisture circulation patterns from the South West and tropical Western Indian Ocean (Fig. 7.6a and 7.6b, Vol.II, pg.35 and 36). These circulation patterns are affected by the position of the ITCZ and sea surface temperatures of the Agulhas Current (Cook *et al*., 2004). The western regions are affected by the degree of upwelling of the Benguela Current (Reason and Mulenga, 1999) and the northward movement of anticyclonic high-pressure systems that bring in moisture-rich westerly winds (Barrable *et al*., 2002).

Such a profound change was possibly due to a change in the strength or temperature of the Agulhas Current or an eastward shift of the Agulhas Retroflection (Fig. 7.6b, Vol.II, pg.36). Summer rainfall along the south eastern coast of KwaZulu-Natal is influenced by the proximity and temperature of the Agulhas Current. Alongshore variations in the rainfall gradient are related to the distance between the coast and the current at the continental shelf edge and this influence extends up to 50km inland (Jury *et al*., 1993), and includes the Sibudu region. A weaker/cooler Agulhas Current and an eastward shift of the Agulhas Retroflection would lower SST's along the eastern coast, resulting in a decrease in summer rainfall and also lower humidity levels (Jury *et al*., 1993; Reason, 2002; Reason and Mulenga, 1999; Tyson, 1999). If SST's are cool in the western southern Indian Ocean (along the southeast coast of South Africa) and warmer in the eastern areas, the air over south-eastern Africa is drier and rainfall decreases (Reason, 2002; Reason and Mulenga, 1999). Proxy evidence from core RC17-69 suggests that a weakening of the Agulhas Current occurred towards the end of MIS 4 and that there was a corresponding decrease in rainfall as indicated by the Tswaing record.

This study highlights the necessity to examine multiple strands of palaeoenvironmental evidence. The use of multiple strands of evidence allow a better understanding of the connections between global climate change events and the impact of these on local environments and human populations. It also indicates the need to ensure that dating of sites is secure before such strands of palaeoenvironmental evidence can be convincingly linked.

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CHAPTER 8. SUMMARY AND CONCLUSIONS

8.1. Introduction

The goal of this research is to develop a methodology to obtain palaeoenvironmental evidence in the form of stable carbon isotope data from archaeological charcoal from the MSA site of Sibudu Cave. This is a multi-step process. Several key requirements need to be satisfied to ensure that the carbon isotope data from the archaeological charcoals reflects an environmental signal. First the tree species must have an adaptive strategy that leads to the fractionation of carbon isotopes in response to the environment. This time series record in the rings of the tree must be preserved during the charcoal formation process. The environmental signal must also be unaffected by post-depositional processes. Ultimately the validity of the isotopic interpretation was tested by comparing it with additional proxy environmental evidence from the site. Sibudu Cave is well-suited for such research as the site has yielded a suite of environmental data from sediments, botanical and faunal remains providing detailed insights as to how the local environment has changed at various times during the past 75ka. In addition, the MSA deposits have been dated using the OSL technique, producing a precise chronological sequence to contextualise the available environmental and cultural evidence with other sites around the world.

A series of isotopic analyses and combustion experiments performed on modern tree species from KwaZulu-Natal and the Eastern Cape provinces elucidated the impact of the charcoalification process. Isotope data derived from trunk discs and cores of modern trees from the two provinces provides isotopic evidence of an environmental response to prevailing climatic conditions. The environmental signal is also reflected in the isotopic values of wood tissue of branches from the same trees. The reason for examining $\delta^{13}C$ values from branches is based on ethnographic evidence that modern communities harvest easily accessible deadwood in the form of branches and this was most likely what past inhabitants of Sibudu Cave used as fuel.

In order to demonstrate the validity of the interpretation of isotope values from charcoal, the following analyses and experiments were carried out.

8.2. Modern tree ring $\delta^{13}C$ records

8.2.1. Isotopic analysis of trunk discs and core samples

Three trunk discs of *Mimusops caffra* (Coastal Red Milkwood) and three trunk cores from two specimens of *Podocarpus latifolius* (Real Yellowwood) from KwaZulu-Natal were analysed. A single large trunk disc of *Podocarpus falcatus* (Common Yellowwood) from the Baviaans Kloof in the Eastern Cape was also examined. The results are discussed in Chapters 4 and 5. The samples of the two species of *Podocarpus* were obtained from environments that can be used as modern analogues for past environments at Sibudu. The KwaZulu-Natal samples represent conditions of high moisture availability and the Baviaans Kloof sample represents conditions of low moisture availability.

In order to compare isotope values with environmental variables, it is necessary to determine whether growth rings are laid down on an annual basis as they often have missing or additional growth rings. This was achieved using high precision radiocarbon dating techniques. The dating results highlighted the need for care when counting rings as growth rings of all three species are laid down in a lobate fashion making it possible to obtain incorrect ring counts. Three samples from the largest *M. caffra* trunk were dated at the CSIR laboratory and two samples of *P. latifolius* were dated using AMS techniques at the Centre for Isotope Research, Groningen in the Netherlands. The radiocarbon dates and age models obtained showed that growth rings for both species are laid down on an annual basis and allowed the accurate counting of growth rings for each sample.

To determine the most time-efficient method which yielded a meaningful climatic signal, wood samples of *M. caffra* were pre-treated in two ways and the $\delta^{13}C$ values compared. A set of wholewood samples was pre-treated by Soxhlet distillation. Samples of the same growth rings were further processed to yield α-cellulose. The wholewood and α-cellulose $\delta^{13}C$ values for the same samples showed a significant correlation ($r^2 = 0.75$, $P > 0.0001$, correlation = 0.85), preserving the same record of environmental responses. Wholewood $\delta^{13}C$ values were more negative on average by

1.2‰ with respect to α-cellulose $\delta^{13}C$ values. Based on these results and data from recent literature, wholewood samples were used for the remainder of the study.

The isotopic variability in wholewood samples of *M. caffra* provided a potential measure of past rainfall from 1870 to 2005. The $\delta^{13}C$ values for three specimens were compared with a local climate data set (rainfall, humidity and temperature) to determine which climatic factor had the most influence on $\delta^{13}C$ values. The $\delta^{13}C$ values of *M. caffra* showed a response to a number of dramatic variations in the regional rainfall record with isotope values fluctuating when rainfall deviates from normal (e.g. during tropical cyclones or droughts), indicating that *M. caffra* is adapted for an environment with high moisture availability. There was also isotopic evidence for a "juvenile effect" during the early stages of growth, as well as evidence for anthropogenic influences on the isotopic composition. Through the use of signal processing techniques (wavelets) it was possible to account for these influences and separate the inter-annual climatic response of the trees. The carbon isotope time series from *M. caffra* show a strong inter-annual response to rainfall variability and they allowed the reconstruction of rainfall patterns beyond the instrumental record for the region.

Three trunk cores (Core 2, 3 and 4) taken from two specimens of *P. latifolius*, from Seaton Park (KwaZulu-Natal) were analysed and their $\delta^{13}C$ values compared with the same annual rainfall, temperature and humidity records for the region, as used for the *M. caffra* specimens. A statistical regression approach was adopted to determine which of the environmental variables best correlated with the isotope data. Core 2 correlated with both temperature and humidity at the >99 % and >95% confidence levels, but Core 3, from the same tree, only correlated with humidity at the >99% confidence level. It is suggested that this anomaly may be the result of temperature and the impact of sunlight in the partitioning of resources in the tree. Core 4 correlated with temperature (>99% confidence), but not humidity nor rainfall. The correlation between carbon isotopes and humidity appears to be common to all the samples with the exception of Core 4. This may be a result of the position of this tree within a riparian zone. A trunk disc from a *P. falcatus* from the Baviaans Kloof (Eastern Cape) provided a $\delta^{13}C$ time series from trees growing in a more moisture-restricted environment, providing a contrasting analogue to those from KwaZulu-

Natal. The results for the Baviaans Kloof specimen showed a response to variation in annual rainfall.

Through the isotopic analysis of the modern tree specimens it is possible to demonstrate that their $\delta^{13}C$ time series record an environmental signal reflecting prevailing climatic conditions. These results thus satisfy the first requirement indicated in section 8.1. It was then necessary to demonstrate that the same environmental signals are recorded in the $\delta^{13}C$ values of branches from the same trees.

8.2.2. Isotopic analysis of *Podocarpus* branch samples

The $\delta^{13}C$ values of the core samples of *Podocarpus latifolius* were compared with $\delta^{13}C$ values of two corresponding branches. The isotope values showed that the same environmental response to temperature and humidity was recorded in both the core samples and the branches (see Chapter 5).

To demonstrate that an environmental signal was preserved in charcoal produced from these branches, three additional *P. latifolius* branch samples were collected from the same site. Two discs from each branch were combusted under oxidising and reducing conditions and their respective $\delta^{13}C$ values compared with $\delta^{13}C$ values from corresponding un-burnt discs. $\delta^{13}C$ values for the reduced and oxidised branch charcoal were more negative with respect to the source material, but reflected the same temperature/humidity response to prevailing climatic conditions as was recorded in fresh wood. The $\delta^{13}C$ values of trunks and branches both preserve the same environmental record. In the case of the KwaZulu-Natal, the carbon isotope values correlated with humidity and temperature data from the region. Therefore it is possible to obtain a record of past climatic conditions from the growth rings of branches. These results indicated that a meaningful environmental signal may be preserved in branch charcoal, thereby satisfying the remaining requirements listed in section 8.1, namely an adaptive strategy that leads to the isotopic fractionation of carbon isotopes in response to the environment that is recorded in fresh wood and charcoal from the same individual.

8.3. Combustion experiments

There is no clear understanding in the scientific literature of the changes that take place in the carbon isotope ratios of wood charcoal during the combustion process. In order to ensure that the $\delta^{13}C$ values obtained from burnt *P. latifolius* branches were a true reflection of the original source material, an additional combustion experiment was conducted (Chapter 5). A single green *P. latifolius* branch was analysed to determine the range of variability in the carbon isotope values of the products released from wood tissues during stages of combustion. An untreated aliquot was placed directly into a Thermo Flash Elemental Analyser (1112 series) integrated via a Thermo Finnigan Con-flo III system with a Thermo Delta V Plus Isotope Ratio Mass Spectrometer. The temperature of the furnace was increased in a step-wise manner (5° to15°C) and the $\delta^{13}C$ values of released products measured at each temperature increment, until the sample was completely combusted.

The results indicate a two-step process by which the carbon isotope composition changes during combustion and thus is linked to a pyrolitic distillation of chemical compounds within the wood. The main factor affecting the isotope composition of charcoal is the combustion temperature. Normal wood-fuelled fires burn at an average temperature of 450°C to 500°C and although overall $\delta^{13}C$ values of charcoal found at this temperature are systematically more negative relative to the green wood from which it is formed, this temperature range does not obscure the environmental signals. The results from this experiment demonstrate that branches and carbonised wood preserve a record of local environmental conditions. It is therefore possible to use $\delta^{13}C$ data from archaeological charcoal as a palaeoenvironmental proxy.

8.4. Validation of archaeological $\delta^{13}C$ data with other palaeoenvironmental proxies from Sibudu Cave

8.4.1. Sampling and analysis of archaeological charcoal

Samples of archaeological charcoal that had previously been identified using wood anatomical features and a number of reference collections (Allott, 2004; 2005; 2006)

were selected from a number of layers representing three MSA industries at Sibudu Cave. These layers include the Howiesons Poort, the post-Howiesons Poort and the late MSA industries, for which the chronology has been well established using OSL dating techniques. The genus *Podocarpus* was selected because of its predominance in certain of the MSA layers at Sibudu Cave. *Celtis* charcoal was sampled from the same archaeological layers as *Podocarpus* charcoal on the assumption that the different environmental sensitivity of this genus should provide a more robust interpretative framework than the *Podocarpus* alone. Methodological details of the analyses are discussed in Chapters 5 and 6.

To ensure that the $\delta^{13}C$ values obtained from the Sibudu archaeological charcoal were not due to post-depositional contaminants, subsets of *Podocarpus* and *Celtis* charcoal from the three MSA periods were further pre-treated using the acid-alkali-acid method (AAA) and re-analysed (See Chapter 6). This pre-treatment ensures that any post-depositional contaminants such as humic substances and or carbonates are removed. The $\delta^{13}C$ range, variance and standard deviations of the AAA samples were very similar to the original samples and this indicates relatively little post-depositional contamination of the Sibudu charcoals.

8.4.2. Stable carbon isotope-based palaeoenvironmental interpretations

The interpretation of the $\delta^{13}C$ analyses of archaeological charcoal is based on both the absolute values that were obtained, as well as the variability. Since the values have been demonstrated to be responsive to the environment, the distribution of isotope values is an indication of the range of adaptive responses that the trees required in the past. During the Howiesons Poort occupation of Sibudu (65ka-62ka), $\delta^{13}C$ values for both *Podocarpus* and *Celtis* suggest that the environment was one of high moisture availability. Conditions were well-suited for the presence of *Podocarpus* and this was indicated by the wide range and variance and high standard deviations of the isotopic data. The isotopic data suggest that *Podocarpus* was able to utilise a broad variety of adaptive responses to prevailing conditions and the results correspond well with those from the modern data from Seaton Park. The isotopic results obtained for the *Celtis* samples have a very low range and variance, suggesting that this genus was not well-

adapted to the environment and may have been at the wet limit of its adaptive response.

The $\delta^{13}C$ values for *Podocarpus* and *Celtis* from the post-Howiesons Poort (~58ka) reveal a very different picture, indicating that available moisture levels were much lower than those of the Howiesons Poort. Stable carbon isotope values for *Podocarpus* stand out with much lower mean values and a very small range and variance, representing an isotopic record of markedly changed environmental conditions. It appears that *Podocarpus* displayed a highly restricted adaptive response and may have been limited to refuge areas in the region. The Baviaans Kloof provides an analogous environment for this period, although conditions may have even drier at Sibudu during the post-Howiesons Poort. The *Celtis* isotope data show that this genus was well-adapted to conditions of lower moisture availability and most likely had a wider local distribution than in the Howiesons Poort.

There is evidence for further environmental changes during the late MSA (~48ka). The $\delta^{13}C$ values for *Podocarpus* and *Celtis* suggest that there was low moisture availability at this time. Conditions were warmer than those at ~58ka, and it was less moist than between 65ka and 62ka. Carbon isotope values for *Podocarpus* have a higher range and variance than those of the post-Howiesons Poort at ~58ka, but are still lower than those of the Howiesons Poort 65ka to 62ka samples. This suggests that *Podocarpus* was less restricted in its adaptive responses to prevailing environmental conditions than during the post-Howiesons Poort. The *Celtis* data show a similar pattern to that of the post-Howiesons Poort, which suggests that the genus was also well adapted to the local environment of the time.

Both genera that were analysed from Sibudu showed similar trends in their isotopic composition over time, but each has markedly different environmental adaptations. This adds support to the earlier conclusion that the charcoal isotopes do not reflect post-depositional contamination or charcoal formation processes.

8.4.3. Comparison of Sibudu palaeoenvironmental proxies

Since the Wadley excavations at Sibudu Cave began in 1998, a multi-disciplinary approach has been applied to the various material recovered from the MSA layers. Consequently researchers have developed a detailed understanding of the environmental and cultural sequences over the last 75 thousand years. There are several factors that need to be considered when examining the environmental data. First, the site was not occupied continuously, so there are large gaps of as much as 10ka in the environmental record. Second, a mosaic environment was present around the site throughout the MSA. This persistent mosaic environment was most likely a product of site aspect and the presence of the Tongati River (at least during the periods that Sibudu was occupied). These would have allowed the localised occurrence of certain species which would not have otherwise have survived when the areas surrounding the site were drier. Third, the sediment of the site is largely anthropogenic in origin, which may complicate environmental interpretations of proxy data (Goldberg *et al*., 2009; Pickering, 2006). Despite these limitations it is still possible to use the range of proxy data to reconstruct past environmental and climatic conditions during periods of site occupation.

Botanical, faunal and sedimentological evidence from the Howiesons Poort layers indicate that the climate was cooler than at present, and conditions were also humid. A substantial evergreen forest was supported although areas of slightly more open woodland communities were also present in the area. This supports the isotopic data which show that *Podocarpus* was well-adapted to the prevailing environmental conditions, whilst *Celtis* was at its limit of adaptive responses. The post-Howiesons Poort (~58ka) layers provide evidence for oscillating climatic conditions where substantial changes in the composition of the various plant and animal communities took place. The Howiesons Poort predominantly forested environment was drastically reduced and open woodland and grassland communities developed. However, it is important to note that due to the continual presence of water in the Tongati River, evergreen riverine forest persisted. Stable carbon isotope values from *Podocarpus* charcoals from the post-Howiesons Poort suggest that these forests were reduced to sheltered refuge areas and the genus showed a very limited adaptive response. Proxy environmental data from the late MSA (~48ka) layers show an initial period of

warming followed by a cooler period. The combined seed, phytolith and charcoal data indicate an overall increase in deciduous woodland within close proximity to the site. Carbon isotope values for both genera became more negative again at ~48ka suggesting greater moisture availability once more. At ~58ka and ~48ka, *Celtis* $\delta^{13}C$ values suggest a wider range of adaptive strategies were available for the genus which is suited to woodland and savanna environments.

Stable carbon isotope data from archaeological charcoal thus have the potential to provide an additional source of proxy evidence for localised environmental changes during the MSA occupations at Sibudu Cave. The currently available suite of environmental proxy data from botanical, faunal and sedimentological data provides supporting evidence for the isotope-based interpretations, thereby validating the isotopic results.

8.5. Environmental change during the Sibudu MSA in a regional and global context

The isotopic evidence developed here, and the corroborative evidence from other environmental proxies, provide a very coherent indication of past environments at Sibudu Cave. These can be compared with regional and global climate changes (Chapter 7). The focus is on events that occurred at the end of the Howiesons Poort occupation through the beginning of the post-Howiesons Poort that corresponds to the end of Marine Isotope Stage 4 (MIS 4) and the early phase of MIS 3 at about 60ka.

At this time the local Sibudu environment changes from a predominantly cool, humid forested environment to a drier, open grassland/woodland mosaic community with remnants of riverine forest. Forested communities require high moisture levels (rainfall and humidity) and are also constrained by the local substrate (Eeley *et al*., 1999; Mucina *et al*., 2006). Under colder and drier conditions evidenced for early MIS 3, the forested areas surrounding Sibudu would have been reduced. This shift in local vegetation communities may have been due to a change in the strength of the Agulhas Current and an eastward shift of the Agulhas Retroflection. Summer rainfall along the south eastern coast of KwaZulu-Natal is influenced by the proximity and temperature of the Agulhas Current. Alongshore variations in the rainfall gradient are

related to the distance between the coast and the current at the continental shelf edge and this influence extends up to 50km inland (Jury et al., 1993), including the Sibudu region. A weaker Agulhas Current and an eastward shift of the Agulhas Retroflection would lower sea surface temperatures along the eastern coast, resulting in a decrease in summer rainfall and also lower humidity levels (Jury et al., 1993; Reason, 2002; Reason and Mulenga, 1999; Tyson, 1999). If sea surface temperatures are cool in the western southern Indian Ocean (along the southeast coast of South Africa) and warmer in the eastern areas, the air over south-eastern Africa is drier and rainfall decreases (Reason, 2002; Reason and Mulenga, 1999). Proxy evidence from marine core RC17-69, off the eastern coast of KwaZulu-Natal, suggests that a weakening of the Agulhas Current occurred towards the end of MIS 4 and there is a corresponding decrease in rainfall as indicated by the Tswaing record. At approximately 58ka (post-Howiesons Poort) it appears that local environmental conditions were unfavourable and Sibudu Cave was abandoned for almost 10 000 years until 48ka (late MSA) when the site was re-occupied.

Proxy palaeoenvironmental data from six other MSA sites from South Africa were also examined to see if a similar change in climate was recorded. This inter-site comparison highlighted difficulties associated with differences in excavation methods, data standardization or insufficient data and the fact that some sites do not have well resolved age models. However it was possible to determine some general regional patterns of environmental change across the country at the transition between MIS 4 and MIS 3. Environmental evidence suggests that the eastern regions experienced greater precipitation than the more arid western regions.

Isotopic and chemistry data from Antarctic ice cores, deep sea cores and speleothems indicate that glacial conditions similar to those during the Last Glacial Maximum (MIS 2) prevailed in the southern hemisphere during MIS 4. Evidence for a shift to an ameliorating climate in MIS 3 is also apparent in these data. This is reflected in the range of proxy evidence from the Howiesons Poort (65ka-62ka) layers at Sibudu. However, during MIS 3 temperatures began to rise slowly. Rainfall data from the Tswaing Crater indicates that overall rainfall began to decrease during this time, in a response to decreasing insolation levels. Evidence from the Tswaing Crater needs to be used with care, due to the age model that has been extrapolated for this site. A

range of southern African palaeoenvironmental proxies (e.g. pollen sequences and speleothems) and modern meteorological records combined with various Global Circulation Models have indicated that the eastern portion of South Africa responds differently to climate change from that of the western regions (Barrable *et al*., 2002; Cook *et al*., 2004; Scott *et al*., 2008). The eastern portions are influenced by moisture circulation patterns from the South West and tropical Western Indian Ocean affected by the position of the ITCZ and sea surface temperatures of the Agulhas Current (Cook *et al*., 2004) (Fig, 7.6a and 7.6b, Vol. II, pg.35 and 36). The western regions are affected by the northward movement of anticyclonic high-pressure systems that bring in moisture-rich westerly winds and the degree of upwelling of the Benguela Current (Barrable *et al*., 2002).

During the period 70-50ka, local and regional climate change records from southern Africa show that in the eastern regions, conditions were overall colder and drier and colder and wetter in the western regions than present. MSA sites located in the western and south-western regions were abandoned earlier and for longer than MSA sites from the eastern region of South Africa because local environments were less suitable for human occupation than those in the east.

This inter-site comparison demonstrates the strength of the multi-disciplinary approach, which utilised multiple strands of palaeoenvironmental evidence to understand the connections between global climate change events and the impact of these changes on local environments and human populations between MIS 4 and early MIS 3. It also highlights the necessity for the secure dating of sites to allow the convincing linkage of palaeoenvironmental evidence.

8.6. Avenues for further research

During the course of this study it became apparent that there are a number of areas where further research is necessary, particularly if the use of stable carbon isotopes of charcoal are to be used as a source of environmental data in the southern African context. Based on currently published results, the charcoal formation process and its effect on stable carbon isotopes are still not well understood. Researchers have produced conflicting results on the effects of combustion temperatures and duration,

type of plant material combusted and chemistry of the source material on the carbon isotope composition of charcoal. The majority of charcoal isotope and tree ring isotope research has been carried out in Europe on a limited number of taxa. It is therefore necessary to expand the available southern African data. Further experimental work is required to identify the range of volatile compounds released during combustion. As the objective is to obtain proxy environmental evidence from $\delta^{13}C$ values of charcoal, it would be useful to develop a standardised database of isotopic values for a range of southern African woody species, from both identified archaeological and modern charcoal. There is a potentially substantial source of identified archaeological charcoal that can accessed from a variety of archaeological sites across southern Africa, along with a range of supporting proxy environmental evidence from these site. Of particular interest are MSA sites such as Rose Cottage, Klasies River Mouth, Boomplaas Cave, Blombos Cave and Diepkloof rock shelter. These sites will be able to provide suitable charcoal for isotopic analysis, representing the two main climatic zones in southern Africa, namely the eastern summer rainfall region and the western winter rainfall region. Such a database would provide complementary material to the growing collection of southern African tree ring isotope records.

At present there are limited isotopic time series derived from southern African tree ring records, representing a range of taxa from different environments. These currently include examples of the following genera; *Podocarpus* (Yellowwood), *Celtis* (White Stinkwood), *Acacia*, *Widdringtonia* (Cedar), *Mimusops* (Milkwood), *Breonadia* (Matumi) and *Adansonia* (Baobab). The carbon isotope time series obtained from these taxa have all shown that an environmental response (e.g. rainfall, humidity or temperature) is recorded in annual growth rings. It would be advantageous to expand the number of tree ring time series in systematic fashion, combining oxygen ($\delta^{18}O$) and carbon ($\delta^{13}C$) isotope analyses with traditional dendrological methods. In this way, it will be possible to develop and extend climatic records beyond the instrumental range. Ideally such data should be standardised for the region and the predictive skill of reconstructed climatic variables be tested using standard metrics (National Research Council, 2006). The species analysed should be representative of those with a limited or restricted range, as well as those with a wide

range. The species should also be sourced from both main climatic zones of southern Africa.

This research has demonstrated the methodology and the potential of doing palaeoenvironmental research through combining stable carbon isotope analysis of archaeological and modern charcoal. Coupled with an increased number of indigenous tree ring isotope time series it will be possible to create a powerful database of proxy environmental records for the recent past and deeper in time.

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