Geochemical insights into the influence of Holocene sea level change on the evolution of the Mkhuze River Delta, Lake St Lucia, northern KwaZulu-Natal

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Declaration

I, Caldin Higgs, declare that this thesis is my own, unaided work, except where referenced and otherwise acknowledged. This thesis is submitted in fulfilment of the requirements for the degree of Master of Science at the University of the Witwatersrand, Johannesburg. I have not submitted this work for examination at any other university.

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Abstract

The Mkhuze River discharges into the most northern part of Lake St Lucia, via a contemporary bayhead delta. The delta formed in response to sea level rise during the last deglaciation and today exerts great influence on the functioning of Lake St Lucia, one of the largest estuarine systems in Africa and a globally important conservation area. A sediment core (11.5 m) was extracted from the distal end of the delta to examine the geomorphic evolution of the Mkhuze River Delta and links with variations in Holocene sea level and climate. Radiocarbon and optically-stimulated luminescence dating show that the core captured the entire Holocene infill and documents changes in sedimentation over the last ~13.8 kyr. Grain size and high resolution XRF analysis indicates that initiation of the modern delta occurred since ~7200 cal yr BP , when deglacial sea-level rise reached present-day level. Initial Holocene aged sediments are dominated by clay and silt material that was deposited when seawater intruded into Lake St Lucia *via* a palaeo-river connection to the ocean at Leven Point. The influx of silt and clay material was accompanied by the emergence of an onshore proto-barrier that created a sheltered lagoonal environment and promoted the accumulation of fine fluvial sediment. The presence of discrete, coarse-grained horizons enriched in zircon identifies a period of increased marine palaeostorm activity between 4700 and 2500 cal yr BP. This period is characterised by the presence of discrete shell fragment accumulations and is interpreted to reflect a strongly positive Indian Ocean dipole anomaly, which resulted in warmer sea surface temperatures and an increase in regional cyclone activity and frequency. The upper part of the core is characterized by generally fine silt and is marked by a decrease in sedimentation rate that corresponds to a phase of lateral delta progradation. The last ~1700 cal yr BP years of the record identify with subtle changes in grain size that can be attributed to a strengthening in El Niño–Southern Oscillation (ENSO) activity, which is known to be associated with prolonged drought and wind erosion in eastern South Africa. This study highlights the usefulness of coastal geochemical records in identifying environmental changes and related climate signals at a regional scale.

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Chapter 1: Introduction

Sediment cores can be used to examine long-term variations in climate dynamics and sedimentary processes (Hodgson *et al.*, 2005; Woodruff *et al.*, 2009; Kylander *et al.*, 2011; Gilli *et al.*, 2013). Past environmental changes are reconstructed using a combination of different proxy data, which include geochemistry (Kylander *et al.*, 2011), microfossils [e.g. diatoms (Gomes *et al.*, submitted) and foraminifera (Jennings and Weiner, 1996)], pollen (Scott *et al.*, 2012) and charcoal (Swain, 1978). Sediment archives from river delta systems are of particular interest for high-resolution reconstruction as they can capture changes from both the river catchment area and the adjacent ocean.

Water systems on the northern KwaZulu-Natal (KZN) coastal plain were developed following a rise in sea level that accompanied the transition to the present interglacial climate. Sediments stored in the shoreface migrate inland with sea-level rise, forming transgressive barriers that isolated estuaries and wetlands from the open sea (González-Villanueva *et al.*, 2015). The diverse Holocene morphological features along the coast include barrier lagoons, lakes, estuaries and wetlands, many of which occupy incised valleys that have since flooded and filled. These systems thus contain sedimentary records that document their geomorphic response to sea-level variations and changes in the coastal palaeo-environment. Furthermore, the eastern coastline of southern Africa remained marginally unaffected by glacio-isostatic processes associated with melting ice sheet during the post-glacial period, and is therefore an important region for understanding eustatic sea level change (Norström *et al.*, 2012).

Lake St Lucia is the largest waterbody on the KZN coastal plain that is characterised by three main depositional basins: False Bay, North Lake and South Lake. The lake is classified as a shallow estuarine lake, attaining an average depth of approximately 1.5 m (Stretch and Maro, 2013). Lake St Lucia has been subjected to frequent marine transgressions and regressions that transitioned the lake from an open marine-dominated embayment system to a lagoonal environment and today it is characterised by lacustrine conditions (Gomes *et al.*, submitted). The Mkhuze River, the largest supplier of sediment and freshwater to Lake St Lucia, that discharges into the northern part of the lake via a contemporary bayhead delta.

The Mkhuze River Delta has been subject to varying degrees of fluvial sediment supply, marine influence and changes in Holocene sea level. These changes and the changes in the coastal palaeo-environment are likely recorded in sediments from Mkhuze Delta. Due to its sheltered location and well-preserved state, the Mkhuze Delta represents a promising site for understanding Holocene sedimentary patterns in response to changes in climate and sea level.

Aims and Objectives

This study is aimed at understanding the Holocene evolution of the Mkhuze River delta using a combination of geochemical and sedimentological analyses.

The main objectives of this study are to:

- Identify changes in sediment deposition and source using geochemical analysis.
- Relate changes recorded in the sedimentary record to the evolution of Lake St Lucia.
- Link the results from this study with previously published research to examine regional variations in sea level and climate.

Chapter 2: South African sea level history and palaeoenvironmental indicators recorded in lake sediments

2.1. Regional Quaternary sea level history

The South African coastline and shelf is well positioned for research into sea-level rise, as the coastline consist of two trailing margins that were tectonically stable during the late Quaternary (Hendey and Volman, 1986). In addition, the eastern coastline of southern Africa remained relatively unaffected by glacio-isostatic processes associated with melting ice sheet during the post-glacial period, and is therefore an important region for the understanding eustatic sea level change (Norström *et al.*, 2012). Evidence of past sea-level changes along the coastline of southern Africa are indicated in dates obtained from beachrocks, marine sediments and wave-cut platforms (Hendey and Volman, 1986; Ramsay, 1995; Miller, 1996; Compton, 2006; Strachan *et al.*, 2014). For the east coast, Ramsay and Cooper (2002) developed a sea level curve covering the Late Quaternary based on indicators from the South African coast and shelf (Fig. 1).

Several studies incorporated a last glacial maximum (LGM) along the South African coastline that reported depths of excess -100 m (Fairbanks, 1989; Pether, 1994; Ramsay and Cooper, 2002). By using seismic stratigraphic evidence, studies suggested that sea level fell to approximately −120 m below present-day sea level. This occurred during a regression that started approximately 25 000 years BP (Fairbanks, 1989; Pether, 1994; Ramsay and Cooper, 2002). The regression climaxed approximately 17 000 years BP, which in turn became the LGM (Ramsay and Cooper, 2002). Following this LGM, climatic amelioration in global temperatures resulted in rapid rise in sea levels and overall deglaciation.

A rapid (8 mm y^{-1}) rise in sea level following the LGM (Termination I), occurred before reaching present-day levels ~7300 cal yr BP (Fig. 2). A deceleration in sea level rise following this was the primary forcing factor responsible for the development of most modern estuaries and lagoons. Sediments stored in the shoreface, migrated inland with sea-level rise forming transgressive barriers that isolated coastal systems from the open ocean (González-

Villanueva *et al.*, 2015). Ramsay (1995) indicates that sea level reached a highstand at 3.5 m ~5300 cal yr BP (Fig. 2). Evidence from other coastal areas within South Africa generally support a mid-Holocene highstand, although the timing and amplitude remain uncertain (Miller *et al.*, 1993; Ramsay, 1995; Compton, 2006; Strachan *et al.*, 2014). Following the mid-Holocene highstand, sea level returned to present day levels, with Ramsay (1995) proposing a drop-in sea level to – 2m below amsl (~3000 cal yr BP). A final rise in sea level to +1.5 m occurred ~1610 cal yr BP, followed by stabilisation near present day levels ~900 cal yr BP.

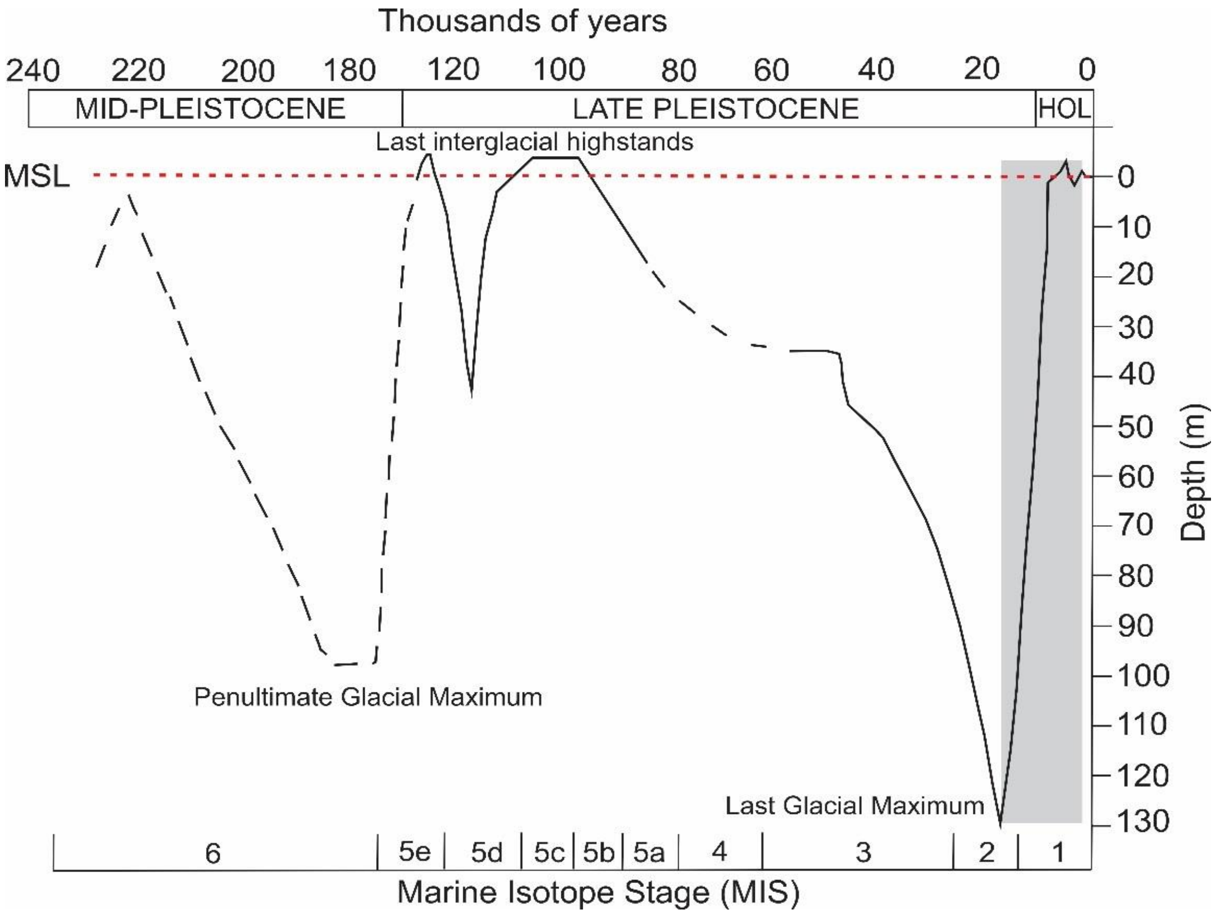


Figure 1: Sea-level curve for past 200 000 years based on available sea-level indicators from the South African coast and shelf (Ramsay and Cooper, 2002).

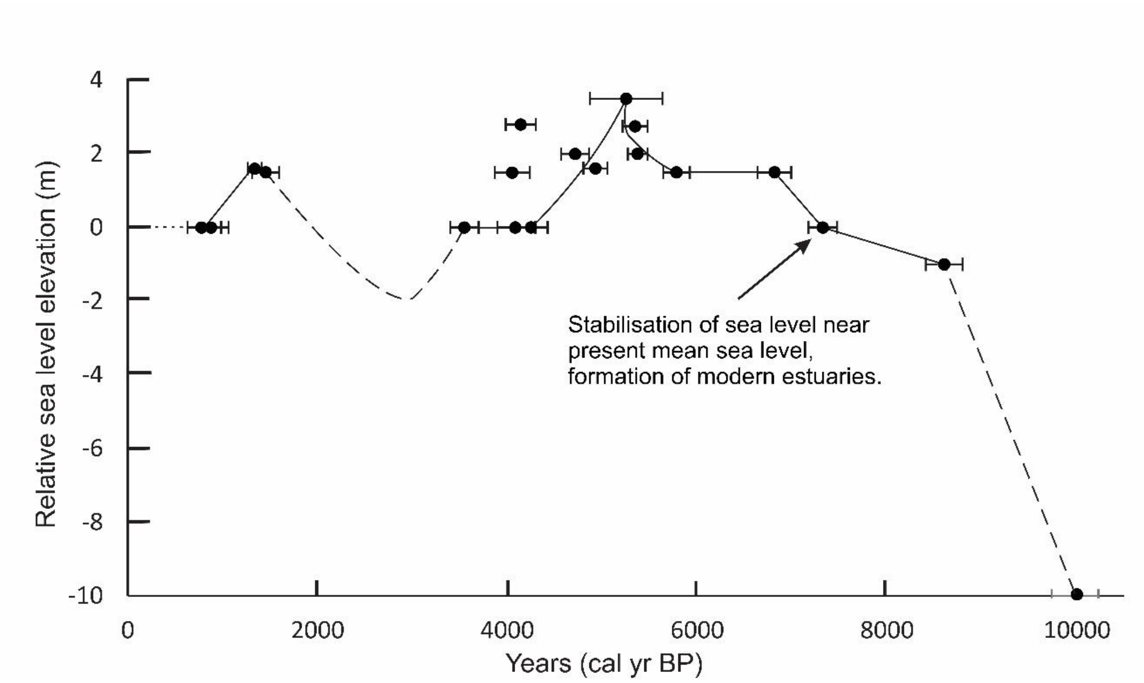


Figure 2: Fluctuations in sea level along east coast of South Africa coastline over the Holocene (Ramsay, 1995). All dates were calibrated using the Southern Hemisphere atmospheric curve SHCal.13.

The continuation of sea level rise and fall (Figs. 1 & 2) has played an important role in altering the South African coastline's environment and climate. It has therefore played a major role in the formation of the coastal water bodies found along the eastern coastline of South Africa. The influence the sea level has had on surrounding landscapes, involves determining the role of multiple processes. For example, it determines the lower limit of continental denudation and thus controls erosion potential of rivers flowing into the sea (Mastronuzzi and Sansò, 2003; Botha *et al.*, 2013).

2.2. Formation of coastal lakes along the east coast of South Africa

The late Quaternary evolution of the north-eastern coastal waterbodies is complex and poorly understood due to several factors (Fig. 3). These factors include lack of suitable dating materials, the sparseness of fossil remains and the widespread reworking of older sands (King, 1972; Davies, 1976; McMillan, 1993; Botha, 1997; Wright *et al.*, 1999; Wright *et al.*, 2000). The formation of the large coastal waterbodies along northern KZN can be attributed to Mio-Pliocene sea-level lowstands, which caused rivers on the coastal plain to incise into underlying Cretaceous and Palaeocene sedimentary sequences (Wright *et al.*, 2000). Seismic evidence obtained from the three coastal lakes (Lake St Lucia, Lake Sibaya and Kosi Bay) show that the original bases of these lakes were scoured to -40 m during the late Miocene/Early Pliocene regressive phase (Van Heerden, 1987; Wright and Mason, 1993; Miller, 1996; Wright *et al.*, 1999; Benallack *et al.*, 2016).

At the last interglacial (~125000 years BP), all three systems displayed similar morphologies, comprising large protected embayments with widespread tidal-flats that were protected by barrier archipelagos (Wright *et al.*, 2000). Fossil coral evidence from the False Bay region of Lake St Lucia indicate the presence of shallow clear water conditions,

that would have formed by a tranquil environment created by a protected barrier (Botha *et al.*, 2013). The marine transgression following the LGM had a profound influence on low lying basins such as St Lucia. Sand barrier initiation in response to littoral sediment supply and aeolian deposition eventually caused a constriction and subsequent closure of channel inlets (Wright *et al.*, 2000). At Lake St Lucia, impounded water levels rose well above present mean sea-level and flooded low-lying swamps and river valleys both north and south of the lake (Wright *et al.*, 2000). Rising water levels allowed the lake to merge with the adjacent Mfolozi system, resulting in both systems sharing a river outlet to the sea at the present river mouth location (Wright *et al.*, 2000; De Lecea *et al.*, 2015).

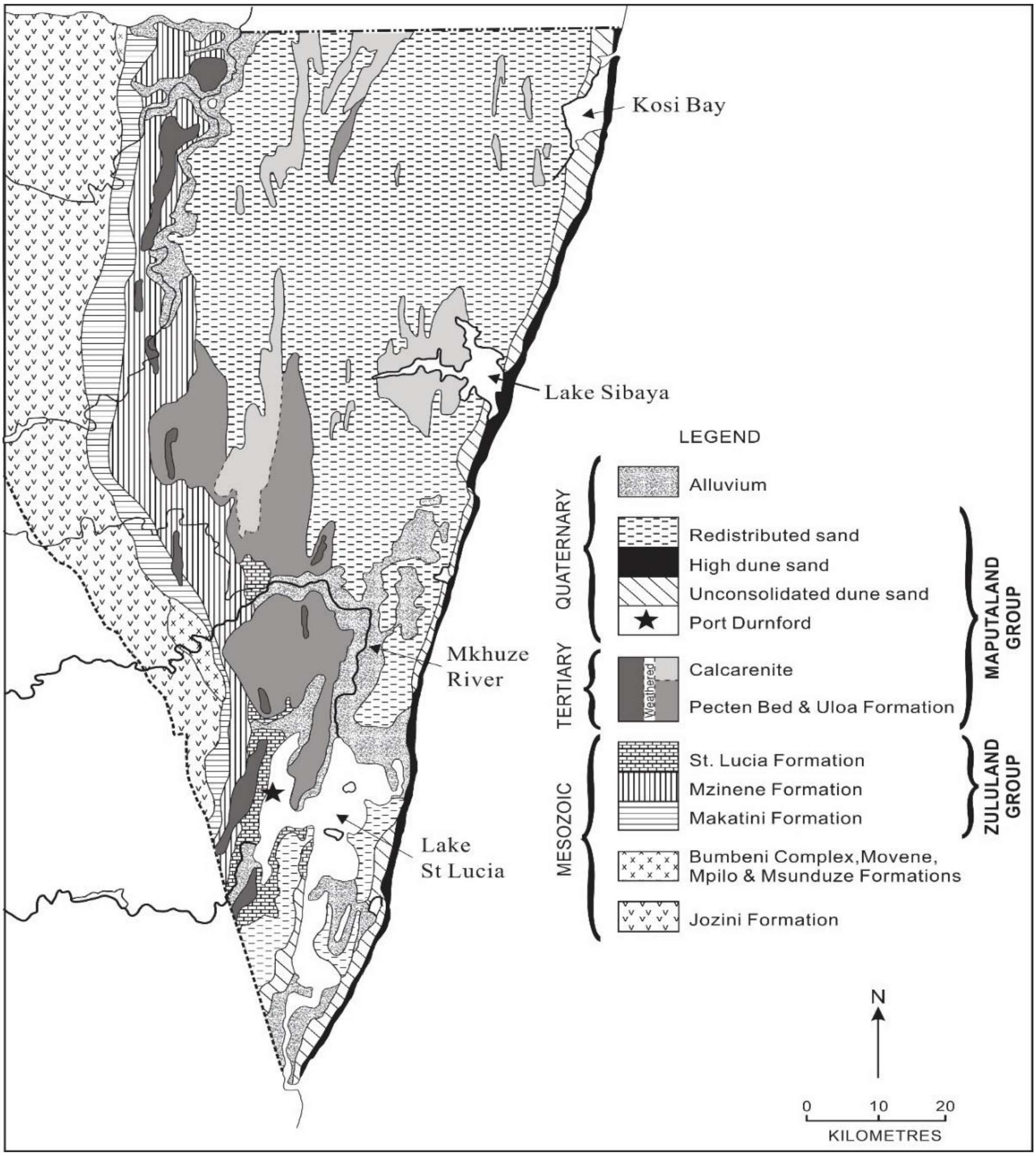


Figure 3: Regional map showing the main coastal waterbodies found on the northern KwaZulu-Natal coastal plain (Wright et al., 2000).

2.3. Sediment records as archives of palaeoenvironmental information

In order to understand changes in climate and environment over geological timescales, long-term records are required (Willis and Birks, 2006; Brewer *et al.*, 2012). Predicted future climate change as well as increasing impacts from human activities has placed a growing importance on palaeoenvironmental research. Palaeoenvironmental studies are able to provide perspective on recent observations and help identify mechanisms driving environmental change.

Lake deposits often offer valuable prospects for reconstructing past environmental change, as they record variations that have occurred both within the waterbody itself (in-situ) and from the surrounding catchment area (ex-situ) (Foster and Walling, 1994; Zolitschka *et al.*, 2015). It is often important to differentiate between these two depositional sources, in order to reliably interpret geochemical records. In such cases, a multidisciplinary approach that incorporates the use of multiple proxies is often needed. A number of physical (e.g. grain size, magnetism), biological (e.g. diatom, pollen) and chemical (e.g. elemental, isotopes, biomarkers) proxies are available for palaeoenvironmental reconstruction. This section focuses on some of the physical and inorganic geochemical proxies available as these were employed in this investigation.

2.3.1. Physical proxies

Grain size datasets from lake sediments can be used as a proxy to interpret changes in depositional conditions related to sediment provenance and transport energy (Beierle *et al.*, 2002; Holz *et al.*, 2007; Schillereff *et al.*, 2014). Grain size changes can provide an indication of the dominant mode of transport, with coarser grain size indicative of transport via hydrological pathways and finer grain sizes indicative of aeolian transport (Wohlfarth *et al.*, 2008). Grain size data are also traditionally used as an indicator of river discharge and are thus a possible proxy for paleoprecipatation (Schillereff *et al.*, 2014).

Loss on ignition (LOI) is a simple method for estimating the organic and carbonate content of sediments. Loss on ignition concentrations are generally influenced by both the production of biomass and subsequent degree of degradation, and can thus reflect changes associated with the origin of organic matter, biogeochemical processes and post-depositional conditions (Snyder *et al.*, 2004).

2.3.2. Inorganic geochemical proxies

Variations in sediment geochemistry can be used to examine multiple sediment sources (variable inputs from terrestrial and marine sources), biotic processes, particle sorting, changes in hydrology and catchment processes (Dellwig *et al.*, 2000; Minyuk *et al.*, 2007; Kylander *et al.*, 2011). These processes themselves reflect and respond to palaeoenvironmental changes. In palaeoenvironmental studies, relative changes in elemental ratios are often more informative than absolute concentrations. Elemental ratios can be used to identify sediment provenance, marine influences, as well as changes in transport and geochemical processes. In variable settings such as coastal lakes, it is often useful to compare relative variations against a conservative element that is not affected by both biological and post-depositional processes (Schropp *et al.*, 1990). In addition, absolute concentrations can be affected by possible dilution and matrix effects (Beckhoff *et al.,* 2007). Aluminium is often used as a reference element in such cases, as it is usually associated with alumino-silicates, behaves conservatively in the environment and it is not affected by biological or redox processes (Schropp *et al.*, 1990). Elemental ratios such as Rb/Al, Ti/Al, Fe/Al are all usually associated with silicates and thus changes in their abundance are indicative of temporal variations in minerogenic input to the lake (Haberzettl *et al.*, 2008).

Sediment geochemistry is useful in tracing environmental changes over time, induced by both natural and anthropogenic causes (López-González *et al.*, 2006). The geochemistry of the sediment can be strongly influenced and controlled by grain size of the dominant mineral host and its particle size sorting (Gierlowski-Kordesch, 2004; Kylander *et al.*, 2011). Often Si and Al are represented as a ratio, as Si is found in coarser sediments such as quartz and Al found in clay minerals. Changes in Si/Al ratios can therefore indicate transport energy with higher values representing coarser sediments deposited under higher energy conditions (Kylander *et al.*, 2011). Like Si, Zr is normally associated with medium to coarse sediment fraction, an example is its heavy mineral form zircon. In finer sediments, Zr/Rb ratio can be used as a proxy for changes in grain size, with lower values representing fine-grained material and higher values representing the coarse-grained material (Dypvik and Harris, 2001; Kylander *et al.*, 2011).

Coastal lakes receive sediments through fluvial, marine and aeolian processes, which provide information on the climatic conditions prevalent in the region surrounding the lake. One of the climatic conditions revealed in lake sediment is chemical weathering, which is an important feature in the hydrochemical cycle of elements (Warrier and Shankar, 2009). Good indices of chemical weathering are ratios of K/Al and K/Rb for example (Zabel *et al.*, 2001; Pattan *et al.*, 2005). This is due do that larger cations like Al and Rb remain in the source rock, while smaller cations like K, Sr and Na are selectively leached (Nesbitt and Markovics, 1980). The smaller cations are lost from source rocks during chemical alteration and become less abundant than immobile elements such as Al or Ti (Hu *et al.*, 2015). Therefore, these elements are often used as indicators of warmer and/or wetter climates in addition to overall weathering intensities (Gierlowski-Kordesch, 2004).

In lake sediments, depending on the catchment rock composition Ca and Sr can be related to either carbonate weathering, bioclastic material or in-situ precipitation (Kober *et al.*, 2007; Shand *et al.*, 2007; Kylander *et al.*, 2011). The latter occurs when the chemical concentration of lake water reaches carbonate saturation, which may result from evaporation and lower lake levels (Cohen, 2003).

Chapter 3: Study Area

3.1. Regional Background

Lake St Lucia (Fig. 4) is one of the largest and most important estuarine systems in Africa, covering a surface area of 350 km^2 with an average depth of approximately 1 m. The lake forms part of the iSimangaliso Wetland Park, which gained international recognition when it was declared a Ramsar site and listed as a UNESCO World Heritage site in 1999 (Taylor and De la Harpe, 1995). Lake St Lucia is the largest protected estuarine habitat for hippos, crocodiles, aquatic birds, estuary-associated fish and invertebrates on the continent (Whitfield and Taylor, 2009). Lake St Lucia has received significant research attention through understanding its hydrology, evolution, climate, conservation and management (Alexander *et al.*, 1986; Wright and Mason, 1993; Cyrus *et al.*, 2010; Humphries *et al.*, 2010; Humphries *et al.*, 2011). However, few studies have focussed on understanding the geomorphic evolution of the system and the controls governing its response to changing environmental conditions (Whitfield and Taylor, 2009).

Lake St Lucia comprises three interconnected basins, namely False Bay, North Lake and South Lake (Fig. 4). The lake compartments are separated from the ocean by a Pleistocene-Holocene barrier dune complex, with the only contemporary link to the ocean via an ~21 km long sinuous channel known as the Narrows. Freshwater and sediment input into these basins is supplied by five major rivers (Hluhluwe, Mkhuze, Mfolozi, Nyalazi and Mzinene) that feed into Lake St. Lucia and collectively drain a catchment with an estimated area between 8900 to 9065 km^2 (Begg, 1978). The Mkhuze River is the primary contributor of fresh water for the northern parts of Lake St. Lucia. During the drought period of 2002-2007, the river dried up and ceased flowing. This resulted in an increase in salinity for the northern parts of Lake St. Lucia, with the only source of freshwater from groundwater (Whitfield and Taylor, 2009).

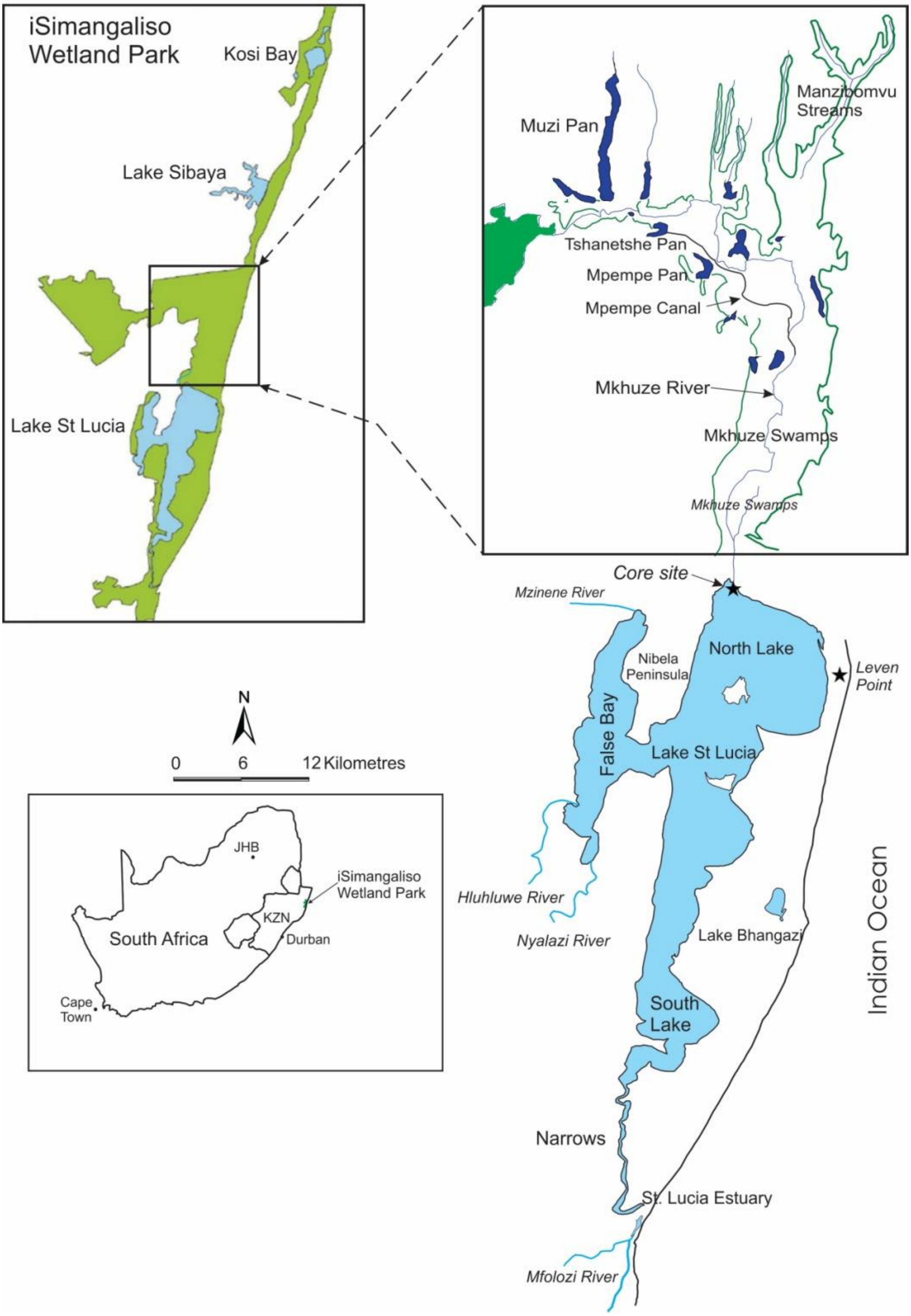


Figure 4: Regional map indicating the main features associated with the Mkhuze swamps and Lake St. Lucia.

3.2. Mkhuze river hydrology

The Mkhuze River is strongly seasonal, flowing during the wet summer, but often drying up over winter months. Mean annual flow is variable, ranging between 211 and 326 × 10^6 m^3 (Stormanns *et al.*, 1987). The input of freshwater from Mkhuze River is one of the main factors responsible for the water balance and salinity variations in North Lake (Whitfield *et al.*, 2006). Flooding in this area is highly variable and usually occurs in response to cyclonic activity. During flooding, the Mkhuze River overtops its banks and inundates the surrounding floodplain. This plays an important role in recharging the floodplain lakes and wetlands (Humphries *et al.*, 2010). The most severe flood events on record occurred in 1984 (Cyclone Domoina) and 1987 (cut-off low pressure system). The distal section of the river is associated with an extensive wetland system, comprising of a range of seasonally flooded swamps, riparian floodplains and shallow lakes (Stormanns *et al.*, 1987). The extensive Mkhuze wetland system acts an important filter to remove river sediments from the water before it enters North Lake (Whitfield and Taylor, 2009). The western portion of the wetland does not fall under the protection of the iSimangaliso Wetland Park authority and has therefore been impacted by human activity (Whitfield and Taylor, 2009).

3.2.1. Human influences

The Mkhuze floodplain has been affected to some extent by human activities. The floodplain has gone from having zero agricultural influence in 1937 to having 1385 ha in 1996, therefore, experiencing an exponential increase in agriculture (Neal, 2001). Cultivation in the Mkhuze floodplain first occurred in the mid to lower floodplain that ran along the western fringe of the wetland near human settlements and subsequently concentrated on high lying alluvial ridges (Ellery *et al.*, 2013). This was achieved through clearing of riparian forests that turned the land into cultivated subsistence farming (Ellery *et al.*, 2013). This agriculture transformation was most prevalent during droughts, and recently a change from subsistence farming to intensive farming of sugar cane has occurred (Neal, 2001; Whitfield and Taylor, 2009). As a result, the transformation may compromise the sediment-retaining abilities of the swamp. If this occurs, a rapid influx of sediment into Lake St. Lucia would cause infilling of large parts of the lake, resulting in the demise of the aquatic ecosystem (Whitfield and Taylor, 2009).

One of the most noticeable human impacts on the Mkhuze wetland system is the diversion of freshwater via the construction of two canal systems, upstream of the Mkhuze Swamps (Fig.4). The Mpempe canal was constructed in the 1960's in response to hypersaline conditions that developed in Lake St. Lucia as a result of a severe drought (Alexander *et al.*, 1986). The canal was constructed in an effort to increase the freshwater supply to the lake by bypassing the northern portion of the Mkhuze River floodplain, thus shortening the water flow route into Lake St. Lucia (Humphries *et al.*, 2010). The Tshanetshe canal was initially excavated in 1986 by a local farmer clearing a large zone of riparian forest. Over time this canal was subjected to rapid erosion resulting in ~80% of water flow from the northern parts of the floodplain being diverted into this canal (Ellery *et al.*, 2013). Efforts to restore water flow down the original Mkhuze River channel have proven unsuccessful (Ellery *et al.*, 2003).

3.3. Climate

The coastal plain of KZN is characterised by a relatively humid subtropical climate with warm, wet summers and cooler, drier winters (Hunter, 1988). The warm Agulhas Current and Indian Ocean plays an important role in the precipitation occurring along northern KwaZulu-Natal (De Jager and Schulze, 1977). Annual rainfall typically varies between 900 and 1200 mm yr^{-1} and is associated with the Intertropical Convergence Zone (ITCZ) and tropical easterly flow, which transports moisture from the Indian Ocean during the austral summer (Tyson, 1999). The region experiences a 10 year cyclical wet/dry pattern, linked to both the migration of the ITCZ and the El Niño Southern Oscillation (ENSO) (Tyson and Preston-Whyte, 2000; Stretch and Maro, 2013). Heavy rainfall events are usually associated with cut-off low-pressure systems moving northwards up the coast. The region is also affected by tropical cyclones that move through the Mozambique Channel and can cause severe flooding (Hunter, 1988).

3.4. Geological history

The Mkhuze River drains a catchment comprising of various sedimentary strata, including the Dwyka Group, Ecca Group, Lower Beaufort Group, as well as Pongola Supergroup granites and Lebombo rhyolites (McCarthy and Hancox, 2000).

The formation of the Mkhuze wetland has been influenced by periodic sea-level changes since the LGM (~25000 - 17000 yr BP) when sea was lowered to -130 m relative to present day levels (Ramsay, 1995; Tooth and McCarthy, 2007). This drop in sea level resulted in the exposure of the continental shelf, which caused rivers to incise deep valleys into Cretaceous bedrock along the pre-existing tectonic lineament faults (Ramsay and Cooper, 2002). A rise in sea level during the Holocene (~6000 years BP) prompted the blocking of major rivers and caused back flooding of the incised valleys. An outlet at Leven Point was maintained through much of the Late Pleistocene/Holocene transgression (Green, 2009), but ultimately sealed between ~7100 and 6200 cal yr BP (Benallack *et al*., 2016). The erosion and deposition of unconsolidated shelf deposits resulted in the formation of a barrier, which isolated both Lake St Lucia and the Mkhuze River from the ocean. Progradation of the Mkhuze Delta over the past 6500 years is estimated to have occurred at an average of 3 m yr^{-1} (McCarthy and Hancox, 2000). This process is controlled mainly by major flood events, which are capable of transporting large amounts of sediment over vast distances (McCarthy and Hancox, 2000).

Chapter 4: Methods

4.1. Field work

Two continuous 11.5 m parallel sediment cores (MkD-1 and MkD-2) were extracted in April 2015 from the distal end of the Mkhuze bayhead delta (27°53'08.3"S, 32°29'07.0"E; Figs. 5 and 6) using a floating platform and piston coring system (Fig. 5). Water depth at the time of coring was <0.5 m. The MkD-1 core was split longitudinally and logged according to standard sedimentological procedures. One half of the core was sub-sampled for geochemical and sedimentological analyses, the other half was preserved for various non-destructive scanning measurements. The MkD-2 core was split open in a dark room (OSL lab, Wits University) to obtain samples for OSL dating.

Figure 5: Field image showing the coring system used at the core site.

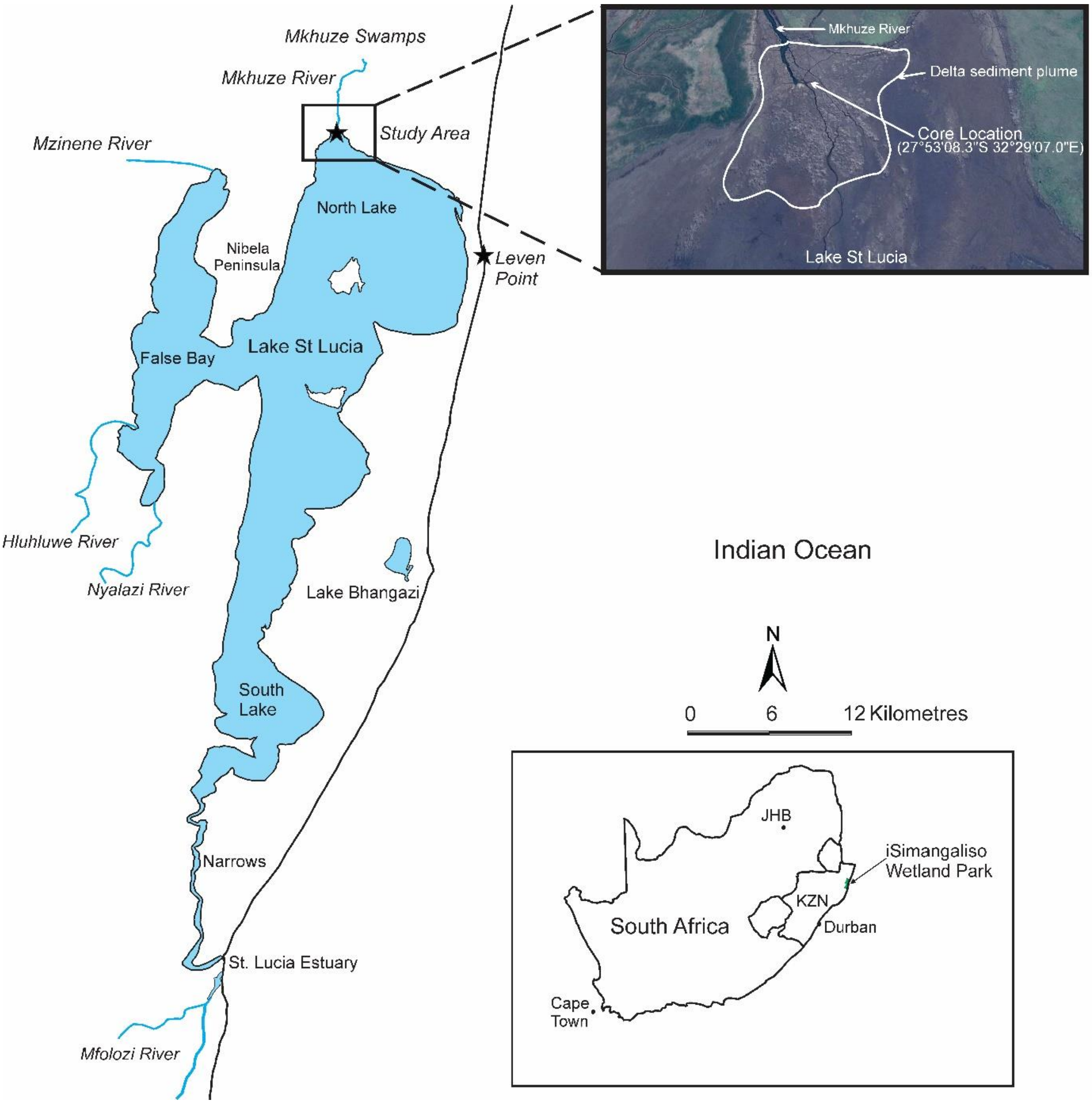


Figure 6: Map indicating the study area and core location.

4.2. Sediment dating and age modelling

Six bulk organic samples were selected for radiocarbon dating using accelerator mass spectrometry (AMS) to provide a down core chronology. A sample was not taken from the upper meter of the profile because of concerns over sediment reworking near the surface. Analyses were carried out by Beta Analytic Incorporated, Florida, USA following standard HCl and NaOH pretreatments. Dates were calibrated using the Southern Hemisphere atmospheric curve SHCal.13 (Hogg *et al.*, 2013). Bayesian age-depth modelling was performed using the Bacon 2.2 source code and the R statistical software program (Blaauw and Christen, 2011).

One sample was collected from a sandy deposit near the base of the core (1120 cm) for OSL dating. This sample contained insufficient organic carbon for AMS dating. The sample was pre-treated with 33% HCl and 20% H_2O_2 to remove organics and carbonate, and then sieved to isolate the quartz grains. The quartz grains were etched with 40% HF for 40 mins and prepared for equivalent-dose determination. Measurements were performed on a Risø TL/OSL Reader, following the Single Aliquot Regenerative (SAR) dose protocol of Murray & Wintle (2003). The luminescence age was obtained by dividing the palaeodose by the mean total dose rate. The error for luminescence age was estimated by combining the systematic and experimental error associated with both the D_e and dose rate values.

4.3. Analysis

4.3.1. Physical analyses

Multi-sensor core logger (MSCL) measurements were performed at 1 cm step intervalsusing a Geotek MSCL at the MARUM Centre for Marine Environmental Sciences (University of Bremen, Germany). Grain size analyses were performed at 5 cm resolution on wet samples at the University of KwaZulu-Natal. Measurements were made using a Malvern Mastersizer 2000 (measuring range: 0.02–2000 µm) following dispersion and sonication in water.

Loss on ignition was measured on subsamples taken at 10 cm intervals. The sub-samples were milled and weighed prior to combustion at 550 ^{o}C. Difference in mass before and after combustion was used to estimate organic carbon content. Where possible, porewater samples were extracted by centrifuging 5 cm^3 sediment sub-samples. Electrical conductivity was measured on collected porewater samples using a Thermoscientific conductivity meter, calibrated against an appropriate standard.

4.3.2. Geochemical analyses

X-ray fluorescence (XRF) core scanning measurements were conducted using an Avaatech XRF scanner at the MARUM Centre for Marine Environmental Sciences (University of Bremen, Germany). The cores underwent sample preparation, through the careful removal of the exposed surface sediment, followed by the placement of a protective film layer.

Following the sample preparation, the cores were measured at a sample resolution of 1 cm. Measurements of the lighter elements (Al, Si, S, K, Ca, Ti, Mn and Fe) were conducted using a generator setting of 10 kV and a current of 0.2 mA, while 30 kV and a current of 1 mA was used for the heavier elements (Br, Rb, Sr and Zr). Sampling time for both runs was 20 s. To calibrate element intensities to concentrations, 59 discrete samples were analysed by energy dispersive X-ray fluorescence (ED-XRF) using a PANanlytical Epsilon 3-XL. Subsamples were dried at 60 ^{o}C, milled and then compressed into discs for analysis. The quality of the measurements was assessed by replicate analysis of USGS certified reference material, Mag-1. Elemental concentrations measured (Si, Ti, Ca, K, Zr and Fe) were within the 95% confidence interval of the certified values.

4.3.3. Minerology

Scanning electron microscope-energy dispersive X-ray (SEM-EDX) analyses were performed at University of the Witwatersrand on four bulk subsamples using an FEI Nova 600 microscope. Samples (430cm, 459 cm, 969 cm and 1120 cm) were selected based on their bulk geochemical composition. The samples were dispersed in water, mounted onto Al stubs and coated with 10 µm Au-Pd.

Chapter 5: Results

5.1. Chronology

The core extracted from the Mkhuze Delta consists mostly of clay and silt dominated sediments underlain by a basal sandy unit comprising medium to fine sand. The six radiocarbon dates obtained from the upper (970 cm) section of the core indicate that the sedimentary infill is stratigraphically consistent (Table 1). The dating of two samples from 854 cm and 970 cm yielded overlapping ages, possibly related to rapid sediment deposition at this time. The OSL date obtained from near the bottom of the core (1125 cm), yielded an age of 13.80 ± 3.55 ka, indicating that the basal sandy deposit is likely late Pleistocene in age.

Table 2: Radiocarbon and calibration data for MkD-1

Lab-Code	Depth (cm)	Elevation relative to MSL (cm)	^{14}C age (yr BP)	95% probability range	Calibrated Age (cal yr BP)
Beta 439022	150	50	1890 +/- 30	Cal AD 80 to 98 (3.8 %) Cal AD 111 to 240 (91.6%)	1775 ± 64
Beta 439023	266	- 66	3020 +/- 30	Cal BC 1371 to 1359 (1.5%) Cal BC 1297 to 1056 (93.9%)	3123 ± 123
Beta 416422	479	- 279	4630 +/- 30	Cal BC 3501 to 3428 (26.4%) Cal BC 3381 to 3317 (40.6%) Cal BC 3237 to 3109 (27.6%)	5299 ± 32
Beta 416423	745	- 545	5420 +/- 30	Cal BC 4333 to 4222 (61.1%) Cal BC 4210 to 4154 (18.45%) Cal BC 4133 to 4062 (15.8%)	6227 ± 55
Beta 416424	854	- 654	6060 +/- 30	Cal BC 5001 to 4826 (92.7%) Cal BC 4818 to 4801 (2.7%)	6863 ± 87
Beta 439024	970	- 770	6010 +/- 30	Cal BC 4946 to 4729 (95.4%)	6787 ± 108

The calibrated radiocarbon ages generally display a linear relationship with depth (Fig. 7). A general decrease in sedimentation rate is observed from 0.25 cm yr^{-1} near the base to ~0.1 cm yr^{-1} near the top of the profile.

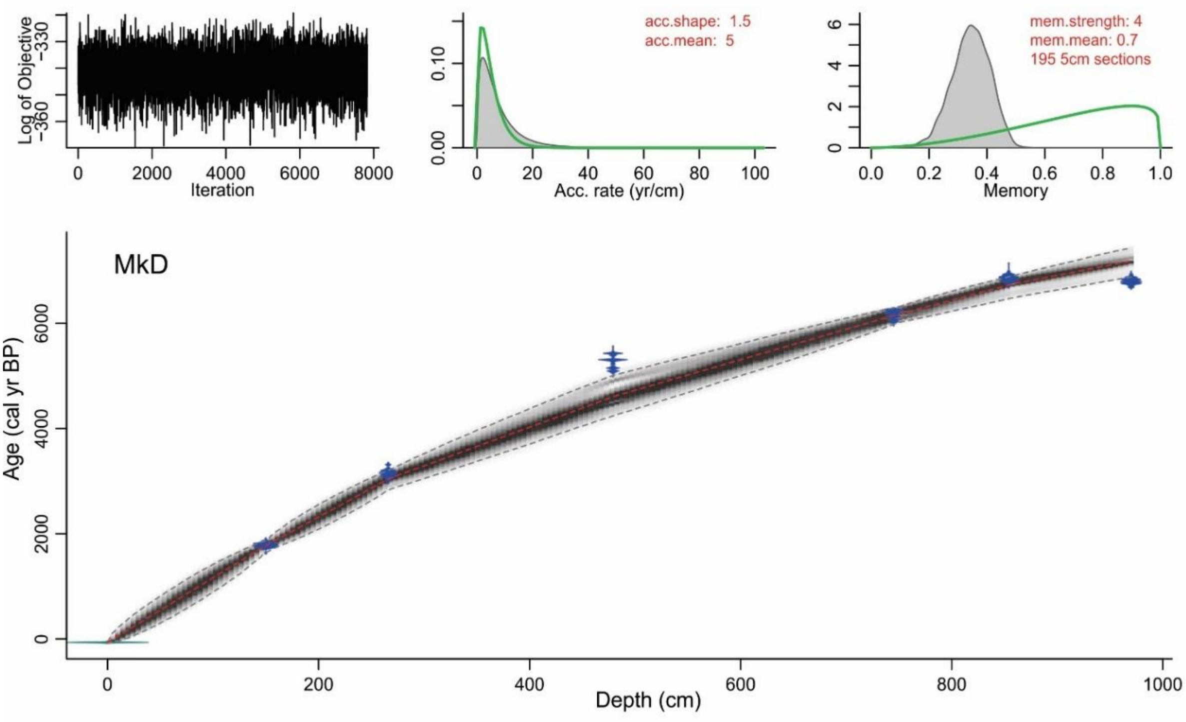


Figure 7: Bayesian age-depth model for MkD-1.

5.2. Core description

The basal unit (1150 – 980 cm) comprises cohesive orange-brown sand, with isolated shell fragments (Figs. 8 and 9). This deposit is sharply overlain by clay and silt dominated sediments that are finer grained and more organic-rich than the lower unit. From 980 cm to the top of the core, a number of shell hash horizons are evident, with significant accumulations occurring between 950 - 830 cm, 570 – 490 cm and150 cm to the top of the core. Shell hash layers are usually associated with lower organic matter contents and an increase in grain size. A number of discrete coarse-grained horizons are seen between 550 cm and 250 cm, and are associated with frequent shell hash accumulations (Fig. 9). The upper 250 cm of the profile is characterised by generally finer material, although variable grain sizes are observed within the uppermost 150 cm. This material contains abundant rootlets and occasional shell fragments.

Porewater electrical conductivity reveals dramatic fluctuations through the core (Fig. 9). Significant enrichments are observed between 960 – 660 cm and 380 – 200 cm.

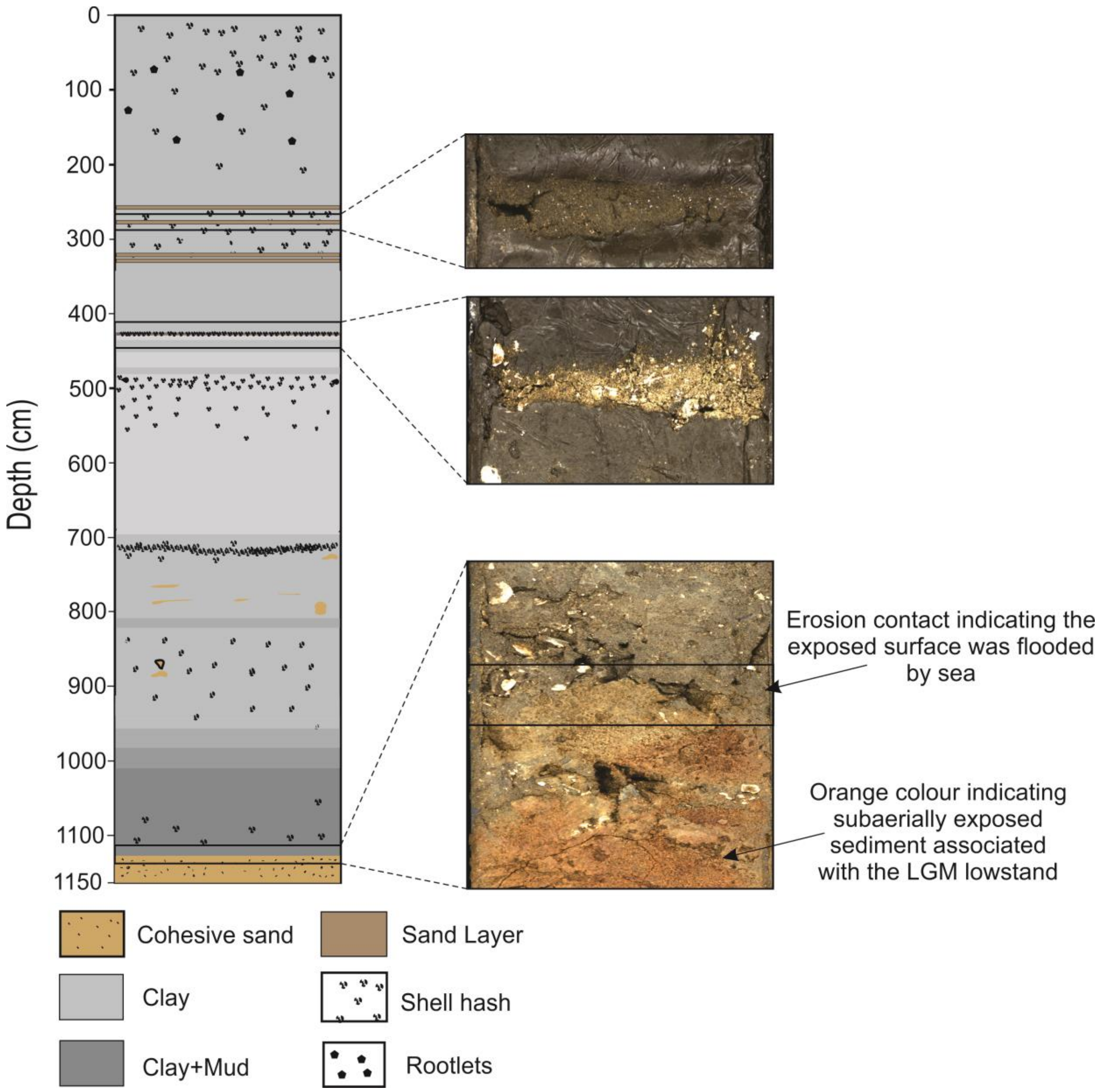


Figure 8: Core log and photographs showing major changes in lithology.

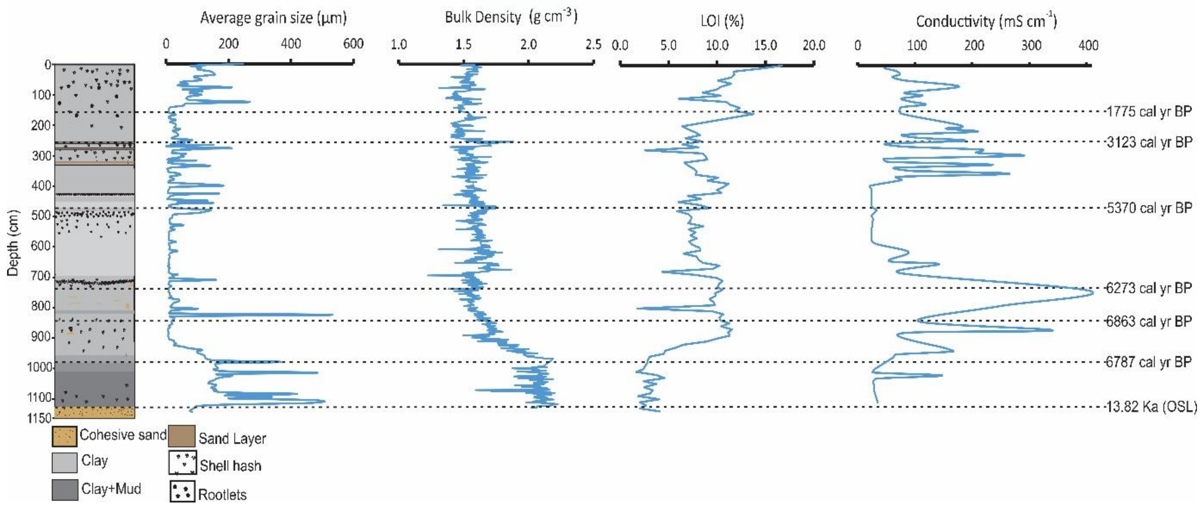


Figure 9: Variation in grain size, bulk density, LOI and porewater conductivity through MkD-1.

5.3. Elemental correlations

Lake sediments contain complex matrices that are subjected to various processes and to avoid artefacts introduced by varying sediment composition, elemental data are compared with a conservative element. Al was chosen as reference element as it is commonly associated with alumino-silicates and is not affected by biological and redox processes (Schropp *et al.*, 1990).

Variations in K, Ti and Rb abundances all show good correlation with Al (Fig. 10), indicating their association mainly with clay minerals and detrital inputs. Relative variations in these elements is thus controlled by silicate sources and reflects the deposition of minerogenic material (Kylander *et al.*, 2011).

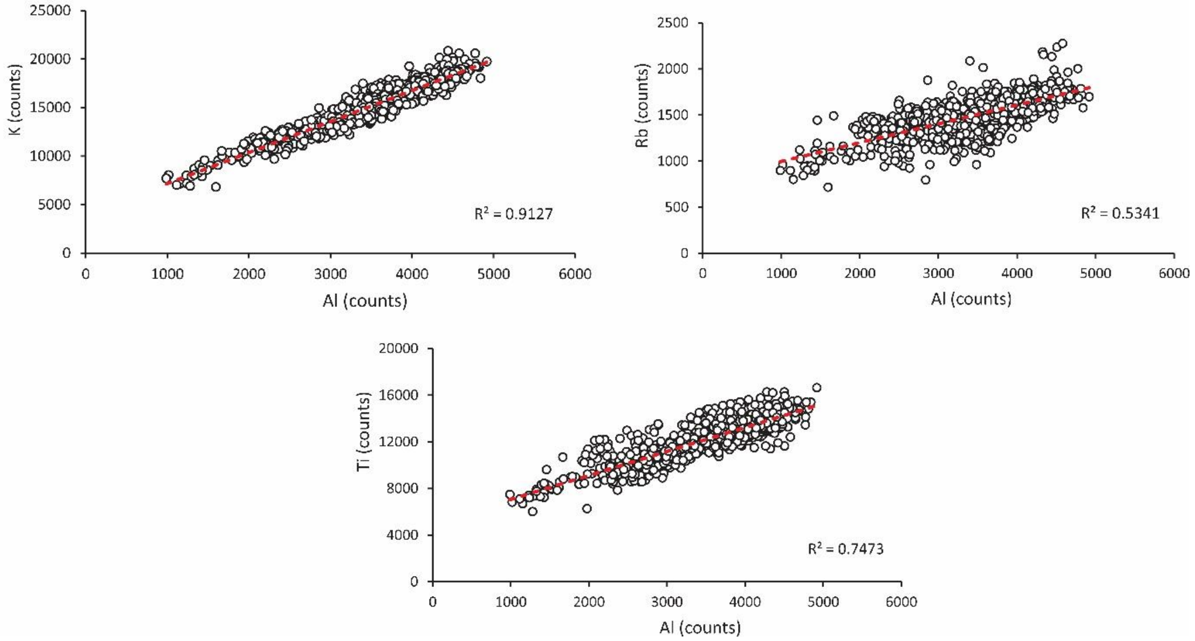


Figure 10: Relationship between Al and selected minerogenic elements.

Plots of S, Fe, Ca and Zr against Al show poor correlation (Fig. 11). Little or no correlation between Al and these elements suggests that their behaviour is not controlled by silicate sources. These chemical components thus likely originate from another source or are introduced via a different process.

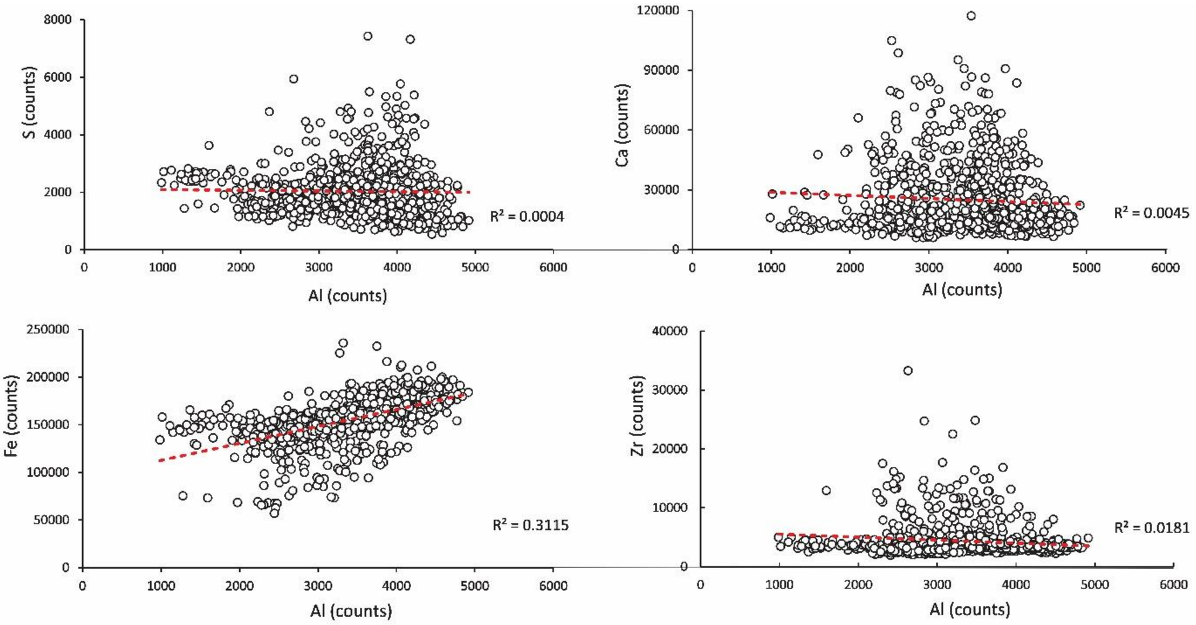


Figure 11: Relationship between Al and S, Ca, Fe and Zr.

5.4. Downcore geochemistry

Down core variations in selected elemental ratios are presented in figures 12 and 13. Variations in minerogenic indicators (Fig. 12) show similar trends. The most notable feature corresponds to the basal sandy deposit (1150 – 980 cm) which shows depletion in Al relative to K, Si and Ti. The material overlying this deposit is characterised by substantially lower ratios, which remain relatively constant through the profile. This material is clearly geochemically distinct from the underlying basal sand unit. Moderate increases in K/Al and Ti/Al ratios, over the uppermost meter suggest an increase in the accumulation of fine material near the surface, while discrete increases in Si/Al (430, 322 and 267 cm) suggest a change in depositional conditions during these periods.

Variations in Ca, Sr, S and Zr ratios (Fig. 13) all show departure from the overall trends observed in Fig. 12. Ca and Sr show similar trends which suggest that carbonate is the dominant phase controlling their abundance. Strong increase in Ca and Sr at 1120 – 1045 cm, 580 – 437 cm, and 380 - 245 cm correspond with shell hash layers identified in the core. Sulphur ratios are variable through the profile, showing periods of strong enrichment at 977, 790, 504 and 48 cm. The Zr/Al ratio indicate that the underlying basal sand is relatively enriched in Zr. The overlying mud is characterised by substantially lower ratios, although notable discrete increases in Zr are observed between 1135 – 985 cm,490 – 430 cm and 337 - 233 cm (Fig. 13).

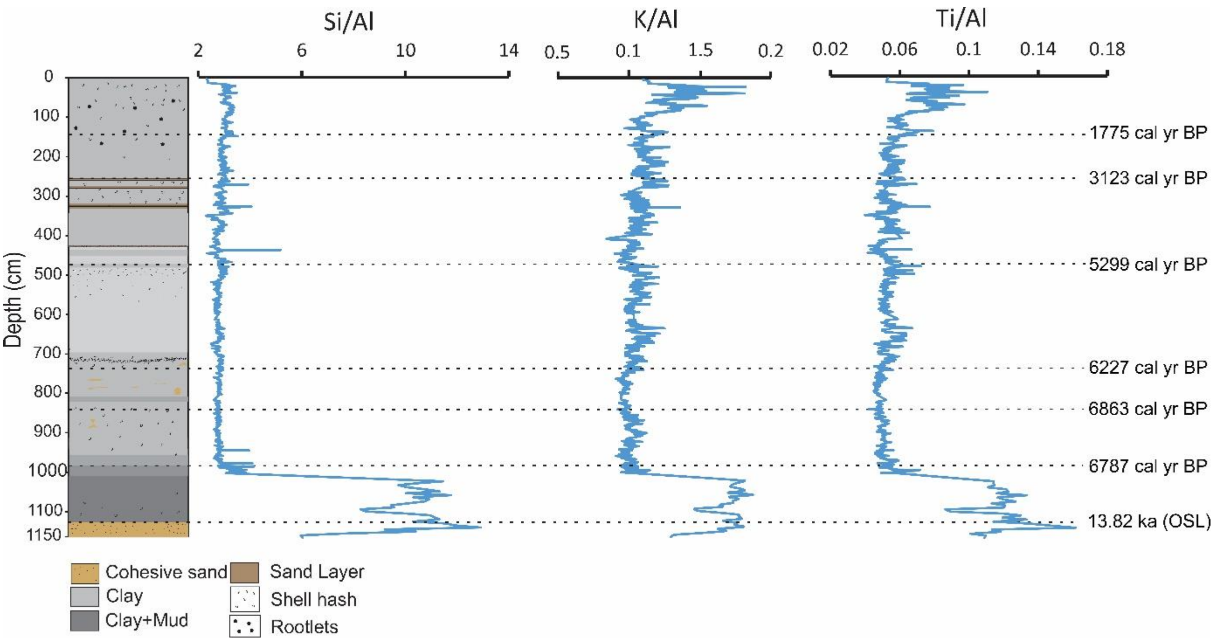


Figure 12: Variations in minerogenic input through the core.

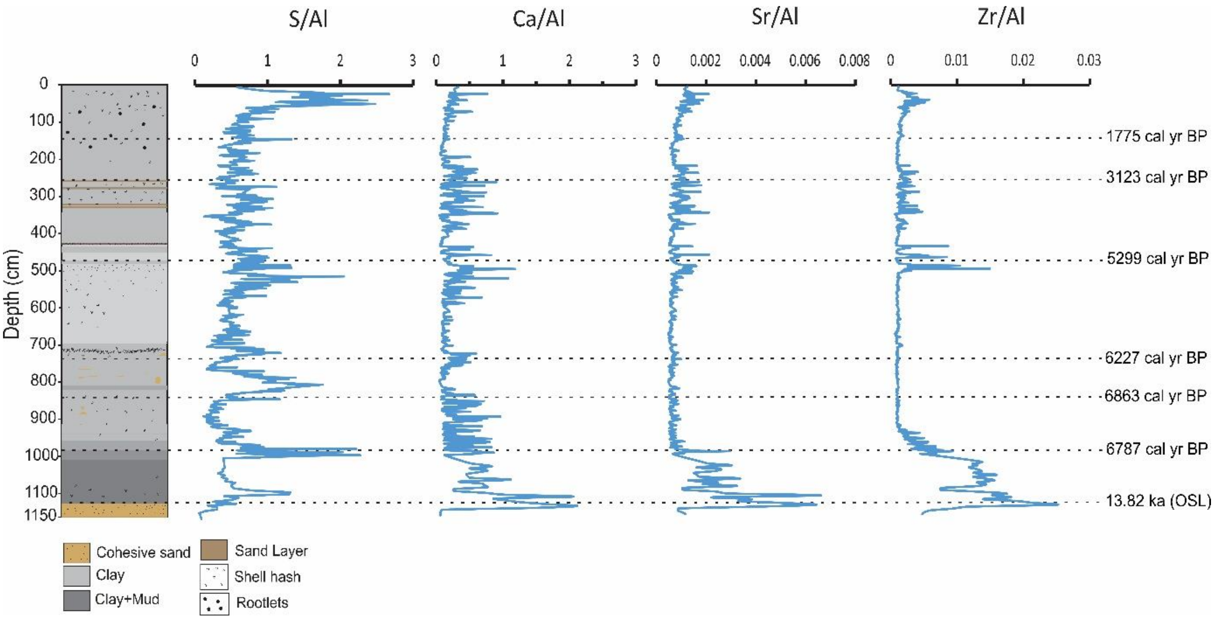
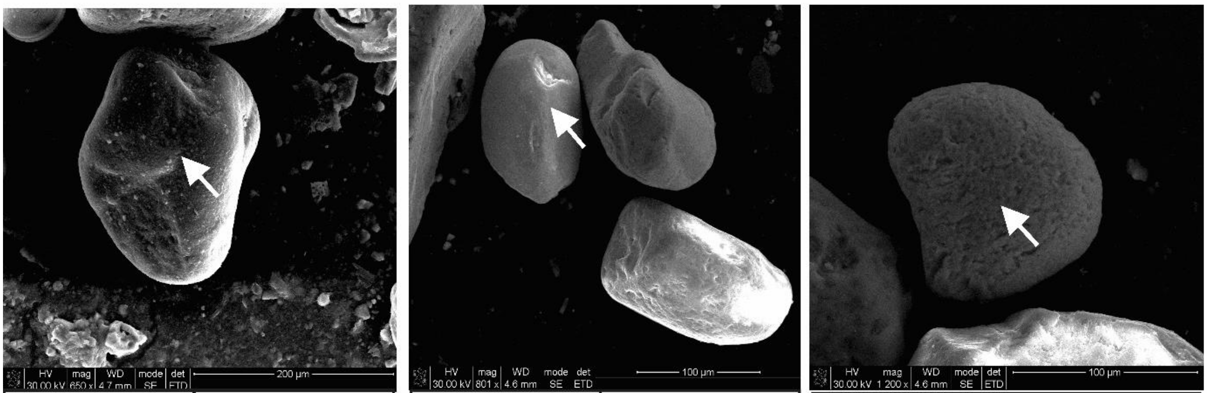


Figure 13: Variations in palaeoenvironmental indicators through the core.

Examples of SEM-photographs along with their geochemical composition are presented in Fig. 14. Samples from 430 cm and 459 cm reveal the presence of large quartz grains (Fig. 14a), along with the presence of zircon (Fig. 14b) and minor quantities of ilmenite (Fig. 14c). The heavy-mineral and quartz grains are well-rounded, indicative of transport under high-energy conditions.



Element	Weight %
O	54.9
Al	4.98
Si	32.66

Element	Weight %
O	32.34
Si	17.16
Zr	49.26

Element	Weight %
O	36.75
Ti	27.89
Fe	32.53

Figure 14: SEM images and EDX results from selected subsamples A) Quartz (4.3 m), B) Zircon (4.59 m) and C) Ilmenite (4.59 m).

Chapter 6: Discussion

6.1. Introduction

Lake St Lucia forms part of a series of coastal lakes found along the eastern coastline of South Africa. These coastal lakes occupy incised valleys that were last exhumed during the LGM and have subsequently been filled by fluvial sediment. The nature of the sedimentary infill is influenced the sediment supply and determined by interactions between local (e.g. fluvial processes), regional (e.g. climate and tidal currents) and global (e.g. sea level) factors. Mkhuze River Delta deposits therefore contain a mix of sediments of different origin that reflect varying depositional processes. Analysis of the sedimentary record obtained from the Mkhuze River Delta thus presents an opportunity to gain insights into these processes, as well as the forcing factors that have shaped the Holocene geomorphic evolution of the delta. In this chapter, geochemical proxies are used in an attempt to understand major influences on sedimentary processes over the past ~7000 cal yr BP.

6.1.1. Depositional energy

Sediment geochemistry is often strongly influenced by both the grain size and particle sorting during deposition (Kylander *et al.*, 2011) and geochemical proxies can be useful tools for characterising depositional energy conditions (Kylander *et al.*, 2011). Zirconium and Si are usually associated with coarser silt and sand size fractions; zircon and quartz, respectively. Because grain size and heavy minerals are known to be good indicators of high-energy depositional conditions, enrichments in Zr and Si can therefore serve as geochemical proxies for increased depositional energy.

In general, there is broad agreement between mean grain size, Si/Al and Zr/Al through MkD-1 (Fig. 15). The presence of coarse material enriched in quartz and zircon prior to ~7200 cal yr BP suggests that this unit was likely deposited under high energy conditions. Sharp declines in grain size, Si/Al and Zr/Al at ~7200 cal yr BP signifies a transition to a more tranquil depositional environment. Intermittent increases in grain size and the presence of multiple clay balls at 6500 cal yr BP suggests that tranquil conditions may have been interrupted by occasional high energy episodes. This is supported by radiocarbon dating which indicates relatively high deposition rates 6200 to 6800 cal yr BP (Fig. 15).

A distinct shift in depositional energy occurs at 4600 cal yr BP, where several discrete increases particularly in grain size and Zr/Al are evident (Fig 15). This points to a period of increased depositional energy between 4600 and 2900 cal yr BP. Thereafter, tranquil conditions return with the last ~2900 years of the record characterised by relatively low Si/Al and Zr/Al ratios. This is supported by a decrease in overall sedimentation rate. Noticeable increases in grain size are evident from ~1700 cal yr BP, which suggest intermittent periods of high energy. Caution, however, should be exercised when interpreting changes over the last 1000 years, as surface sediment may have been subjected to bioturbation and reworking.

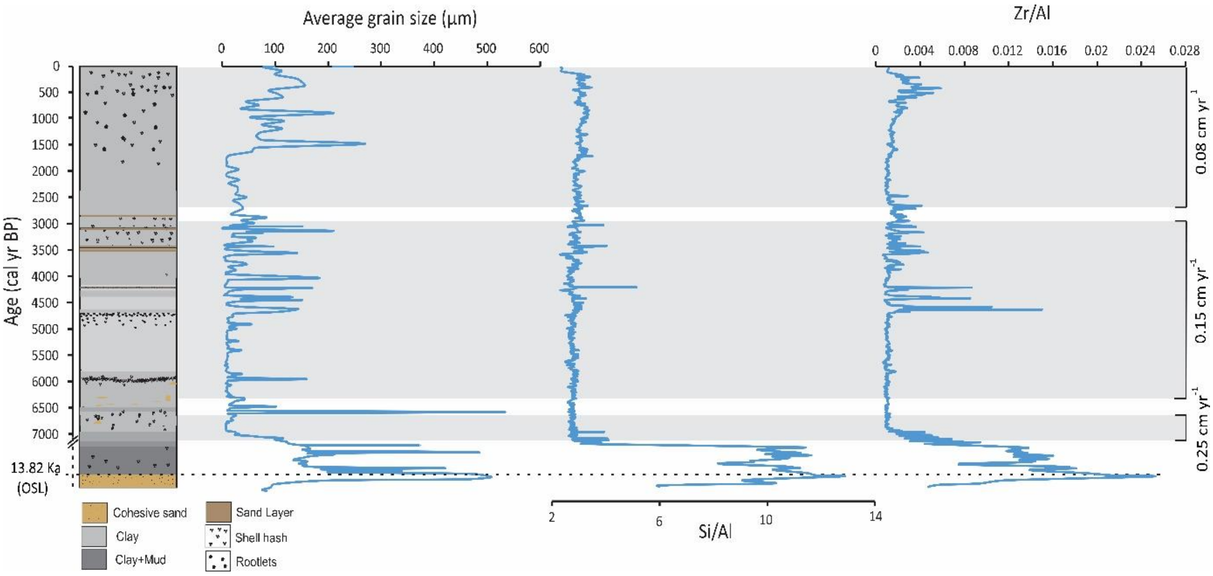


Figure 15: Variations in indicators of depositional energy (grain size, Si/Al and Zr/Al) with age.

6.1.2. Sediment sourcing

Sourcing sedimentary inputs is important in interpreting depositional mechanisms and associated climatic conditions. Due to its environmental setting, the Mkhuze delta has been subject to varying degrees of fluvial sediment supply and marine influence, and therefore has likely received a mix of sediments from different origins.

Grain size analyses of material from MkD-1 reveal contributions from two likely sources; a local sand dune endmember and a fine-grained fluvial end member (Fig. 16). Core material from Zr-enriched horizons contains a coarse-grained component that is characteristic of present-day coastal dune sand. Finer-grained core material, which is not enriched in Zr, displays a grain size distribution that is comparable with Mkhuze River sediment. Figure 17 highlights the contribution from these two end-members. Mkhuze River sediment is characterised by clay/silt dominant sediments and low heavy mineral enrichment, while local dune material is dominated by quartz and is enriched in Zr. This suggests that accumulation of Mkhuze River sediment dominates during tranquil periods, while the local dunes are an important sediment source during high energy events.

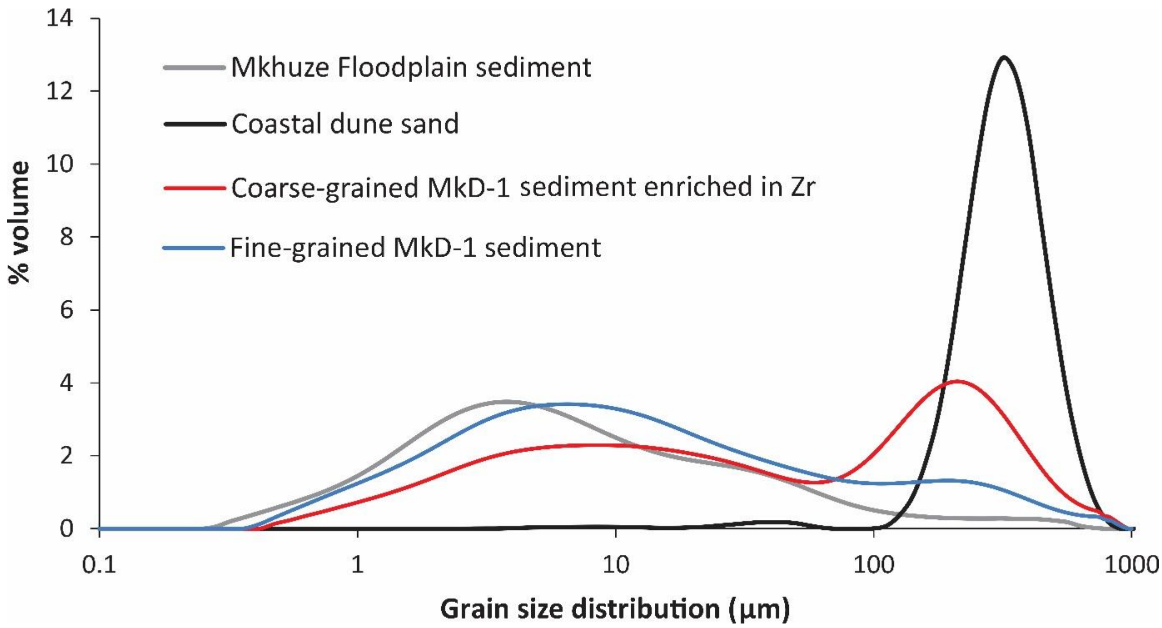


Figure 16: Variation in grain size distribution of MkD-1 sediments representative surrounding dune material and Mkhuze river sediments

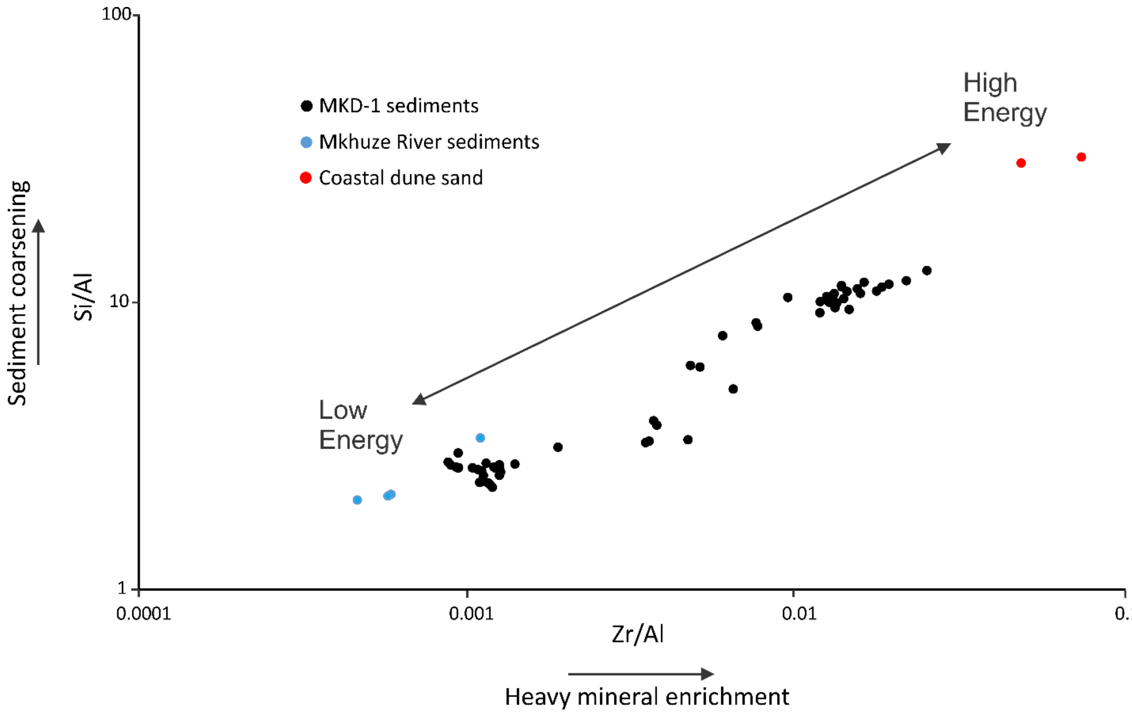


Figure 17: Variation in Zr and Al abundances. The composition of samples from MKD-1 represents the relative mixing of two local end members; dune sand and Mkhuze River sediment.

6.1.3. Marine influences

Sea water is relatively enriched in sulfate compared to freshwater. Seawater inundation results in increased salinity and sulfate concentrations in coastal lakes (Riley and Chester, 2013; Schoepfer, 2013). The S/Al ratio may therefore be a good indicator of marine influence when linked to conductivity. A recent study, using sulfur isotope ratio to determine marine influences at Lake St Lucia has shown that while bacterial sulfate reduction dominates in sediments, there are significant links to marine influences (Gomes *et al.*, submitted).

There are several periods of increased values in the conductivity, Sr/Al and S/Al ratios (Fig. 18). Conductivity appears to have an inversely proportional relationship with the S/Al ratio (7200, 6300 and 4700 cal yr BP). The presence of *Saccostrea forskahlii* shells observed from 6300 – 7000 cal yr BP indicates a marine dominated environment. During this period, the two significant increases in conductivity (6800 and 6200 cal yr BP) may indicate hypersaline conditions. From 6000 – 5000 cal yr BP, the decrease in conductivity and S/Al imply a decrease in marine influences and potential freshwater input. Additionally, the presence of a preserved intact *Paphia textile* shell and *Brachidontes virgilia* in the core imply shallow estuarine conditions. The distinct shell layer at 4100 cal yr BP in a period devoid of other shells (5000 - 4000 cal yr BP) suggest a high energy event which transported this material. From 4000 - 1000 cal yr BP, the variability in conductivity, S/Al and Sr/Al suggest multiple marine influences. A *Soletellina lunulata* valve present at 1100 cal yr BP may indicate shallow estuarine conditions occurred during this period.

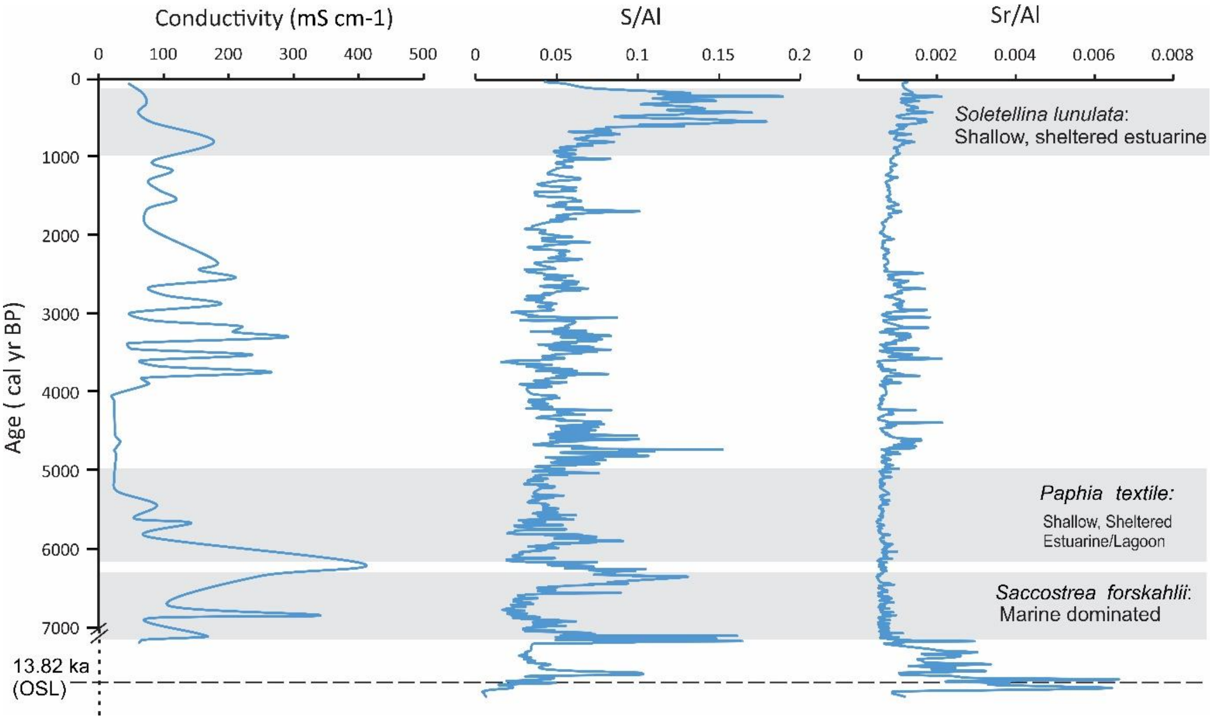


Figure 18: Variations in sediment S/Al, Sr/Al and porewater conductivity vs age.

6.2. Palaeoenvironmental reconstruction

The core extracted from the Mkhuze River Delta captured the entire Holocene sequence and documents sedimentary infilling over the last 7000 years. Variations recorded in geochemical composition reflect regional climate signals that can be used to interpret the influence of palaeoenvironmental changes on the geomorphic evolution of the Mkhuze delta.

6.2.1. Initial marine transgression

Modern coastal sedimentary deposits developed in response to eustatic sea-level rise after the LGM. The Holocene rise in sea-level is clearly recorded within the MkD-1 sequence and indicated by a sharp contact between the upper clay-silt dominated sediments and the underlying late-Pleistocene sands. This contact separates oxidised orange stained basal palaeo-soil with overlying erosional marine mud, which indicates inundation of the surface land after a prolonged period of subaerial exposure. The initiation of Holocene sediment accumulation ~7200 cal yr BP coincides with the stabilisation of sea-level around present day MSL (Fig. 19). Sea-level rise resulted in the back-flooding of the Mkhuze River Delta, creating an environment that favoured the deposition of fine fluvial material. Marine inundation of the Mkhuze River valley is supported by the presence of the intertidal zone species *Saccostrea forskahlii approximately 6200 cal yr BP*, while low energy sedimentation is indicated by sharp decreases in mean grain size, Zr/Al and Sr/Al.

Sea-level rise was also responsible for triggering barrier formation. A sedimentary record from North Lake (NL-1, Fig. 20) documents a shift from marine sand to silt-dominated sedimentation ~6200 cal yr BP (Benallack *et al.*, 2016) in response to barrier development at Leven Point. This restricted tidal flow and promoted the establishment of a low-energy depositional environment, as revealed by the presence of draped, low amplitude seismic reflectors (Benallack *et al*., 2016). The earlier establishment of depositional conditions that favoured the accumulation of Holocene muds at MkD-1 is likely explained by the site's more sheltered position (Fig. 20). The sequence documents a period from 7200 to 4700 cal yr BP that is characterized by the relatively rapid accumulation of fine-grained fluvial sediment, with the presence of intact *Paphia textile* shells indicating the establishment of a shallow, sheltered lagoon (Fig. 18).

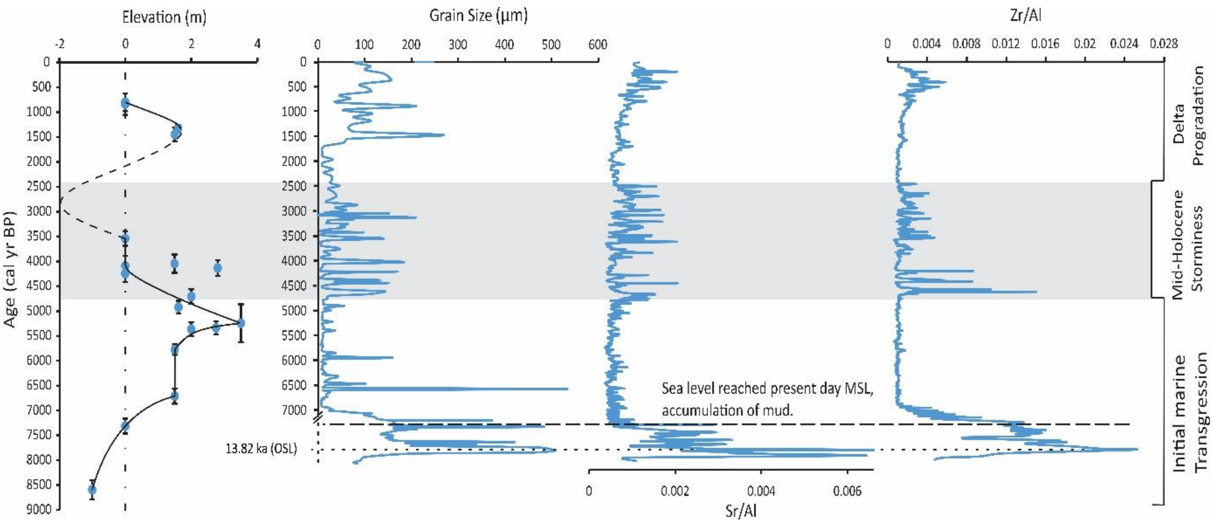


Figure 19: Relationship between reconstructed sea-level and key sedimentological and geochemical indicators from MKD-1. Modified sea level curve from Ramsay (1995) with calibrated age data using ShCal 13.

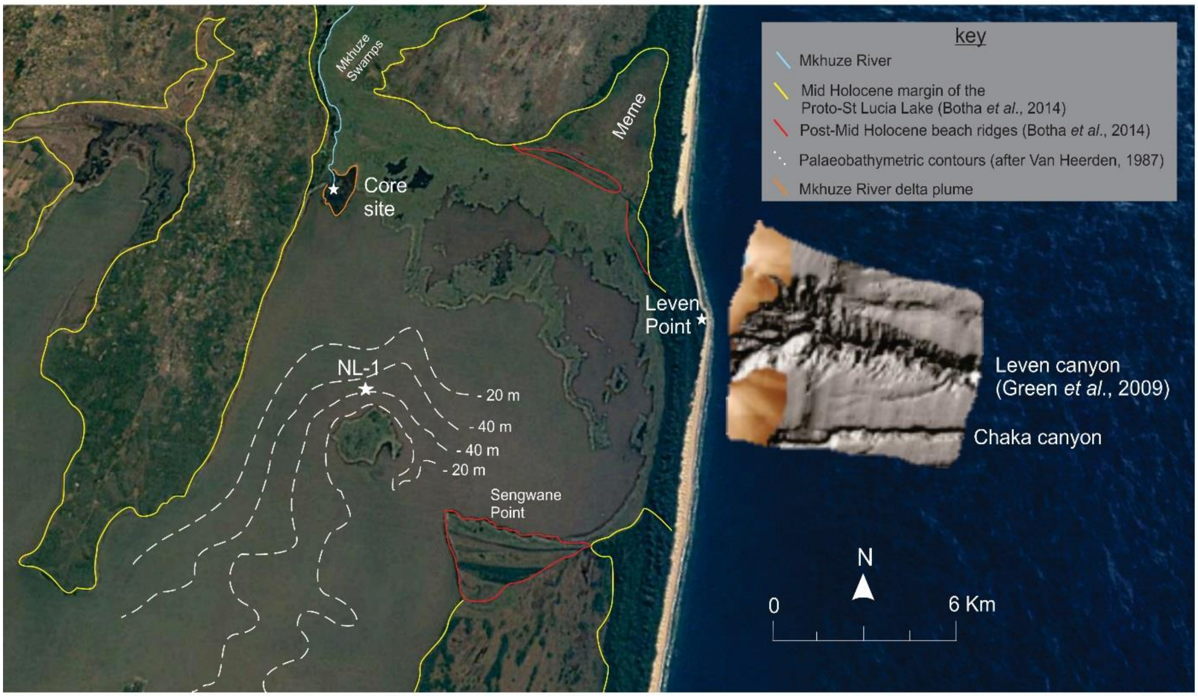


Figure 20: Map showing the inferred palaeo-inlet at Leven point with the position of core sites MkD-1 and NL-1 indicated

6.2.2. Mid-Holocene storminess

From 4600 to 2900 cal yr BP, sediments are notably coarser than the underlying material. The coarsening is accompanied by an enrichment in heavy minerals (indicated by Zr/Al) and shell fragment accumulations (indicated by Sr/Al), consistent with deposition under high-energy conditions. This suggests that barrier stabilization was temporarily interrupted by episodes of enhanced storminess. Evidence of increased storminess during the mid- to late Holocene is found in North Atlantic estuarine and coastal sedimentary archives (Billeaud *et al.*, 2009; Sorrel *et al.*, 2012). Locally, enrichment in sediment $\delta^{34}S$ values and an increase in marine planktonic species 4550-3500 cal yr BP point to intermittent sea water intrusions into North Lake, likely as a result of storm surge events and barrier inundation (Gomes *et al.*, submitted). Collectively, this evidence suggests an increase in regional palaeostorm activity that resulted in the mobilisation of local dune sand. Climatic variability during this period could be related to a strongly positive Indian Ocean dipole anomaly which resulted in warmer sea surface temperatures and an increase in cyclone activity and frequency (Webster *et al.*, 2005).

6.2.3. Delta progradation

From ~2500 cal yr BP, depositional energy declines. This is associated with a decrease in sea level, which Ramsay (1995) inferred from a lack of preserved sea-level indicators. A sequence of sandy beach ridges found at Sengwana and Makatana (Fig. 20) documents St. Lucia lakes' area shrinking in response to declining sea levels, supporting a lower sea level at this time (Botha *et al.*, 2013). OSL dating of the coastal barrier show that the barrier was likely still mobile until ~2000 years ago (Porat and Botha, 2008). Further barrier construction during a final phase of sea-level rise ~1500 ca yr BP was likely responsible for permanently separating North lake from the ocean. Stabilisation of sea level near present day MSL around 900 cal yr BP promoted lateral progradation and a decrease in vertical accumulation rates.

Increases in grain size after 1700 cal yr BP may be associated with the deflation and deposition of coarser sediment under periods of prolonged drought. This is consistent with evidence supporting a significant strengthening in ENSO activity across the Southern Hemisphere (Moy *et al.*, 2002; Gagan *et al.*, 2004; Macreadie *et al.*, 2015). Warm phases are typically associated with drought and wind erosion in eastern South Africa and have been linked to similar desiccation events at Lake St Lucia (Humphries *et al.*, 2016).

Chapter 7: Conclusions

This study highlights the use of geochemical analysis in the reconstruction of environmental changes associated with the evolution of the Mkhuze River Delta. Variations in grain size and geochemical composition document changes in sediment source and particle sorting that reflect changes in the palaeoenvironment. Grain size and high resolution XRF analysis reveal that initiation of modern delta sedimentation occurred ~7200 cal yr BP, when sea-level rise during the Holocene reached present-day levels. Back-flooding of the Mkhuze River valley and subsequent establishment of a proto-barrier in the vicinity of Leven Point promoted the relatively rapid deposition of fine fluvial sediment characterised by low Zr/Al and Si/Al ratios. A shallow, sheltered lagoon is inferred by *Saccostrea forskahlii approximately 6200 cal yr BP* coinciding with the low Si/Al and Zr/Al values. Distinct increases in grain size accompanied by enrichments in zircon (Zr/Al) and shell fragment accumulations (Sr/Al) identifies a period of increased marine palaeostorm activity between 4700 and 2500 cal yr BP. Storm deposits identified within the sedimentary architecture are tentatively linked to a strongly positive Indian Ocean dipole anomaly which resulted in warmer sea surface temperatures and an increase in cyclone activity and frequency. Further barrier accretion and stabilization of sea level promoted lateral delta progradation and transitioning to a lacustrine environment. Subtle changes in grain size over the last ~1700 cal yr BP are linked to periods of prolonged drought that is consistent with evidence supporting a strengthening in ENSO activity.

The core from the Mkhuze River Delta provides unique insight into the global (sea level) and regional (palaeostorm activity and ENSO) forcing factors that have influenced sedimentation and the evolution of Lake St Lucia. Understanding past changes in environmental conditions may help in evaluating the response of coastal systems to future environmental stressors, while knowledge of the long-term functioning of coastal systems enables better management strategies to be developed. Recommended future work could involve confirming some of the interpretations made in this study using other proxies (e.g. diatoms, foraminifera and biomarkers).

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