



# Geology Honours Project

The effects of weathering on coal quality in the Limpopo and Soutpansberg Coalfields.

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## **Abstract**

The petrographic and chemical properties of weathered coals and their fresh counterparts were investigated using stored coal from the Limpopo Coalfield (Vele Colliery) and the Soutpansberg Coalfield (Makhado Project) of South Africa. The coal quality was determined through, proximate analysis, ultimate analysis and x-ray fluorescence (XRF). The petrographic characteristics of the coals were determined through routine vitrinite reflectance and a maceral point count. In addition, the abnormal condition analysis (ACA), the chemical index of alteration (CIA) and the chemical index of weathering (CIW) were used to assess the extent of weathering on a petrographic and chemical basis, respectively.

The proximate analysis and ultimate analysis yielded variable results, however, based on the major oxide chemistry determined by XRF, the majority of the coals from both localities were classified as moderately weathered on the CIA and CIW. As a consequence of the short duration of exposure to secondary weathering (less than two years for coals from the Limpopo Coalfield and approximately 5 years for coals from the Soutpansberg Coalfield, extreme weathering features such as well-developed oxidation rims, devolatilization pores and plastic deformation were not detected through the ACA. On average, fresh organics and fresh minerals constituted 94% of the coal, whereas early stage weathering features only accounted for 6% of the coal. At low levels of weathering the free swelling index (FSI) was the most sensitive parameter to weathering as values were comparatively reduced.

Although signs of weathering were detected through the analytical techniques, no definitive correlations could be made between the observations and the external and internal factors affecting weathering such as the initial base line quality of the coal when it was initially exposed to the atmosphere during mining, the effect of particle size, and the role of ambient weather conditions (temperature and solar radiation) during outdoor storage. Therefore, in order to better constrain the development of weathering features during the early stages of outdoor storage, a follow up study incorporating these factors is required to better understand and characterise early-stage weathering features.

**Keywords:** Secondary weathering, coal quality, low temperature oxidation, weathered coal, fresh coal

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## List of Acronyms

as:	As received
ad:	Air dried
BM:	Bottom Middle
CIA:	Chemical Index of Alteration
CIW:	Chemical Index of Weathering
CoAL:	Coal of Africa Limited
FGD:	Flue-gas Desulphurisation
HCC:	Hard Coking Coal
inc. mm:	Including mineral matter
MCZ:	Main Coal Zone
MKB:	Main Karoo Basin
MS:	Middle Seam
NAR:	Net as received
ROM:	Run of mine
TL:	Top Lower
TM:	Top Middle
vol%:	Volume percent
wt%:	Weight percentage

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## 1. Introduction

Weathering is a physical and chemical process that breaks down rocks into their constituents (Marchioni, 1983). In the context of coal, weathering has great implications on the quality of the end products that are used in various coal markets (Steyn and Minnitt, 2010; Taole, 2014; Stefaniak *et al.*, 2015). Weathered coals are generally perceived to be of low quality and are therefore often overlooked and not beneficiated (Belaid *et al.*, 2013). This has led to the accumulation of up to 1.5 billion tonnes of discarded coals in dumps throughout South Africa; in addition, up to 60% of the pillar coals remaining in the Witbank and Highveld coalfields are classified as weathered (de Korte, 2001; Belaid *et al.*, 2013; Jeffrey *et al.*, 2014). These constitute the bulk of South Africa's weathered coal resources (Belaid *et al.*, 2013). In most cases coal is stored outdoors in stockpiles for varying periods of time which expose the coal to the processes of weathering (Wagner, 2007). Weathered coals have the potential to be beneficiated to produce suitable coal products primarily for use at Eskom's power plants (Mahr, 2010; Jeffrey *et al.*, 2014). This will be beneficial to local markets; furthermore, the utilization of weathered coals will reduce the environmental degradation incurred by coal dumps and stockpiles (Reddick *et al.*, 2007; Eberhard, 2011; Belaid *et al.*, 2013).

### 1.1 Coal quality and weathering

Coal is a combustible rock that consists of organic and inorganic matter (Orem and Finkelman, 2003). The organic matter (macerals) are derived from plants whereas the inorganic matter (mineral matter) are derived from the environment in which the coals form (Schweinfurth, 2009). Macerals are the organic remains of plants that are preserved during coalification (coal formation), thus they are useful in identifying the plant species from which the coals are derived (Suárez-Ruiz and Ward, 2008). *Glossopteris* and *Gangamopteris* were the dominant flora in the Karoo Basin during the Permian period and are the main parent material that gave rise to South African coals (Catuneanu *et al.*, 2005; Hancox and Götz, 2014). Coal forms in swampy environments that are characterised by thick vegetation cover, relatively stagnant waters, and anoxic conditions (reducing environment) where the accumulation of plant matter (peat) is faster than the rate of biological and chemical decay (Orem and Finkelman, 2003). These conditions allow peat to accumulate through geological time (~0-1 m per Millennia) without major disturbance by sediment deposition (Tatsch, 1980). The depth of burial and the temperature and pressure conditions affecting peat through time, governs the rank of the resulting coals (Orem and Finkelman, 2003). With increasing temperature and pressure peat progresses from low rank coal, lignite, to high rank coals, meta-anthracites (Tatsch, 1980). Thus the geological environment in which

coal is deposited and lithified determines the initial coal quality (Li *et al.*, 2014). The coal quality can be subsequently affected by weathering and igneous intrusions that increase the temperature of the coals in close proximity of the contact aureole, resulting in devolatilised coals or increase in coal rank (Snyman and Barclay, 1989).

Temperature, water and oxygen content are the major agents affecting the weathering of coal (Cox and Nelson, 1984; Mangena and Du Cann, 2007). When coal seams are exposed at the surface by uplift and erosion, they are subjected to greater fluctuations in temperature, oxygen and moisture content (Snyman and Barclay, 1989; Crelling *et al.*, 1979; Banerjeer *et al.*, 2000). These changes that occur *in situ* are referred to as primary weathering or oxidation, and work together to break down the complex physical and chemical structure of coal (Mangena and Du Cann, 2007; Wagner, 2007). The effect of weathering on *in situ* coal is often minimal however weathering is accelerated during mining and stockpiling of the coal; this is known as secondary weathering or oxidation (Wagner, 2007; Jha *et al.*, 2014). Coal is commonly stored in stockpiles for long periods under various conditions which accelerate the rate of weathering and often lead to spontaneous combustion (Cox and Nelson, 1984; Banerjeer *et al.*, 2000; Misz-Kennan and Fabiańska, 2011). Weathered coals also contribute to environmental pollution, because the by-products of weathered coals in dumps can contaminate the surrounding ground water and soils (Misz-Kennan and Fabiańska, 2011).

## **1.2 The significance of coal quality in South Africa**

Coal quality is increasingly becoming a global issue especially for economies that predominately rely on coal-based energy production (Mahr, 2010; Kinghorn, 2013). South Africa is no exception, more so because it is amongst some of the major coal producing countries who have adopted the practice of exporting their high grade coals, whilst using lower grade coals (35-45% average ash content) to generate electricity locally (Jeffrey, 2005; Nkomo, 2005). Although this practice generates much needed revenue, it exacerbates environmental pollution and creates coal supply shortages for the South African State energy-generating company, Eskom (Nkomo, 2005; Sebitosi, 2008; Jeffrey *et al.*, 2014; Jurgens, 2015).

Optimal energy generation requires good coal quality and efficient technologies (Mahr, 2010; Kinghorn, 2013). One of the major issues affecting Eskom's power supply is its out-dated technologies which produce less energy and consume more coal (Mahr, 2010; Jeffrey *et al.*, 2014). Pulverised fuel beds are the older forms of technologies used to generate electricity in South Africa but, they are unable to accommodate a wider range of coal qualities compared to the newer circulating fluidised beds

(Eberhard, 2011; Jeffrey *et al.*, 2014). All of Eskom's existing 13 coal-fired power stations utilise pulverised fuel bed technology; it is only the newly constructed Medupi and Kusile Power Stations that have installed Flue-gas desulphurisation (FGD) systems that are not only more efficient, but also remove sulphur and carbon emissions (Eskom, 2011; Jeffrey *et al.*, 2014). Approximately R 1 billion is lost yearly at Eskom's Duvha, Matla, and Tututka Power Stations due to the use of low quality coal which reduced their operational capacities (Eberhard, 2011).

The quality of coal and the retail value can be improved to a certain extent by beneficiation; however, low rank coals require more rigorous cleaning at higher operating costs that are usually associated with high water consumption (Cox and Nelson, 1984; Mahr, 2010). Most beneficiation methods in South Africa are wet based and require significant amounts of water; this will prove to be problematic for coalfields in water-stressed regions such as the Limpopo Province (Jeffrey, 2005; Jeffrey *et al.*, 2014). In spite of these challenges, a cleaner production assessment of the ultra-fine coal waste generated in the Witbank Coalfield proved that the wastes can be successfully improved and sold with substantial economic gains (Reddick *et al.*, 2007).

Dry coal beneficiation is not a novel technology in the coal industry, yet it is not widely used in South Africa nor in other coal producing countries compared to China, Germany and India (Dwari and Rao, 2007; de Korte, 2013). In South Africa, recent testing of dry coal beneficiation technologies has been chiefly conducted by Coaltech Research Association (de Korte 2010; Von Ketelhodt and Bergmann, 2010; de Korte, 2013; de Korte 2015). These studies show that the Fuhe Ganfa Xuan mei (FGX), which translates to "compound dry type coal washer", and the X-ray transmission (XRT) sorters are able to clean low quality coals at economically feasible rates in the short term, however for long term coal processing, the use of dense medium beneficiation remains more profitable due to a higher product yield, albeit at the expense of the environment (de Korte 2010, de Korte, 2013; de Korte 2014; de Korte 2015). Because the efficiency of the FGX and XRT sorters greatly depend on the physical properties of coal, the petrographic and geochemical properties of the weathered coals are important in assisting the prescription of a suitable beneficiation technique (Dwari and Rao, 2007; Von Ketelhodt and Bergmann, 2010). A comparison of the performance results of the FGX and XRT are contrasted in Table 1. Details of the experiment can be accessed from de Korte, (2013).

Table 1. Beneficiation efficiency of the FGX sorter versus XRT sorter (de Korte, 2013)

Parameter	FGX	XRT
Feed % Ash	40.4	71.0
Product % Ash	31.9	59.5
Discard % Ash	60.2	81.4
Product Yield %	70.08	47.58
D50 cut-point RD	2.007	2.062
EPM	0.2168	0.2878
Organic Efficiency %	86.8	79.4
Sink in float %	6.78	27.54
Float in sink %	10.94	3.83
Total misplaced %	17.73	31.37
Near-dense material	8.4	1.9

In 2005 it was reported that both the Witbank and Highveld Coalfields only had 9 billion tons of recoverable resources remaining (Jeffrey, 2005). Thus, less developed coalfields such as the Waterberg, Limpopo, and Soutpansberg will become important coal suppliers as early as 2020 (Jeffrey, 2005; Hancox and Götz, 2014). South Africa has, however, considerable amounts of weathered coals in the form of pillar coals, discards coals, long term coal stockpiles, ultra-fine coals and weathered coal zones that can be reworked (de Korte 2001; Belaid *et al.*, 2013). Discard coals are by-products of beneficiation that are considered not suitable for trade, whereas pillar coals are those that have been left as support structures during board and pillar mining operations (de Korte, 2001; Jeffrey *et al.*, 2014). These weathered coals can be reworked and beneficiated to supply much needed coal for electricity generation, however this idea has been met with some scepticism due to the high initial costs associated with the beneficiation of low quality coal and the lower efficiency of dry beneficiation methods which result in a lower product yield compared to traditional wet beneficiation (Lloyd, 2000; Belaid *et al.*, 2013; de Korte, 2013).

## 1.3 The Geology of the Limpopo and Soutpansberg Coalfields

South African coals were deposited during the Permian period and are hosted in the most extensive stratigraphic unit in the country, the Karoo Supergroup (Cadle *et al.*, 1993). The Karoo Supergroup was deposited in a retroarc foreland basin referred to as the Main Karoo Basin (MKB), and comprises sediments derived from glacial, deltaic and fluvial systems (Catuneanu *et al.*, 2005). The sedimentary units indicate progressive climatic warming, whereby glacial conditions prevailed south of the Kaapvaal Craton during the early Permian, and is represented by Dwyka tillites, whereas aeolian sediments (Stormberg Group) were deposited in the late Permian under semi-arid conditions (Catuneanu *et al.*, 2005; Hatton and Fardell, 2012). Sedimentation in the Karoo Basin was effectively terminated due to the rifting of Gondwana which released large volumes of continental flood basalts that intruded into the Karoo sequence as dykes and sills (Catuneanu *et al.*, 2005). The Limpopo and Soutpansberg Coalfields occur in smaller satellite basins of Karoo age, which were formed by a similar tectonic regime to the MKB (Catuneanu *et al.*, 2005; Hancox and Götz, 2014).

### 1.3.1 Limpopo (Tuli) Coalfield

The Limpopo Coalfield is also known as the Tuli Coalfield and occurs within the Tuli Basin (Catuneanu *et al.*, 2005). The Tuli Basin covers an area of 1000 km<sup>2</sup> in South Africa; however it covers a greater spatial extent in Botswana and Zimbabwe (Malaza, 2013). The Tuli Basin is displaced by half graben structures which are separated by the Beit-Bridge Complex (Figure 1). The Tuli Coalfield comprises of Karoo-age sediments namely; Clarens, Bosbokpoort, Klopperfontien, Solitude, Fripp, Mikambeni, Madzaringwe and Tshidzi Formations (Table 2). These sediments unconformably overly the Limpopo Mobile Belt (LMB) and are displaced by the half graben structures (Ortlepp, 1986). The Sediments are typically discontinuous and have a considerably lower maximum thickness ranging between 450-500m compared to the MKB which has a maximum thickness of 6000m (Bordy and Catuneanu, 2002). Dolerite and porphyritic andesite intrusions aged 150Ma are common and have affected the coalfield to variable degrees (Snyman and Barclay, 1989). The lithologies at Vele Colliery have been locally affected by a 15m thick dolerite intrusion occurring with smaller subsidiary dykes (Hancox and Götz, 2014).

Coal seams in the Tuli Basin are distributed between the local equivalents of the Ecca and Beaufort Groups, namely the Tshidzi through to the Clarens Formation (Table 2). The Main Coal Zone (MCZ) is 15 m thick and lies within the Madzaringwe Formation (Hancox and Götz, 2014). The Madzaringwe Formation is approximately 120 m thick and consists of feldspathic, cross-stratified sandstones, siltstone and shale with narrow coal seams (Hancox and Götz, 2014). The nomenclature used by Coal of Africa Limited (CoAL) for the subdivisions of the MCZ at Vele Colliery are as follows; Bottom, Middle and Top Seam Coal (Hancox and Götz, 2014). The Top

and Bottom Seams are further divided into the Bottom Lower (BL), Bottom Upper (BU), Top Lower (TL), Top Middle (TM) and Top Upper (TU) (Hancox and Götz, 2014). The seams are near horizontal and dip slightly at 1-2° N-NE, and consist of interlaminated carbonaceous mudstones and vitrinite-rich coal (Hancox and Götz, 2014). The Middle seam is of principal economic interest because it is the thickest seam ranging between 3-5 m and contains the highest proportions (20 - 45%) of coal (Hancox and Götz, 2014).

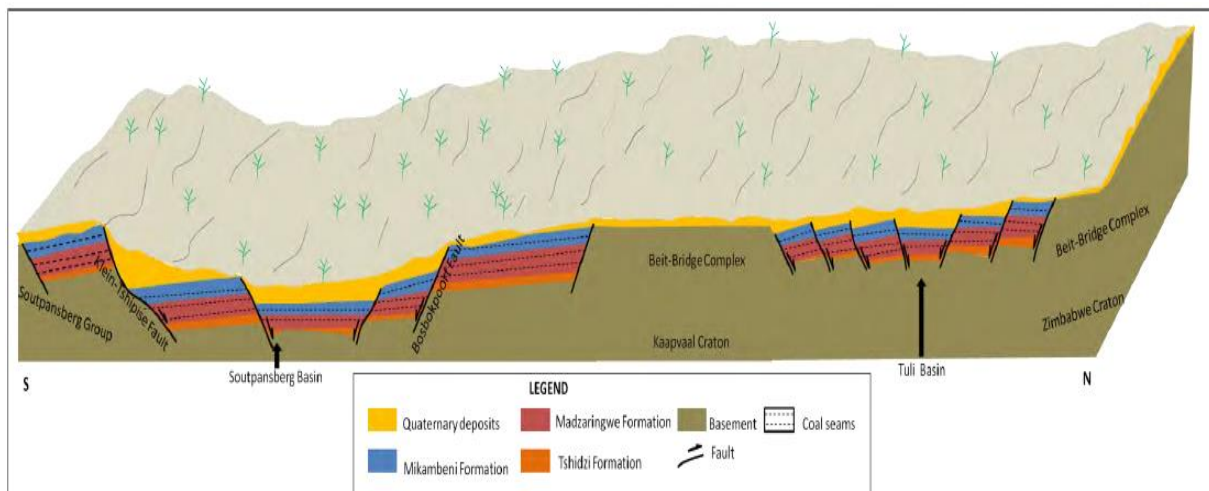


Figure 1. Basin model of the Tuli and Soutpansberg Basins, not to scale (Malaza, 2013)

Table 2. Stratigraphy of the Karoo Supergroup in the Limpopo and Soutpansberg Coalfields after Malaza, (2013)

Formation		Thickness	Lithology
Clarens	Tshipise Member	5-140 m	Fine to very fine-grained, khaki to yellowish sandstones
	Red Rocks Member	~60 m	Very-fine to fine-grained, pinkish to red, argillaceous sandstones
Boshbokpoort		60 m	Red to purple mudstones, minor white siltstone layers and some occasional conglomerates
Klopperfontien		10-20 m	Coarse- grained sandstone and minor conglomerate
Solitude		3.5-170 m	Shallow cross-laminated siltstones and very fine sandstones with grey mudstones
Fripp		5-110 m	Well-sorted, medium- to coarse-grained, white, arkoses, sandstone and course conglomerates
Mikambeni		20-150 m	Alternating black shale, sandstone and coal
Madzaringwe		120-200 m	Alternating feldspathic, cross-stratified sandstone, and siltstone and shale with narrow coal seams
Tshidzi		5-20 m	Diamictite
Limpopo Mobile Belt (LMB)/Beit-Bridge Complex			

### **1.3.2 The Soutpansberg Coalfield**

The Soutpansberg Coalfield trends parallel (E-W) with respect to the Tuli Coalfield and extends from Waterpoort to the Kruger National Park (Malaza, 2013). The three major subdivisions of the Soutpansberg Coalfield are shown in Figure 2 and are referred to as the Mopane, Tshipise and Pafuri sub-basins (Jeffery, 2005). Makhado Project is situated within the Tshipise sub-basin.

The Tshipise sub-basin is centrally located within the Soutpansberg Coalfield and is also structurally controlled by half graben structures that are separated by the Beit-Bridge Complex (Figure 1). The major pre-Karoo faults affecting the Soutpansberg Coalfield trend parallel to the strike (SW-NE) of the LMB and are known as the Tshipise, Klein Tshipise and Bosbokpoort faults (Figure 1) (Malaza, 2013). Coal seams occur in the Madzaringwe and Mikambeni Formation (Table 2).

## **1.4 Current research on weathering and coal quality**

Research on coal quality has long been evolving as an important field of investigation. Early studies presented the fundamentals of how coals weather and the relevance of the effects of weathering on the technological properties of coal (Crelling *et al.*, 1979; Cox and Nelson, 1984). Since then, many weathering and coal quality studies have been done on coals from various regions around the world (Alvarez *et al.*, 1998; Banerjee *et al.*, 2000; Nzoussi-Mbassani *et al.*, 2005; Heriawan and Koike, 2015; Stefaniak *et al.*, 2015; Tamamura *et al.*, 2015). Most of the literature focuses on the behaviour of weathered bituminous coals and coking coals for different technological applications (Montgomery, 1978; Banerjee *et al.*, 2000; Díez *et al.*, 2002; Mangena and Du Cann, 2007; Jha *et al.*, 2015).

As is the case for most coal research in South Africa, more studies on weathered coals have been conducted on coals from the Witbank, Highveld and Ermelo Coalfields (de Korte, 2001; Reddick *et al.*, 2007; Wagner, 2007; Dey *et al.*, 2013; Taole, 2014). These studies focus on investigating the industrial potential and behaviour of the weathered coal. No previous work has been done to document the changes that coal undergoes over time due to weathering in the Soutpansberg and Limpopo Coalfields. Although investigation of coal in the Soutpansberg and Limpopo Coalfields commenced as early as the 1800s and in 1885 respectively, slow progress has been made in terms of research, thus more work remains to be done in the area (Hancox and Götz, 2014). Results from Cindi (2014) indicate that coal quality increases from west to east in the Soutpansberg Coalfield, as coals in the Tshipise sub-basin (west) have lower rank, CV and carbon content with respect to the Pafuri sub-basin which is located east of the Soutpansberg Coalfield. It can be argued that the increase in coal quality may be attributed to the presence of the Klein Tshipise Fault, a boundary between the Tshipise and Pafuri sub-basins, because it formed after the coals were deposited (Malaza, 2013). Mahlangu (2014) classified the coals

at Vele Colliery as medium rank C/B bituminous coal, however the study was limited to vitrinite and maceral analyses and it was therefore recommended that a detailed geochemical investigation involving XRF, proximate analysis, ultimate analysis, and CV analysis be conducted. This study encompasses these methods and will therefore contribute towards a better understanding of the characteristics of weathered coals in the Northern Coalfields.

## **1.5 Problem Statement, Aims and Objectives**

In this study it is hypothesized that the extent of weathering diminishes the quality (and uses) of coal; characterising the effects of weathering may inform the beneficiation of such coal and improved beneficiation could render this coal useful. Therefore, the aim of this study is to investigate the effects of weathering on coal quality in the Limpopo and Soutpansberg Coalfields.

The specific objectives were to:

- a. Determine the geochemical and petrographic properties of the freshest to the most weathered coal; and
- b. Determine the potential use(s) of these coals based on their properties.

## **2. Materials and Methods**

### **2.1 Location of the research area**

Samples used in this study were collected from CoAL's Makhado Project and Vele Colliery which are located in the Limpopo Province of South Africa (Figure 2). Makhado Project is located 65km southwest of Musina whereas Vele Colliery is located approximately 48km northwest of Musina in the extreme northern edge of the Limpopo Province (CoAL, 2015). Both localities belong to the Vhembe Municipality which is characterised by semi-arid conditions in which extreme temperatures greater than 35°C can occur (Mpandeli, 2014). Rainfall in the district is variable such that the area around Makahdo receives 500-800mm mean annual rainfall whereas the area where Vele Colliery is located experiences far less mean annual rainfall of 300mm (Mpandeli, 2014).

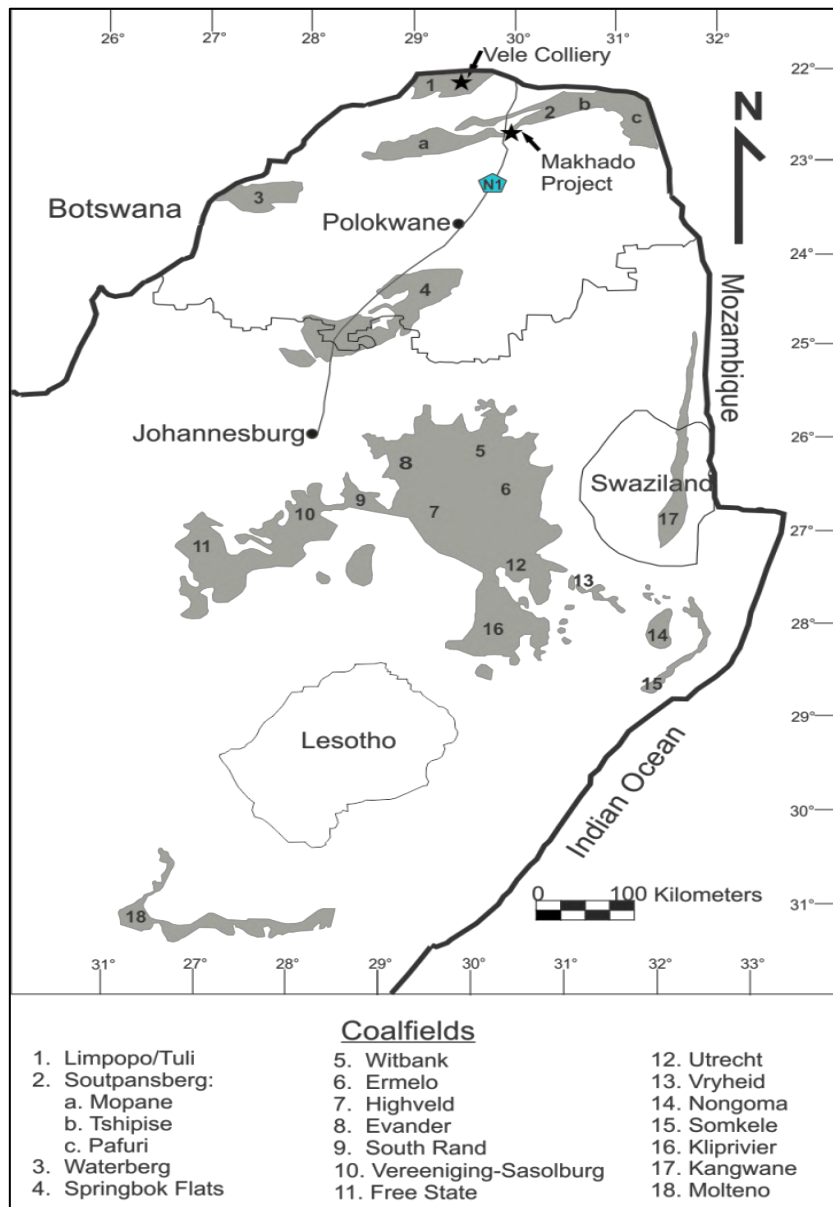


Figure 2. Location of the Limpopo and Soutpansberg Coalfields relative to other South African Coalfields modified after Council for Geosciences, (2015)

## 2.2 Materials collected

All the samples obtained for this research project were generously provided by CoAL on the basis of availability. Fresh and weathered samples were collected from stockpiles at Makhado Project and Vele Colliery. These stockpiles had been stored outdoors for varying periods of time and were sampled to represent, as best as possible, a spectrum of the freshest to the most weathered coal for this study. A comprehensive list of the samples collected and their descriptions is provided in Appendix 1. All samples from Makhado Project are denoted M, and those from Vele Colliery are denoted V.

Two stockpiles that had been stored since 2010 were sampled at Makhado Project, from stockpile 1 and stockpile 5 (Figure 3). Stockpile 1 had been processed and washed, whereas stockpile 5 was run of mine coal (Sparrow, 2015, pers. comm.). The uppermost layer i.e. the first  $\pm 5$  cm (weathered) and lower layer (fresh) of stockpile 1 were sampled. The uppermost coals in stockpile 5 were collected as weathered coals for the study, and were dull grey in colour and rich in disseminated rusty red (oxidised) siderite (Figure 4C). Weathered coal from stockpile 5 were relatively dry when they were collected, however, they were quite moist during sample preparation (Figure 4D). The sample referred to as “bright coal” (M3) in this study was vitrain rich and characterised by many bright coal bands. A distinct peacock tarnish similar to that observed in chalcopyrite was observed in some of the bright coal samples (Figure 4A & 4B).



Figure 3. Coal stockpiles at Makhado Project yard (Photo by John Sparrow)

Only one stockpile was available for sampling at Vele Colliery and was sampled using the same method applied at Makhado Project i.e. sampling of the uppermost weathered layer and lower fresh layer. The stockpile at Vele Colliery had been processed and since stored outdoors as of 2013. Furthermore, a coal sample was taken from each of the TM, TL, MS and BM coal seams of borehole OV125157A at Vele,

which was drilled in February 2014. These borehole cores were drilled near the mining operations and had similar properties as the processed stockpile coal (Sparrow, 2015, pers. comm.).

The author is aware that *in situ* coals undergo primary weathering; however, these process are usually less intense compared to secondary weathering, hence *in situ* coal is relatively more fresh (assuming it has not been significantly thermally altered by intrusions or percolating fluids) compared to coal that has been mined and subsequently stockpiled for extended periods (Jha *et al.*, 2014). *In situ* coal (fresh) was not obtainable for this study due to flooding and unstable walls in the pit at the time of sampling. Therefore, in addition to the fresh stockpile layer (V2), a sample that was drilled in 2010 and subsequently preserved at 4°C (V1) was provided to represent the “freshest” sample amongst the Vele coals.

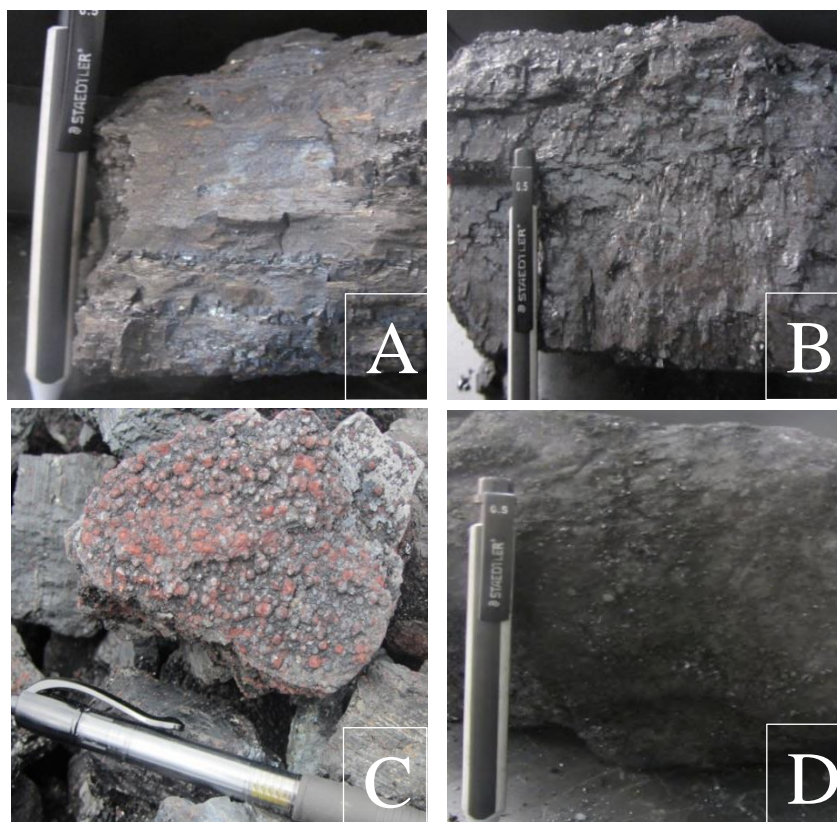


Figure 4. Coal samples collected from the ROM stockpile in Makhado, showing (A) peacock tarnish in bright coal (M3), (B) vitrain rich, bright coal (M3), (C), rusty red disseminated siderite in weathered coal (M4), (D) moist weathered coal (M4).

Coal from borehole OV125157A at Vele were regarded as weathered because they had been removed from the ground and subsequently stored in the core shed. Calcite veins and disseminated pyrite were common secondary mineralisation affecting the coal seams from borehole OV125157A (Figure 5). To a lesser extent, minor quartz veins and vugs occurred in the coal seams (Figure 5A). The MS and TL seams were dull and banded when observed in split core whereas BM and TM coals were bright and massive.

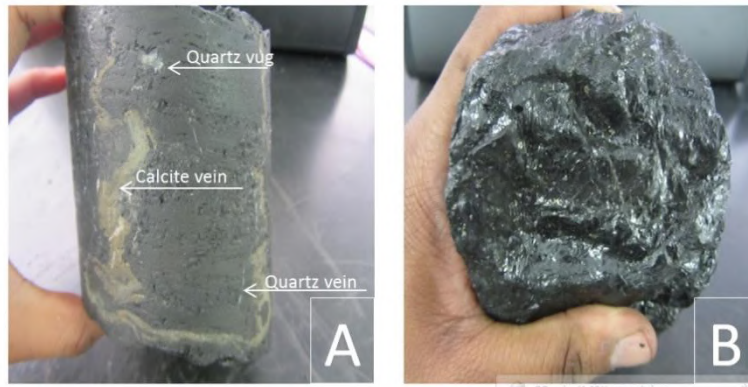


Figure 5. Mineral occurrence in Vele coals: (A); MS coal (V6) with secondary pale-brown calcite veins and white quartz veins and vugs, (B) disseminated pyrite in the bright TL coal seam (V5)

### 2.3 Sample preparation

The samples were prepared by crushing, milling and splitting. Crushing was conducted at the University of Johannesburg (UJ) Doornfontein Campus. The samples were crushed using Jaw, Roll and Cone crushers to reduce the sample sizes to less than 5mm. A rigorous sample splitting procedure is illustrated in Figure 6 and was followed in order to reduce the samples to manageable sizes whilst ensuring that a representative sample was prepared for the various analyses. The samples were split using the cone and quartering method whereby the main sample was initially divided into four quarters. Each of the quarters was then split into four smaller quarters. Of these, only two quarters (2 and 4) were then mixed together and split again, whereas the remaining quarters were stored for emergency use. This was repeated 3 times for each sample to obtain representative samples from each round of splitting. A composite sample was made by mixing the 4 representative samples derived from the splitting of the main sample. The representative sample was finally divided into four quarters of which quarters 2 and 4 were milled down to 0.25mm for geochemical analyses, and quarters 1 and 3 were milled down to 2.00mm for petrographic analysis. Finally, the milled representative samples were split into 8 smaller units for the various analyses using the Eriez MACSALAB 8 way microsplitter.

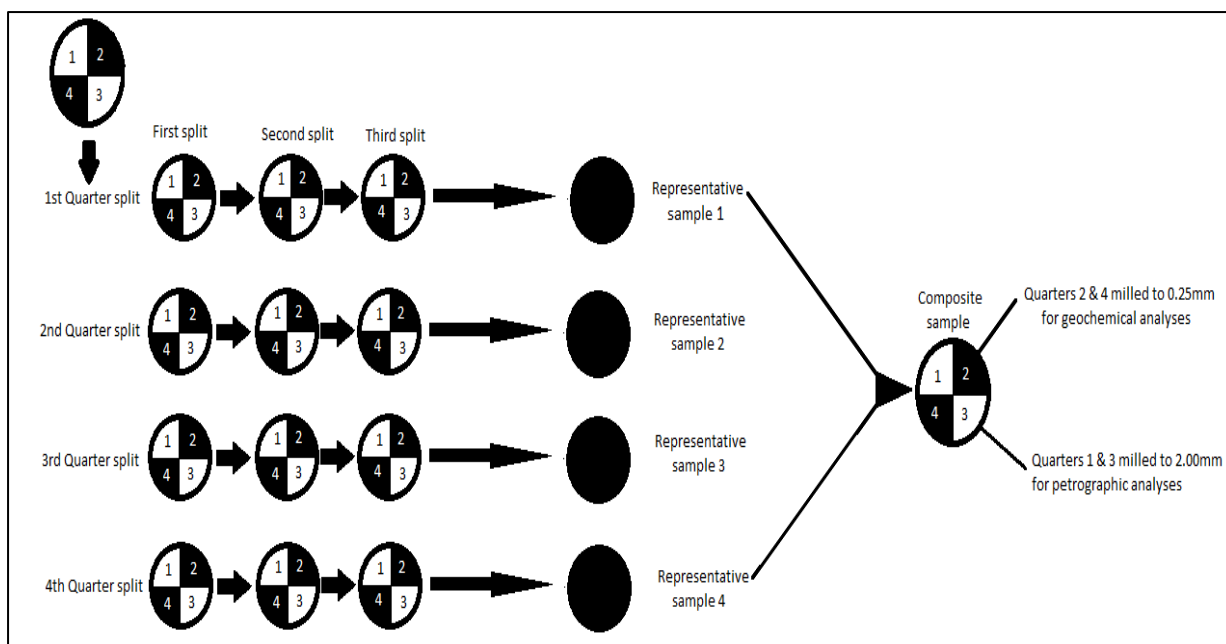


Figure 6. Cone and quartering method.

## 2.4 Analytical Methods

The properties of weathered and fresh coals are best studied using a combination of various petrographic and geochemical analyses described below:

### 2.4.1 Petrographic Methods

For the purposes of this project, the petrographic analysis of coal entailed, a maceral point count and vitrinite reflectance to determine the composition and rank of the coal (Crelling *et al.*, 1979; Lõ and Cardott, 1995). In addition, the Abnormal Condition Analysis was conducted to petrographically quantify observed weathering features in each sample. Photographs of features in the weathered and fresh coal were captured in plane polarised light to characterise the petrographic properties of weathered and fresh coal. The procedure followed for the petrographic analyses was done according to SABS-ISO 7404 standard.

Polished coal blocks were prepared by mixing the milled coal samples (-2mm) with 7g of EpoFix Resin and 1g of EpoFix Hardener in plastic moulds and leaving them overnight to harden at room temperature. The hardened blocks were then ground and polished using various grinding and polishing surfaces using the Struers TegraPol-11 polisher. Lastly the polished blocks were cleaned in a water bath for 5 minutes to remove the lubricants used during the polishing processes. A Zeiss AXIO petrographic microscope, fitted with Fossil Hilgers software and digital cameras was used to examine the polished coal blocks

under reflected light. The magnification was enhanced to 500 times through the use of an oil immersion objective lens.

#### **2.4.1.1 Maceral Point Count**

The objective of the maceral point count is to determine the bulk composition of the organic components that constitute the coal, as well as the minerals present within the coal (Misz-Kennan, and Fabiańska, 2011). Only 3 major maceral groups were identified for the scope of this investigation, namely vitrinite, inertinite and liptinite, plus mineral matter. A colour camera was utilised to clearly identify the macerals and minerals present within the sample. A total of 500 particles appearing under the cross hair were analysed using an automated stage with a specific step-size to ensure the whole surface of the block was covered. The results are reported on a volume percentage basis (section 3.1.1).

#### **2.4.1.2 Vitrinite Reflectance**

Vitrinite reflectance is determined by measuring the intensity of light that is reflected from the surface of vitrinite particles using a monochrome camera. Firstly, the software was calibrated to measure an intensity of reflected light at 0.90 %, using a yttrium-aluminium-garnet (YAG) standard. One hundred reflectance readings were then taken at random vitrinite (Rrandom) particles as the stage moved in a predetermined raster pattern across the block surface, and the results are presented on a histogram. The mean value of reflectance and the standard deviation were also produced for each sample. The mean reflectance value was used to determine the rank of the coal.

#### **2.4.1.3 The Abnormal Condition Analysis (ACA)**

Weathering can be petrographically distinguished by the presence of oxidation rims, pores, fractures (cracks), brown colouration and high relief on the surface of coal particles (Lõ and Cardott, 1995; Wagner, 2007; Misz-Kennan and Fabiańska, 2011). Wagner (1999) developed the ACA as a tool to detect the afore mentioned weathering features and others on a volume % basis. Detailed description on the procedure is provided in Wagner (1999). For the purposes of this study the ACA involved a volume % estimation of the proportion between fresh and weathered particles, and the volume % of secondary alteration features namely particles with uneven edges, cracked edges, prevalent cracks, oxidation rims and colour changes.

## 2.4.2 Geochemical Methods

Coals are classified based on their composition (type), the degree of metamorphism (rank), the amount of impurities (grade), as well as the amount of volatile matter (Tatsch, 1980). The quality of the coals quality was determined from the properties examined in the ultimate and proximate analyses, namely moisture, fixed carbon, ash, volatile matter as well as nitrogen, hydrogen, total carbon and total sulphur (Banerjee *et al.*, 2000; Suárez-Ruiz and Ward, 2008; Liu *et al.*, 2014). The analyses below were chosen to provide information that allowed distinction between fresh and weathered coals on a chemical basis.

### 2.4.2.1 Proximate analysis

This analysis is used to determine the content of ash, carbon, moisture, and volatiles for the fresh and weathered coal samples (Suárez-Ruiz and Ward, 2008). The analysis was conducted on an air dried (ad) basis according to ASTM D5142 standard using a TGA 701 Leco thermogravimetric analyser.

### 2.4.2.2 Ultimate analysis

Ultimate analysis is used to determine the chemical concentration of carbon (C), hydrogen (H), nitrogen (N), sulphur (S) in coal (Suárez-Ruiz and Ward, 2008). The oxygen (O) content was calculated by subtracting the sum the empirically derived elements from 100% using equation 1 bellow:

$$\text{Equation 1: } \% O = (100 - (C + H + N + S)) \quad (\text{Snyman, 1998})$$

This analysis was performed by The Council for Geoscience according to ASTM D5373-08 standard on an air dried basis (ad).

### 2.4.2.3 Calorific value (CV)

The calorific value is a measure of the heat released by coal when it is ignited and is thus an important property for thermal coals which are mainly used for electricity generation in South Africa (Steyn and Minnitt, 2010). The analysis was conducted using the DRYCAL modular bomb calorimeter situated in the Clean Coal Technology Research Laboratory, University of the Witwatersrand. Each sample was weighed to 0.40g in a crucible and inserted into the bomb. The bomb was then filled with oxygen until the crucible reached a pressure of 2400 kpa. The bomb was then placed in the calorimeter for approximately 6 minutes during which the sample was ignited and the energy released recorded in MJ/kg.

#### **2.4.2.4 Free swelling index (FSI)**

The FSI is an important property of coking coals which tests the ability of the coal to increase its volume by swelling into a carbon rich mass at temperatures between 350°-1400°C (Suárez-Ruiz and Ward, 2008). This is an important property of coking coal which makes it useful in the metallurgical industry where the coke is used to facilitate the conversion of raw iron ore into a metal (Schweinfurth, 2009). The samples were tested at ALS labs in Witbank according to the ISO 501 standard on an air dry basis (ad).

#### **2.4.2.5 X-ray florescence spectrometry (XRF)**

Major oxides and trace elements in the coals were determined using XRF at the School of Geosciences Earth Laboratory. The milled coal was heated overnight at a temperature of 600°C. The ash produced by this process was analysed for trace elements. Pressed pellets were made directly from the milled samples to determine the occurrence of major oxides in the coal.

### 3. Results

Reported in this section are the results of the various analyses performed for this research project. All samples from Makhado Project are denoted M, and those from Vele Colliery are denoted V.

#### 3.1 Petrographic Results

##### 3.1.1 Maceral Point Count

The maceral point count values for Makhado and Vele coals are presented in Figure 7 and the values recorded in Appendix 2.

The maceral point count showed that coal from Makhado and Vele were predominantly vitrinite rich with lesser amounts of inertinite and liptinite (Fig. 7). At Makhado liptinite only occurred in the bright coal (M3, 0.5%); however, liptinite was present in all of the Vele coals, albeit in smaller proportions relative to vitrinite (Fig. 7). Generally, Vele coals contained more mineral matter (15.8-40.2%) compared to Makhado coals (6.1-34.3%). The fresh coals at Makhado had less mineral matter (M2, 6.1% & M5, 8.7%) compared to the weathered coals (M1, 9.4% & M4, 34.3%). The fresh stockpile coal (V3) at Vele had the highest content of mineral matter (40.3%) with respect to the fridge coal (V1, 18.9%), weathered stockpile coal (V2, 33.3%) and the borehole coals (15.8-25.3%).

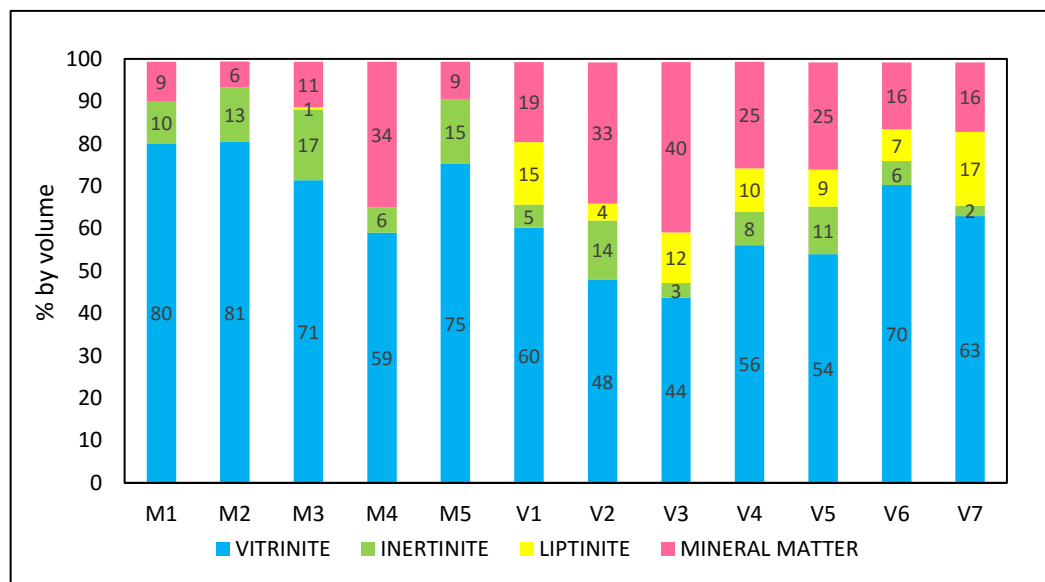


Figure 7. Maceral point count results for Makhado Project and Vele Colliery coal samples. Actual percentages appear within the bars

### 3.1.2 Vitrinite Reflectance

The random reflectance readings for Makhado and Vele coals are presented in Table 3 and the vitrinite reflectograms are presented in Appendix 3.

The random reflectance readings indicated that coals from Makhado were more highly reflecting (0.69-0.81%) than Vele coals (0.66-0.69%). Weathered coals at Makhado (M1 & M4, 0.81%) were more highly reflecting than their fresh counterparts (M2, 0.73% & M5, 0.78%). Overall, Vele coals had a narrow reflectance range such that the fridge coal (V1) and borehole coals (V4 and V6) had the same random reflectance of 0.69%. Furthermore, the weathered stockpile coal (V2), fresh stockpile coal (V3), and borehole coal (V7) had the same random reflectance reading of 0.66%. The fridge coal (V1) coal had a higher random reflectance (0.69%) than the weathered and fresh stockpile coals (V2 & V3, 0.66%).

The vitrinite reflectograms of Makhado coals had a slightly broader range in the reflectance values (0.55-1.05%) compared to Vele (0.4-0.9%). Relatively broad histogram ranges were only observed in Makhado's weathered stockpile 1 coal (M1, 0.6%), fresh stockpile 2 coal (M5, 0.55%) and the fresh Vele stockpile coal (V3, 0.5%). Based on the vitrinite reflectance measurements the coals from both localities were classified as medium rank C bituminous coal (Table 3).

Table 3. Vitrinite reflectance values of Makhado Project and Vele Colliery coal samples.

CoAL property	Sample ID	Random reflectance	SD	Rank category
Makhado Project	M1	0.81	0.088	Medium rank C (Bituminous)
	M2	0.73	0.088	Medium rank C (Bituminous)
	M3	0.69	0.064	Medium rank C (Bituminous)
	M4	0.81	0.064	Medium rank C (Bituminous)
	M5	0.78	0.075	Medium rank C (Bituminous)
Vele Colliery	V1	0.69	0.062	Medium rank C (Bituminous)
	V2	0.66	0.082	Medium rank C (Bituminous)
	V3	0.66	0.078	Medium rank C (Bituminous)
	V4	0.69	0.065	Medium rank C (Bituminous)
	V5	0.68	0.045	Medium rank C (Bituminous)
	V6	0.69	0.051	Medium rank C (Bituminous)
	V7	0.66	0.045	Medium rank C (Bituminous)

### 3.1.3 The Abnormal Condition Analysis (ACA)

The volume percentage (vol%) of petrographic weathering features are recorded in Appendix 4 and graphed in Figure 8. Photomicrographs of the observed weathering features are shown in Figure 9.

Makhado's fresh stockpile 1 coal (M2) showed the most signs of weathering compared to all other coals from Makhado and Vele (Fig. 8). Apart from prevalent cracks and colour change, other alteration features were absent in all the coal samples from Makhado and Vele (excluding M2). Weathered coal from stockpile 5 (M4) had the highest occurrence of alteration minerals (18%); furthermore, sample M4 was unique in that it predominantly consisted of very small coal particles (Figure 9C). The dominant alteration mineral was orange-red in colour, and had a granular texture. This alteration mineral frequently occurred within the coal matrix as massive or smaller grains (Figure 9C), or as massive grains on the edges of vitrinite particles (Figure 9D). Oxidation rims were strangely only observed in Makhado's fresh stockpile coal (M2), however these oxidation rims were not well developed. At Vele the borehole coals (V4 to V7) virtually had no altered minerals and the only indication of weathering in these coals was the presence of prevalent cracks (0.5-5.4%). Cracked edges on vitrinite particles (Figure 9E) and sometimes on inertinite were key features in the fresh stockpile 1 coal (M2). Jagged or saw-tooth edges were commonly observed in most of the samples such as the TM seam coal (V4) in Figure 9F. These edges were usually sharp but in some places they were scalloped.

Makhado and Vele coals were essentially fresh as they mainly consisted of fresh organic matter and fresh minerals (percentages reported here take into account fresh organic matter and fresh minerals). Vele coals comprise fresh coal particles in the range of 94.6-99%. Notably, the weathered stockpile coal (V2, 99%) had slightly more fresh coal particles than the fridge coal (V1, 98.5) and fresh stockpile coal (V3, 97.1%). In stockpile 1 at Makhado, the weathered coal had more fresh particles (M1, 94.9%) compared to its fresh counterpart (M2, 77%). The opposite was true in stockpile 5 in which the fresh coal had more fresh particles (M5, 97.5%) relative to the weathered coal (M4, 80.5%).

Coal particles in the borehole coals (V4 to V7) appeared fresh and were associated with detrital and framboidal forms of pyrite, together with quartz and minor clay minerals (Figure 9A). The fridge coal had the highest percentage of fresh organic matter (V1, 81.50%) followed by the weathered stockpile coal (V2, 71%) and fresh stockpile coal (V3, 62.35%). At Makhado weathered coal from stockpile 1 had more fresh organic matter (M1, 89.4%) compared to its fresh counterpart (M2, 74.5%), whereas the weathered coal from stockpile 5 had less fresh organic matter (M4, 73.5%) relative to the fresh coal (M5, 94.5%).

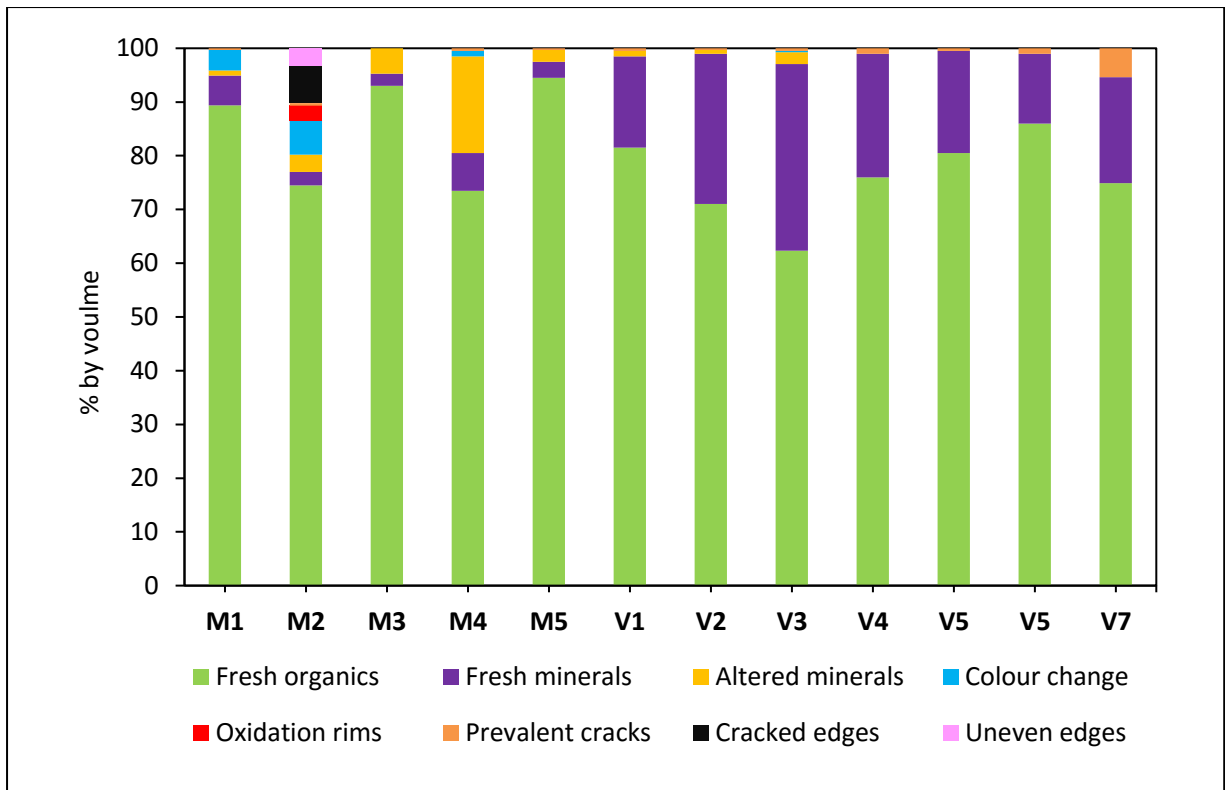


Figure 8. ACA by vol% for Makhado Project and Vele Colliery coal samples.

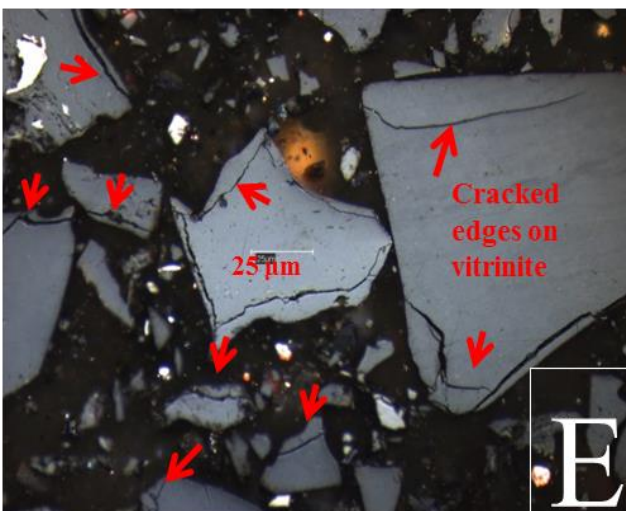
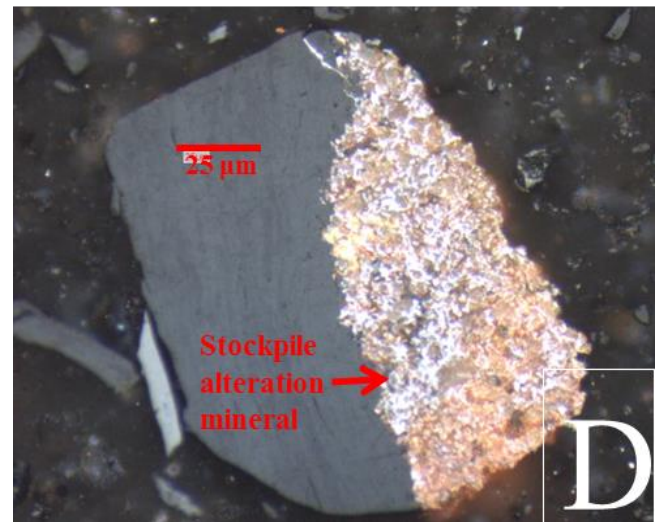
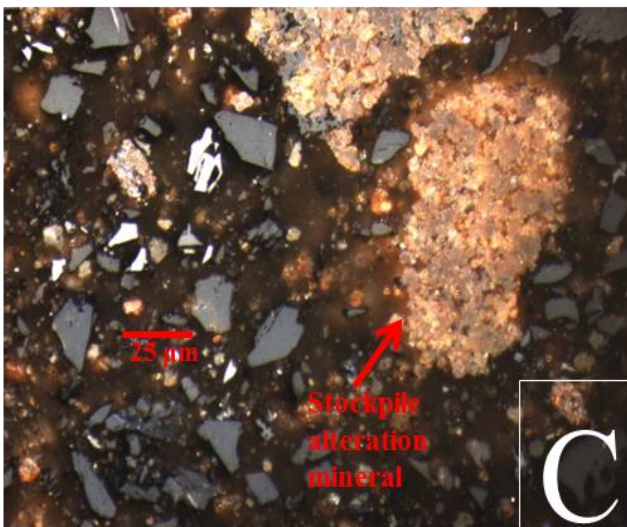
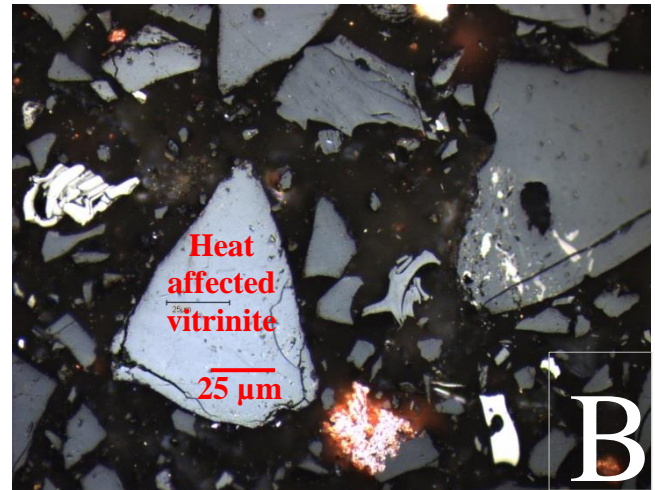
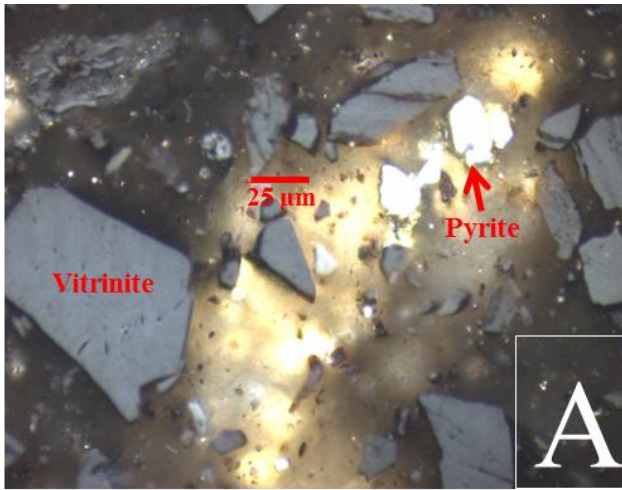


Figure 9. Photomicrographs of coal in reflected light showing (A) Fresh vitrinite particles with pyrite in sample V6, (B) Heat affected vitrinite in sample V2, (C) Small vitrinite particles occurring with small and large grains of the orange-red stockpile alteration mineral in sample M4, (D) Orange-red stockpile alteration mineral developing on the edge of a vitrinite particle in sample V3, (E) Cracks at the edges of vitrinite particles in sample M2, (F) Saw-tooth edge on vitrinite particle in sample V4. (Oil immersion, x 500 magnification).

## 3.2 Geochemical Results

### 3.2.1 Proximate analysis

Proximate results (ad) are shown in Figure 10 and the actual values are reported in Appendix 5.

The moisture content of the weathered stockpile coals (M1, 2.11% & M4, 2.29%) was higher than the moisture in the fresh stockpile coals (M2, 1.39% & M5, 1.21%) and the bright coal (M3, 1.32%). In contrast, the moisture content in the fresh coals at Vele (V1, 1.57% & V3, 1.73%) was higher than in the weathered stockpile coal (V2, 1.52%), but the fridge coal (V1) had less moisture compared to the fresh stockpile coal (V3). Borehole coals (V5 to V7), were moister (1.62-1.67%) compared to the fridge coal (V1) and weathered stockpile coal (V2).

Fixed carbon content was higher in the fresh coals (M2, 54.41% & M5, 50.16%) and the bright coal (M3, 51.43%) compared to the weathered coals (M1, 50.18% & M4, 19.23%). Makhado coals, excluding sample M4 (19.93%), had higher fixed carbon content (50.16-54.41%) than all the Vele coals (27.73%-44.19%). At Vele the weathered stockpile coal (V2, 29.01%) had more fixed carbon than its fresh counterpart (V3, 27.73%) whereas, the fresh fridge coal (V1, 35.87%) had more fixed carbon relative to the weathered stockpile coal (V2) and the borehole coals (V4, 34.18% to V7, 44.19%).

Ash content was higher in the weathered coals at Makhado (M1, 22.72% & M4, 44.05%) compared to their fresh counterparts (M2, 17.68% & M5, 19.60%) and the bright coal (M3, 18.15%); but overall the weathered and fresh coal from stockpile 1 (M1 & M2) had higher ash contents than the weathered and fresh coal from stockpile 5 (M4 & M5). Apart from sample M4, all other coals from Makhado had lower ash contents (17.68-22.72%) compared to Vele coals (23.50-47.45%). At Vele the weathered stockpile coal (V2, 45.89%) had more ash compared to the fridge coal (V1, 34.01%), but it was less than that of the fresh stockpile coal (V3, 47.45%). The fridge (V1) and fresh stockpile (V3) coals had higher ash contents than the borehole samples (V4 to V7) where the ash content decreased with depth from 32.23% to 23.50%.

Contrasting trends were detected for the volatile matter in the weathered and fresh coals at Makhado. In stockpile 1, fresh coal (M2, 26.52%) had more volatiles compared to the weathered coal (M1, 25%) while, in stockpile 5 weathered coal (M4, 33.73%) had more volatiles than its fresh counterpart (M5, 29.03%). Overall, volatile matter was higher in stockpile 5 coal compared to stockpile 1 coal; furthermore, the bright coal (M3) had more volatiles (29.10%) than all coal from stockpile 1. At Vele volatile matter was highest in the fridge coal (V1, 28.55%), and lowest in the fresh stockpile coal (V3, 23.09%) however, the weathered stockpile coal (V2, 23.58%) had more volatiles compared to the fresh

stockpile coal (V3). Volatile matter in the borehole coals (V4 to V7) decreased with depth from 32.16% to 30.67% and was higher than the volatile matter in the weathered and fresh stockpile coals (V2 & V3), the fridge coal (V1) and all coals from Makhado except for sample M4 (33.73%).

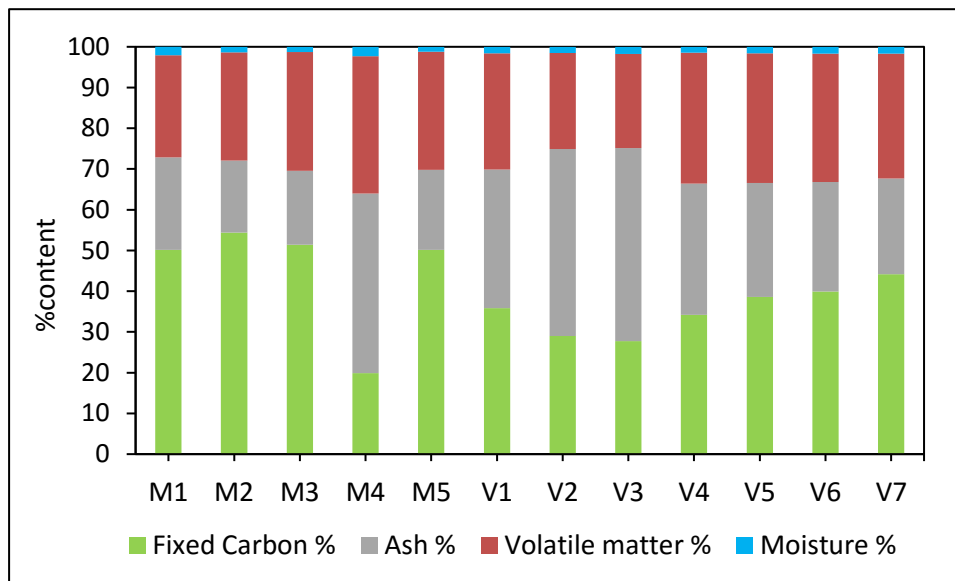


Figure 10. Proximate results (ad) for Makhado Project and Vele Colliery coal samples

### 3.2.2 Ultimate analysis

The elemental compositions of the coals are presented in Figure 11 and the actual values are presented in Appendix 6.

The total carbon content was consistently low in the weathered coals from Makhado's stockpile 1 (M1, 66%) and stockpile 5 (M4, 42%) compared to their fresh counterparts (M2, 71.40% & M5, 69.20% respectively). Although bright coal (M3, 70.40%) had the second highest total carbon content amongst the coal from Makhado, its value was greater than all those at Vele (41.10-64.00%). Apart from borehole coal V4 (53.00%), all other borehole coals (V5 to V7) had more total carbon (58.40-64.00%) compared to the fridge coal (V1, 53.60%).

Weathered coals from Makhado had higher oxygen contents (M1, 27.05% & M4, 54.17%) compared to their fresh counterparts and the bright coal (M3, 22.95%). The reverse is true for Vele coals whereby the weathered stockpile coal had less oxygen (V2, 52.42%) compared to its fresh counterpart (V3, 54.09%), however, the fridge coal had the least oxygen (V1, 40.92%) relative to the fresh and weathered stockpile coals (V2 & V3). The borehole coals had lower oxygen values (V4-V7; 28.99-33.58%) relative to the stockpile coals (V1-V3; 54.09-40.92%).

At Makhado, the hydrogen content was higher in the fresh stockpile coals (M2, 4.31% & M5, 4.28) compared to the weathered stockpile coals (M1, 4.08% & M4, 2.58%), however the weathered (M1) and fresh coal (M2) from stockpile 1 had more hydrogen compared to those in stockpile 5 (M4 & M5). Bright coal (M3, 4.31%) had the same hydrogen content as fresh coal (M2) from stockpile 1, which was higher than the hydrogen contents of all coals from Vele (2.95-4.13%). At Vele, the hydrogen content in the weathered stockpile coal (V2, 3.02%) was intermediate between that of the fridge coal (V1, 3.58%) and fresh stockpile coal (V3, 2.95%). The borehole coals (V5, 3.96% to V7, 4.13%), excluding sample V4 (3.46%), had more hydrogen compared to the fridge coal (V1), fresh stockpile coal (V2) and the weathered stockpile coals (V3).

Total sulphur at Makhado was highest in the fresh stockpile coals (M2, 0.84% & M5, 0.60%) with respect to the weathered stockpile coals (M1, 0.74% & M4, 0.47%), although stockpile 1 (M1 & M2) had higher sulphur values than stockpile 5 (M3, M4 & M5). Bright coal (M3, 0.64%) had more total sulphur compared to coal in stockpile 5 but, was less than the total sulphur values for stockpile 1 coal and also all coal from Vele (0.85-5.78%). Vele's fridge coal (V1, 0.85%) had less total sulphur with respect to the weathered stockpile coal (V2, 0.98%) however; the fresh stockpile coal (V3, 1.12%) had a higher total sulphur content compared to the weathered stockpile coal. The borehole coals (V4 to V7) had considerably higher (1.50-9.14%) sulphur contents compared to the fridge coal (V1, 0.85%), weathered stockpile coal (V2, 0.98%) and fresh stockpile coal (V3, 1.12%).

The nitrogen content of the weathered stockpile coals (M1, 1.53% & M4, 0.78%) was less than the fresh coals (M2, 1.66% & M5, 1.67%). Notably, bright coal (M3, 1.70%) had the highest nitrogen content relative to all other coals from Makhado and Vele. Nitrogen content increased with depth from V4 (0.82%) through to V7 (1.38%), and was higher than the weathered (V2, 0.78%) and fresh stockpile coals (V1, 1.05% & V3, 0.74%).

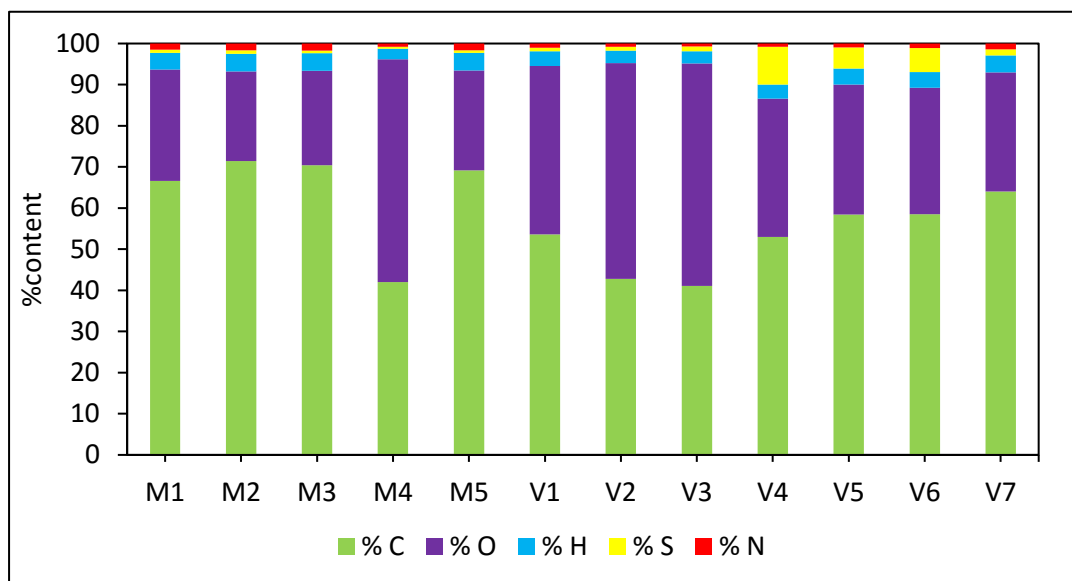


Figure 11. Ultimate results (ad) for Makhado Project and Vele Colliery coal samples

### 3.2.3 Calorific value (CV)

The calorific values of the coals are presented in Figure 12 and the actual values are reported in Appendix 7.

Fresh stockpile coals (M2 & M5) at Makhado had the same CV (27.12 MJ/kg) which was higher than the CV of the weathered stockpile coals (M1, 26.44 MJ/kg & M4, 14.81 MJ/kg). Bright coal (M3) had the highest calorific value (28.09 MJ/kg) with respect to all coals from Makhado and Vele (16.32-25.52 MJ/kg). Apart from the weathered coal from stockpile 5 (M4), Makhado coals generally had higher CVs (26.44-28.09 MJ/kg) than Vele coals (16.32-25.52 MJ/kg). Vele's fridge coal (V1, 20.91 MJ/kg) and the fresh stockpile coal (V3, 18.00 MJ/kg) had higher CV's than the weathered stockpile coal (V2, 16.317 MJ/kg), although the CV of the fridge coal was higher than that of the fresh stockpile coal. The CV's of the borehole coals (V4 to V5) was higher than the fridge coal (V1, 20.91 MJ/kg), weathered stockpile (V2, 16.32 MJ/kg) coal and fresh stockpile coal (V3, 18.00%).

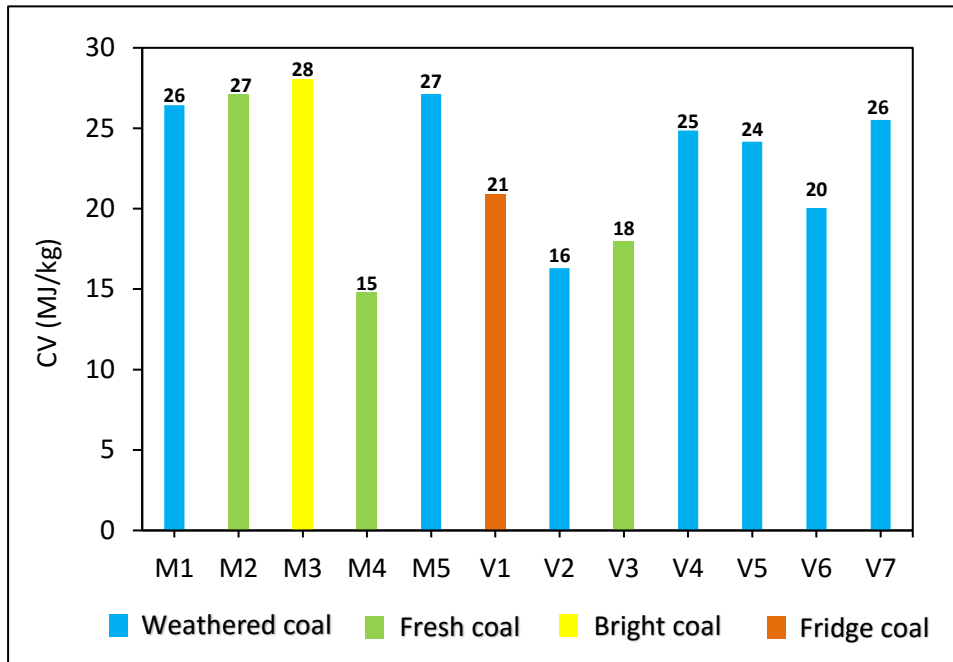


Figure 12. Calorific value (MJ/kg) for Makhado Project and Vele Colliery coals

### 3.2.4 Free swelling index (FSI)

The free swelling index results are plotted in Figure 13 and the actual values are tabulated in Appendix 8.

The FSI values of the weathered coals (M1, 0.0 & M4, 1.5) were extremely low compared to the fresh coals (M2, & M5, 7.5). At Vele the weathered stockpile coal (V2) had a far greater FSI (7.0) compared to the fridge (V1, 1.0) and fresh stockpile coal (V3, 0.0). Notably bright coal (M3) had the highest FSI value (8.0) compared to all other coal from Makhado and Vele. The borehole coals (V4 to V7) had higher FSI values in the range of 2.5 to 4.0 compared to the fridge coal and fresh stockpile coal.

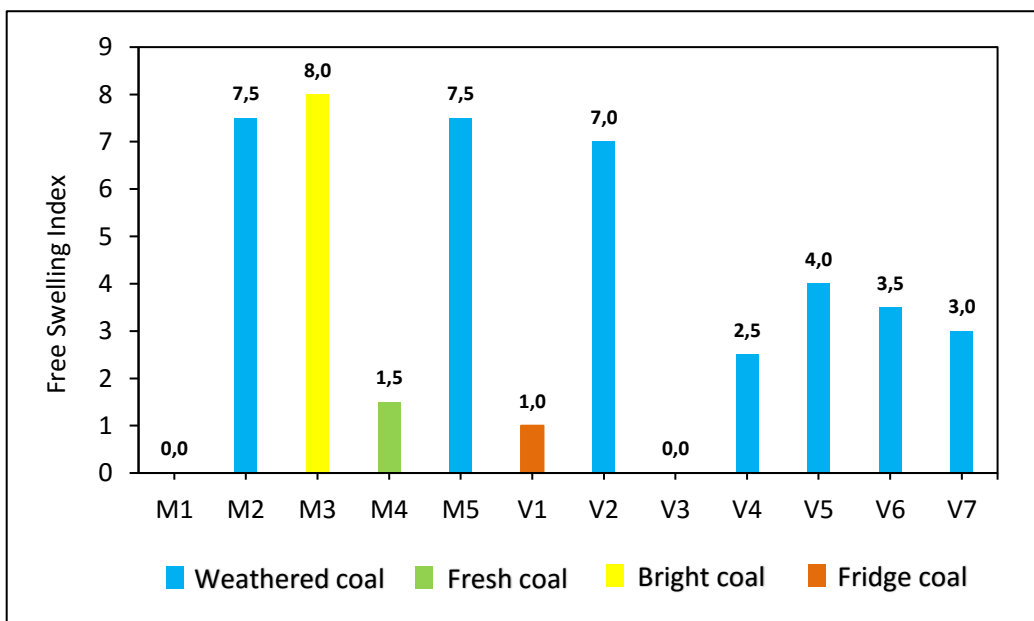


Figure 133. Free swelling index values of Makhado Project and Vele Colliery coal samples

### 3.2.5 X-ray fluorescence (XRF)

#### 3.2.5.1 Major element analysis

The major element analysis indicated that weathered and fresh samples from Makhado and Vele were predominately rich in SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and FeO (Table 4).

Table 4. Major element oxide (wt%) composition, CIA and CIW values of Makhado Project and Vele Colliery coal samples

Sample ID	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	FeO	MnO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	TiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	Cr <sub>2</sub> O <sub>3</sub>	NiO	TOTAL	LOI	CIA	CIW
M1	62.55	22.02	1.08	8.78	0.17	0.65	0.76	0.08	1.19	1.59	0.39	0.08	0.03	99.37	1.11	91.56	96.33
M2	59.15	21.28	1.39	11.29	0.12	0.71	0.85	0.08	1.12	1.9	0.48	0.06	0.03	98.48	1.13	91.21	95.81
M3	54.84	22.12	1.99	16.1	0.25	0.67	0.59	0.04	0.76	1.04	0.36	0.04	0.02	98.82	1.71	94.09	97.23
M4	30.89	13.92	5.27	42.64	0.76	1.54	0.91	0.07	0.51	0.42	0.11	0.04	0.02	96.95	1.51	90.33	93.42
M5	56.61	23.88	1.66	13.45	0.21	0.61	0.52	0.02	0.78	0.95	0.25	0.03	0.02	98.98	1.46	94.76	97.79
V1	60.73	21.23	0.69	5.58	0.2	1.04	3.83	0.07	1.49	0.94	0.07	0.05	0.02	95.94	2.40	79.75	84.48
V2	66.18	22.77	0.34	2.75	0.08	0.86	2.28	0.1	1.86	0.95	0.07	0.03	0.01	98.28	1.91	84.30	90.54
V3	51.18	12.66	2.65	21.49	0.05	0.44	6.85	0.09	1.33	0.47	0.12	0.11	0.07	97.5	3.45	60.49	64.59
V4	27.29	7.49	4.92	39.85	0.1	0.78	11.4	0.05	0.81	0.29	0.17	0.29	0.13	93.49	4.58	37.92	39.55
V5	64.05	22.95	0.44	3.53	0.09	0.87	2.79	0.09	1.77	0.97	0.07	0.04	0.01	97.68	2.48	83.15	88.85
V6	40.41	14.15	2.91	23.53	0.1	0.42	8.85	0.02	1.5	0.5	0.16	0.13	0.05	92.73	2.97	57.71	61.47
V7	68.24	15.81	0.65	5.26	0.08	0.3	5.06	0.03	0.95	0.77	0.05	0.04	0.01	97.26	3.14	72.36	75.65

The chemical index of alteration (CIA) and chemical index of weathering (CIW) have been used extensively to determine the extent of weathering of soils, crystalline rocks and more recently in the silicates that occur in weathered coal fly ash (Price and Velbel, 2003; Haskins, 2006; Bahlburg and Dobrzinski, 2009; Akinyemi, 2011; Akinyemi, 2013). The CIA & CIW are similar in that they quantify the breakdown of feldspar during progressive weathering, but they differ in that the CIW takes into account the fact that potassium (K<sup>+</sup>) is not only leached during weathering but can also be incorporated into new clays (Price and Velbel, 2003; Haskins, 2006). Calculated CIA and CIW values are plotted against SiO<sub>2</sub> (wt%) in Figure 14. The CIA values are calculated using Equation 2 whereby Ca<sup>2+</sup>, Na<sup>+</sup> and K<sup>+</sup> are mobile during weathering (Nesbit & Young, 1984). CIW values are calculated using Equation 3 whereby Ca<sup>2+</sup> and Na<sup>+</sup> are considered mobile elements during weathering (Harnois, 1988).

$$\text{Equation 2: } CIA = \left( \frac{Al_2O_3}{Al_2O_3 + CaO^* + Na_2O + K_2O} \right) \times 100 \quad (\text{Nesbit and Young, 1984})$$

$$\text{Equation 3: } CIW = \left( \frac{Al_2O_3}{Al_2O_3 + CaO^* + K_2O} \right) \times 100 \quad (\text{Harnois, 1988})$$

CaO\* = CaO in silicate minerals

Weathered coal from stockpile 5 (M4), and borehole coals V4 and V6 were classified in the region of low chemical weathering, whereas all other coals from Makhado and Vele were classified as moderately weathered (Figure 14). The CIW values were consistently higher than the CIA values for all coals from both localities (Table 4), and both plots shared the same trend in which the CIA and CIW values for Makhado coals were higher than the Vele coals (Figure 14). Makhado's weathered stockpile 1 coal (M1) and Vele's weathered stockpile coal (V2) had higher CIA and CIW values compared to their fresh counterparts, whereas the CIA and CIW of fresh coal from stockpile 5 (M5, 94.76 & 97.79) were higher than that of the weathered coal (M4, 90.33 & 93.42). The borehole coals (V4 to V7) had lower CIA (37.92-83.15) and CIW (39.55-88.85) values compared to the weathered stockpile coal (V1 79.75 & 84.48). Apart from the weathered stockpile 5 coal (M4) all the other coals from Makhado plotted near kaolinite in the CIW chart, whereas in the CIA chart only the bright coal (M3) and fresh stockpile 5 coal (M5) tended towards kaolinite (Figure 14). On both CIA and CIW charts, all Vele coals occurred within the muscovite field, except for borehole coal (V4) which occurred just below feldspar.

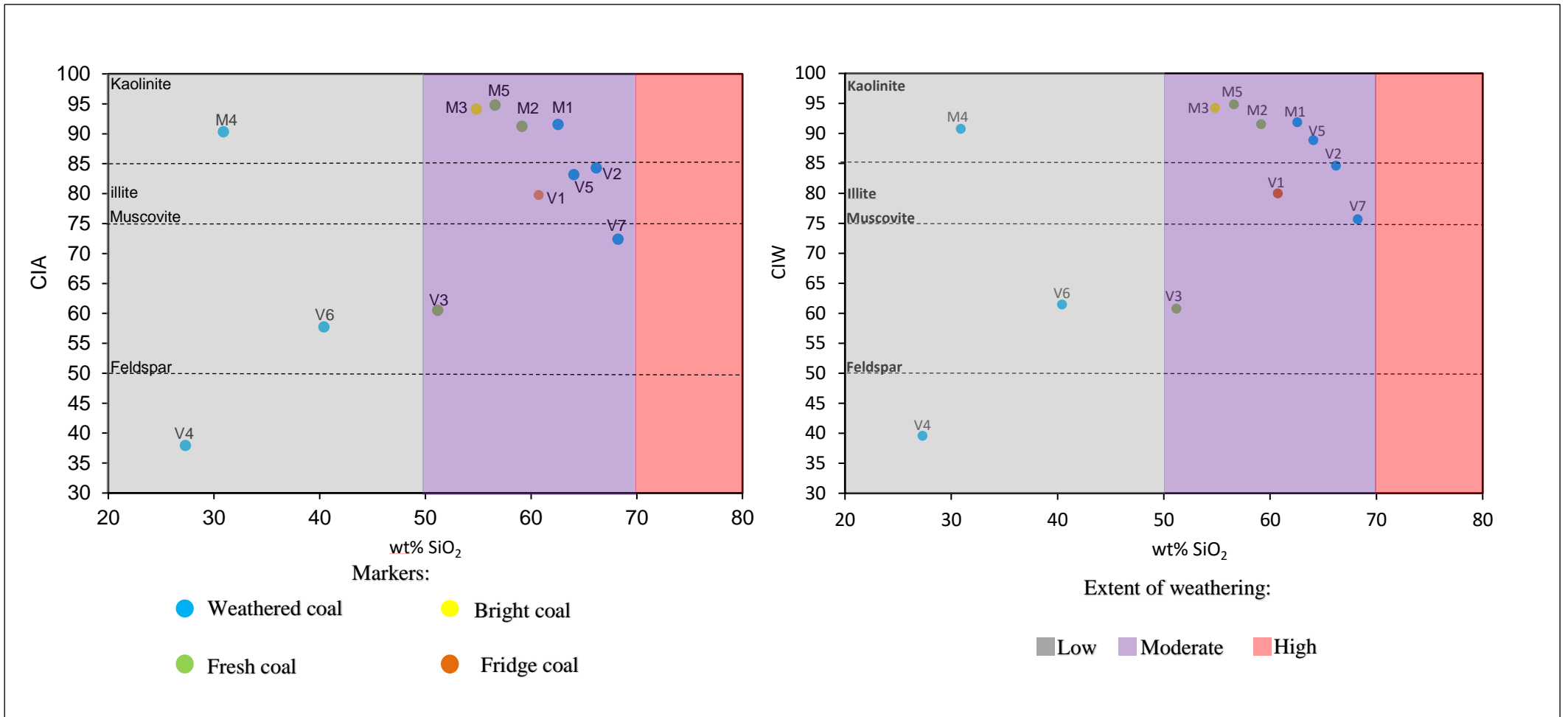


Figure 144. The extent of weathering for Makhado and Vele coals based on the CIA and CIW.

### 3.2.5.2 Trace element analysis

All samples from Makhado and Vele had relatively high levels of barium, moreover the fresh coals from Makhado and Vele were richer in barium compared to their weathered counterparts (Table 5). Overall, fresh coals from Makhado and Vele had more Co, Cu, Pb, Sr, and V compared to their weathered counterparts. Weathered and fresh coal from Makhado's stockpile 1 had very high concentrations of vanadium (M1, 107.63<sub>ppm</sub> & M2, 131.83<sub>ppm</sub>) relative to all other coals from both localities (21.91-70.29<sub>ppm</sub>). Vele's fresh stockpile coal (V3, 103.95<sub>ppm</sub>) and borehole coals V4 (552.01<sub>ppm</sub>), V5 (160.89<sub>ppm</sub>) and V6 (203.37<sub>ppm</sub>) had high concentrations of Cr relative to all the other coals (26.26-94.64<sub>ppm</sub>). Borehole coal V4 had the highest Ni content at 229.16<sub>ppm</sub> compared to all other coals from both localities (20.55-89.46<sub>ppm</sub>). All but the weathered stockpile 5 coal (M4, 43.19<sub>ppm</sub>) from Makhado had high Sr contents (109.81-159.7<sub>ppm</sub>) compared to the Vele coals (38.75-97.11<sub>ppm</sub>). Makhado's stockpile 1 coals (M1, 199.04<sub>ppm</sub> & M2, 219.01<sub>ppm</sub>) and Vele's stockpile coals (V1, 133.27<sub>ppm</sub>, V2 153.24<sub>ppm</sub> & V3 163.61<sub>ppm</sub>) had high Zr relative to all other coals in both localities (30.78- 93<sub>ppm</sub>).

Table 5. Trace element distribution for Makhado Project and Vele Colliery coal samples

ID	Sc	V	Cr	Co	Ni	Cu	Zn	Ga	Rb	Sr	Y	Zr	Nb	Mo	Ba	Pb	Th	U
M1	9.72	107.63	94.64	17.75	42.17	28.93	42.89	20.84	19.15	154.52	24.64	199.04	12.15	7.73	283.26	19.19	7.84	2.58
M2	10.12	131.83	66.12	20.6	36.87	33.57	53.11	25.08	16.64	159.7	23.85	219.01	12.86	6.37	313.5	21.11	7.31	3.05
M3	7.87	45.09	37.86	14.84	28.66	14	11.02	12.67	10.31	141.35	19.67	93	6.19	3.44	239.37	15.12	7.71	1.31
M4	11.9	30.31	76.52	2.37	47.3	28.47	37.13	13.2	14.66	43.19	15.66	72.17	6.21	6.63	112.8	11.71	4.1	d.l.
M5	6.94	35.5	26.26	15.41	27.42	11.73	14.81	10.92	11.46	109.81	19.87	69.74	4.7	3.4	197.47	13.9	7.82	0.87
V1	7.76	58.29	91.51	9.51	34.58	16.18	39.69	13.63	33.58	92.25	21.86	133.27	11.07	7.31	222.94	18.73	9.77	0.31
V2	9.88	62.94	82.1	8.58	29.11	19.58	44.66	16.74	53.92	92.34	25.75	153.24	11.34	5.59	222.88	22.29	12.69	1.49
V3	9.79	70.29	103.95	10.52	45.56	19.83	48.35	16.4	51.8	97.11	26.03	163.61	12.56	9.74	245.37	24.39	12.44	0.36
V4	8.9	34.85	552.01	26.94	229.16	16.16	4.29	4.01	14.24	89.11	28.39	30.78	3.23	50.69	126.38	11.09	2.56	d.l.
V5	5.87	30.8	160.89	33.69	89.46	11.6	18.73	5.04	20.07	50.04	40.35	43.53	3.37	15.06	120.9	13.65	4.34	d.l.
V6	6.51	31.4	203.37	9.93	72.11	11.16	5.24	5.59	22.96	62.91	17.72	42.13	3.9	16.23	121.98	13.37	4.71	d.l.
V7	4.68	21.91	46.14	15.78	20.55	10.22	6.2	6.32	12.71	38.75	16.24	51.98	3.45	3.63	108.87	9.9	4.16	d.l.

## 4. Discussion

### 4.2 Geochemistry

Weathering has commonly resulted in the increase of the inherent moisture content of coal and oxygen content because water is a by-product of the reaction of oxygen with coal (Pisupati and Scaroni, 1993; Martínez and Escobar, 1995). This was found to be true with respect to the weathered coals from Makhado, however at Vele, the fresh stockpile coal (V3) had the highest inherent moisture content, as well as oxygen content. The formation of excess water due to oxidation may also explain the decrease in hydrogen and carbon in the weathered coals relative to the fresh coals of Makhado and Vele. The absorption of moisture results in the formation of humic acids at the expense of organic substances in coal (Ingram and Rimstridt, 1984). Carboxylic (-COOH) and phenolic (-OH) functional groups are the most common humic acids that form when coal is oxidised (Marchioni, 1983). These humic acids are hydrophilic in nature, allowing for more moisture absorption (Xia *et al.*, 2014). The formation of humic acids can also be petrographically distinguished by the presence of oxidation rims around coal particles and scalloped edges however, the oxidation rims observed in Makhado's fresh stockpile 1 coal (M2) were poorly developed and, the occurrence of scalloped edges was rare. Moisture saturated coals have proven to be more difficult to beneficiate because of the difficulty associated with separating the coal from impurities (Cox and Nelson, 1984; de Korte, 2001). Wagner (2007) noted that discard stockpiles that had been stored for extended periods tended to have more inherent moisture compared to younger stockpiles, this was true for the weathered coals at Makhado apart from sample M3 (Wagner, 2007). In contrast, Martinez and Escobar (1995) found that there was no definite relationship between the degree of weathering and moisture content which was also evident in the Vele coals.

Various researchers such as Pisupati and Scaroni (1993) have noted that weathered coals contain more volatiles than fresh coal, possibly as a result of the rearrangement of carbon rings (aliphatic and alicyclic structures) during weathering (Marchioni, 1983; Jha *et al.*, 2015). Apart from sample M4, the volatile qualities of Makhado coal met the thermal domestic product requirements (23-30 MJ/kg) whereas only samples V1, V2, and V3 from Vele had suitable volatile contents for Eskom (20-30 MJ/kg) and domestic coal products (Appendix 9).

The ash qualities of coal from Makhado, except sample M4, were suitable for Eskom's thermal product specifications (25-34 MJ/kg); however, only the borehole samples (V4 to V7) at Vele were suitable to use as Eskom thermal products as per Appendix 9.

Sulphur content is expected to be lower in weathered coal compared to fresh coal due to the oxidation of sulphides into sulphates (Pisupati and Scaroni, 1993; Martínez and Escobar, 1995; Suárez-Ruiz and

Ward, 2008). Oxidation of these sulphides poses environmental risk because of the resultant sulphuric acids that contribute to acid mine drainage (Orem and Finkelman, 2003). The sulphur content of the coals from Makhado was well below the limit (2% sulphur) set by South African standards for coal combustion however; borehole samples from Vele V4, V5 and V6 greatly exceeded this limit (Lloyd, 2000).

The carbon content of weathered coal is known to decline during weathering because the carbon compounds are oxidised into carbon dioxide which is released during spontaneous combustion as oxygen increases (Suárez-Ruiz and Ward, 2008). The carbon content can be correlated to the calorific value in that coal with a high fixed carbon and/or total carbon content tend to have high CV's; for example, the weathered coal from stockpile 5 (M4) had low fixed carbon, total carbon and CV (Vermani, 1995). Banerjee et al., (2000) also experimentally found that carbon and hydrogen decreased in coals that had weathered over a 7-month period. Furthermore, the relatively high calorific values of Makhado and Vele coals could be linked to the highly reactive nature of vitrinite, which was the dominant maceral group in the coals as discussed in the previous section (Misz-Kennan and Fabiańska, 2011).

This also implies that the heating capacity of the coals was not sensitive to weathering at low levels of oxidation because the differences in the CV's of the weathered and fresh coals were not drastic (Appendix 7). Chelgani *et al.*, 2011 and Sharma *et al.*, (2012) have found a strong, positive correlation between the maceral type and CV whereby high vitrinite and liptinite content correlates to high calorific values, but the presence of inertinite and mineral matter reduces the CV. Although not statistically determined in this study, it is observed that bright coal (M3) had the highest calorific value of all the coals sampled in this study, and also had high vitrinite content, whereas weathered stockpile 5 coal (M4) had the lowest calorific value, very low vitrinite content and high mineral matter. The CV's of all the coal from Makhado excluding sample M4 met the heating specifications required for Eskom (21-24 MJ/kg) and domestic thermal products (24.5-27.5 MJ/kg) in South Africa as per Appendix 9. Notably the heating value of the bright coal (M3) was of export thermal coal grade (28.087MJ/kg). At Vele only the borehole coals (except V6) were suitable for domestic end use.

The FSI depends on coal rank and maceral composition of the coal, for example inertinite rich coals tend to have low FSI values compared to vitrinite rich coal (Marchioni, 1983; Suárez-Ruiz and Ward, 2008). This trend was not consistent for the weathered and fresh coals of Makhado and Vele. Weathered coals are also expected to have low FSI values even at low levels of exposure to oxygen because the carbon and hydrogen bonds are easily broken down at low energy levels during weathering, thus the trend observed between Vele's weathered and fresh coals was abnormal (Ndhlovu, 2011). The large differences in the FSI values between the weathered and fresh coals from Makhado and Vele indicated

that the coking properties of the coals were susceptible to low degree weathering compared to the CV. An experimental study conducted by Jha *et al.*, (2015) also found that the FSI was the most effective parameter to document the changes that occur in the early stages of weathering of medium and prime coking coal. The FSI values of the weathered coal at Makhado were well below the typical FSI range (7-9) of Makhado coals (Sparrow *et al.*, 2013). Based on the volatile matter content and FSI criteria used by CoAL (Appendix 10), sample M1 could no longer be used as coking coal, however its fresh counterpart M2 could be used as a medium volatile hard coking coal. Samples M3 & M5 were classified as high volatile hard coking coals, whereas the weathered sample M4 was a high volatile semi-soft coking coal that could be used as coking coal or pulverised coal injection. Vele coals are commonly classified as soft coking coals (Sparrow *et al.*, 2013). All Vele coals in this study were still suitable for use as soft coking coals apart from the fresh stockpile coal (V3) which did not possess any coking properties, and the weathered stockpile coal (V2) which was anomalously classified as hard coking coal. The sensitivity of the coking properties to weathering imply that they should be processed, transported and used before their quality depreciates significantly. The rate at which the various technological properties of coal deteriorate as a result of weathering need to be constrained to ensure the optimum use of coal in terms of efficient performance and monetary value.

Overall, the trends on the CIA and CIW concurred with the expectation that the longer coal is exposed, the more weathering it undergoes, as indicated by the coals from Makhado which had higher CIA & CIW values compared to coals from Vele. Even so, all coals from Makhado and most of the Vele coals were classified as moderately weathered most likely because the margin between the exposure period of the coals in these localities was relatively narrow as coals from Makhado had been stockpiled since 2010 including the Vele fridge coal (V1), whereas the stockpile coals from Vele (V2 & V3) had been exposed since 2013, and the borehole coals since 2014. The distribution of Vele coals on the CIA and CIW chart indicated progressive weathering that was not necessarily linked to the duration of exposure, which suggested that the geochemical properties of the coals required longer periods of exposure in order to show distinct weathering features at varying stages of weathering.

Although the CIA and CIW values are indicative of weathering, it is unclear whether the CIA values actually represent peatification, coalification or post-mining processes because initial baseline quality data was not available to allow for comparison. It appeared that preservation of the fridge coal (V1) had impeded the development of advanced alteration clays (illite and kaolinite) in the duration in which it was stored.

The trace element behaviour in the fresh and weathered coals was highly variable at both localities; however, a couple of inferences could be made. The presence of Ba and Sr in coal is associated with carbonates and corresponded with siderite and calcite that were visible in the hand samples as captured

in Figure 4C and Figure 5A, respectively (Ward, 2002). Rubidium is commonly associated with potassic minerals such as illite, mica and orthoclase (Ward, 2002). Illite represents a high degree in the weathering of silicate minerals within the coals (Bahlburg and Dobrzinski, 2009). This supported the fact that all Vele coals except V2 were less weathered compared to Makhado coals which had been exposed to weathering longer. The sulphides (pyrite) in the coal samples were most likely associated with the presence of Zn, Cu and Pb (Ciesielczuk *et al.*, 2014).

Notably of all the coals from Makhado, the bright coal (M3) retained the best coal quality in terms of relatively high carbon (fixed & total), nitrogen, hydrogen, volatile matter, CV and FSI, whereas the weathered stockpile 5 coal (M4) displayed the poorest quality in terms of the afore mentioned properties. The preservation of coal at low temperature was to some extent able to maintain good coal quality in terms of relatively high nitrogen, hydrogen, carbon (fixed & total), volatile matter and CV as observed in the characteristics of Vele's fridge coal (V1). The characteristics of the fridge coal (V1) and fresh stockpile coal (V3) from Vele were intriguing in that their properties often contrasted each other rather than complement each other because they were both relatively fresh. In most cases the value(s) of the weathered stockpile coal (V2) was intermediate between the fridge coal and fresh stockpile coal, rather it was expected that the value for the various geochemical parameters of the weathered stockpile coal (V2) would be greater or less than the fridge coal (V1) and fresh stockpile coal (V3).

## 4.1 Petrography

A number of anomalous behaviours were exhibited by the fresh and weathered coals in that features that were expected in the weathered coals occurred in the fresh coals and vice versa. This was also evident in the geochemical results. For example, in terms of the petrographic observations it was strange that Makhado's fresh stockpile 1 coal (M2) showed the most degree of weathering. Although it is plausible that sample M2 was not sampled correctly or may have been switched during sample preparation or analysis, the geochemical signature of this sample suggested that it was indeed relatively fresh because it had less moisture, and more hydrogen, fixed carbon, total carbon, CV and FSI with respect to its weathered counterpart (M1).

The process of oxidation produces new minerals such as clays, sulphides and salts (Ward, 2002). The orange-red stockpile alteration mineral was a diagnostic feature of weathered stockpiled coals (Wagner 2015 pers. comm.). This orange-red stockpile alteration mineral could be oxidised siderite as was observed in hand specimen (Figure 4C), however a suitable analytical technique such as X-ray diffraction is required to accurately delineate this mineral. The fact that the weathered coals at Makhado and Vele (V2 with respect to V1) had more mineral matter compared to their fresh counterparts, did not necessarily imply weathering because of those, only the fresh stockpile 1 coal (M2), bright coal (M3) and weathered stockpile 5 coal (M4) consisted of more altered minerals relative to the fresh minerals (Table 5). Therefore, more often than not the geochemical and petrographic features that are considered abnormal are not always true reflectors of weathering unless they are consistently supported by the results of different analyses. In this case, it was most likely due to the fact that the coals investigated in this study were not weathered enough to allow for clear distinction.

The onset of secondary weathering is marked by margin effects such as the occurrence of cracked edges and oxidation rims (Wagner, 2007). The cracks that form on the edges of the macerals may be due to the periodic gain and loss of moisture, which effectively increase the surface area of the coal (Ingram and Rimstridt, 1984). Slackening is the process of grain size reduction that is commonly observed in weathered coals and could therefore be the cause of the small particle sizes of vitrinite and the orange-red alteration minerals that were so prevalent in the weathered coal from stockpile 5 (M4) (Banerjee *et al.*, 2000 Bahlburg and Dobrzinski, 2009). It is possible that the abundance of cracked edges (Fig. 9E) in the fresh stockpile coal 1 (M2), and the slackening effect in sample M4 were as a result of mechanical abrasion during the process crushing and milling, however should this be true then these features should have also been common in the other samples. The rare occurrence of scalloped edges on some of the vitrinite particles suggested that to some extent some of the vitrinite particles were dissolved when humic acids formed at the expense of the organic matter (Marchioni, 1983; Loison *et al.*, 1989; Suárez-Ruiz and Ward, 2008).

Vitrinite is known to be the most reactive maceral followed by liptinite, and they are therefore more susceptible to weathering compared to other macerals (Misz-Kennan and Fabiańska, 2010). On this basis it was expected that the vitrinite rich coals of Makhado and Vele would have undergone significant degree of weathering, more so for Makhado coals which were generally more vitrinite rich and had been exposed longer to secondary weathering (since 2010) compared to Vele coals (since 2013 & 2014).

In ideal geological conditions the reflectance of vitrinite increases progressively with rank during burial, however, it has been documented that higher reflectance in vitrinite particles develops due to exposure to thermal processes such as intrusions, self-heating or even spontaneous combustion (Misz *et al.*, 2007; Misz-Kennan and Fabiańska, 2010). The broad vitrinite reflectograms (Appendix 3) of samples M1, M5 and V3 indicated that the macerals in those samples had been thermally affected, whereas the macerals in the remaining samples of Makhado and Vele were less altered as was indicated by the narrower reflectograms (Misz *et al.*, 2007). This is in agreement with the fact that on average all the coals consisted of 94% fresh organic material and fresh minerals (unweathered), whereas, weathering features only accounted 6% of the coal composition (Appendix 4). Since the higher reflecting vitrinite particles (Fig. 9B) were not observed in significantly large proportions in either of the localities it could be inferred that the coals had experienced limited rates of self-heating (Misz *et al.*, 2007; Ciesielczuk *et al.*, 2014).

Well-developed oxidation rims are commonly associated with advanced stages of oxidation or spontaneous combustion (Misz *et al.*, 2007), thus the presence of juvenile oxidation rims in sample M2 and the absence thereof in the rest of the samples from both localities were indicative of low temperature (~25-50°C) oxidation at a slow rate (Wu *et al.*, 1988; Ciesielczuk *et al.*, 2014). This highlighted the importance of the duration of exposure in that coals that have been exposed for longer periods or to more harsh conditions will show more pronounced weathering features as was the case for Wagner (2007), whereby the discard coals that were analysed had been exposed to secondary oxidative conditions over an extended period of 5-40 years.

## 5. Conclusions

The petrographic and geochemical analyses indicated that coal from Makhado and Vele had undergone low temperature oxidation and were therefore only moderately weathered. It was on this basis that most of the coals met the quality specifications of domestic and Eskom thermal products. The coking properties of the weathered coals at Makhado (except the bright coal, M3) were severely affected by low temperature oxidation, and less so for Vele's weathered coals which had abnormally stronger coking strength compared to the fresh stockpile coals. Hence fresh coals from Makhado, including the weathered stockpile coal from Vele could be used as hard coking coal whereas the weathered borehole coals from Vele could be used as semi-soft coking coals. The inherent properties of the coals greatly affected their response to weathering as was observed in many of the geochemical and petrographic trends, thus the determination of weathering requires a number of analysis to support the validity of the observed weathering indicators.

It is likely that the preservation of coal at low temperature was useful in preserving most of the desirable properties of coal apart from the FSI. Of all the analysis, the FSI was the most sensitive geochemical parameter to low levels of weathering (i.e. drastic differences in coal quality value existed between the weathered and fresh coals). This has implications on mining companies that primarily produce coking coal as great care must be taken to ensure that suitable stockpiling methods are used and that the coal is used well before the desired properties deteriorate. The significance of time in the weathering process came into play in that coals that were exposed for longer periods underwent a greater extent of secondary weathering compared to coal that had been exposed for shorter durations.

Overall the weathered coals from both localities were geochemically marked by higher moisture, sulphur, lower carbon (fixed & total), nitrogen, hydrogen, volatile matter, and higher sulphur and ash. The scarcity of extreme weathering features such as well-developed oxidation rims indicated that the coals from Makhado and Vele had not been exposed long enough for these features to develop. This emphasises the fact that the weathering processes is slow and the inherent properties of the coal together with the storage conditions exert a control in their response to weathering.

## 5.1 Limitations of the study

Weathering is a process of change through time, therefore without initial baseline quality samples (i.e. quality data from fresh *in situ* coal) from which weathering can be detected, it is difficult to attribute any of the observations in this study to secondary weathering with certainty, hence all observations in this research are inferred. Furthermore, since only single point data were collected for this study due to financial constraints, the data were only interpreted based on the raw results because statistical analysis could not be applied. Had there been opportunity to analyse multiple samples per stockpile or per borehole, the following statistical approach would have been followed: the mean and standard deviation would have been calculated for the weathered and fresh samples from both localities, and the significance of the differences between the means compared between the stockpiles and any variation in the boreholes. Finally, a standard student's t-test would be conducted to determine whether the differences or similarities between the weathered and fresh samples from the localities are statistically significant.

## 5.2 Future work

In light of the findings and limitations of this study, further investigation should be carried out in terms of:

1. Constraining the rates at which various coal properties deteriorate to ensure that coal is utilised within a given time-frame so as to ensure optimum performance.
2. Delineating the orange-red stockpile alteration mineral and verifying the presence of other alteration minerals observed in the weathering analysis with the distributions on the CIA and CIW plots through X-ray diffraction.
3. Detailed characterisation of the petrographic and geochemical properties of weathered coals in other coalfields, namely the Waterberg Coalfield and possibly Natal Coalfields.

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## 7. Appendices

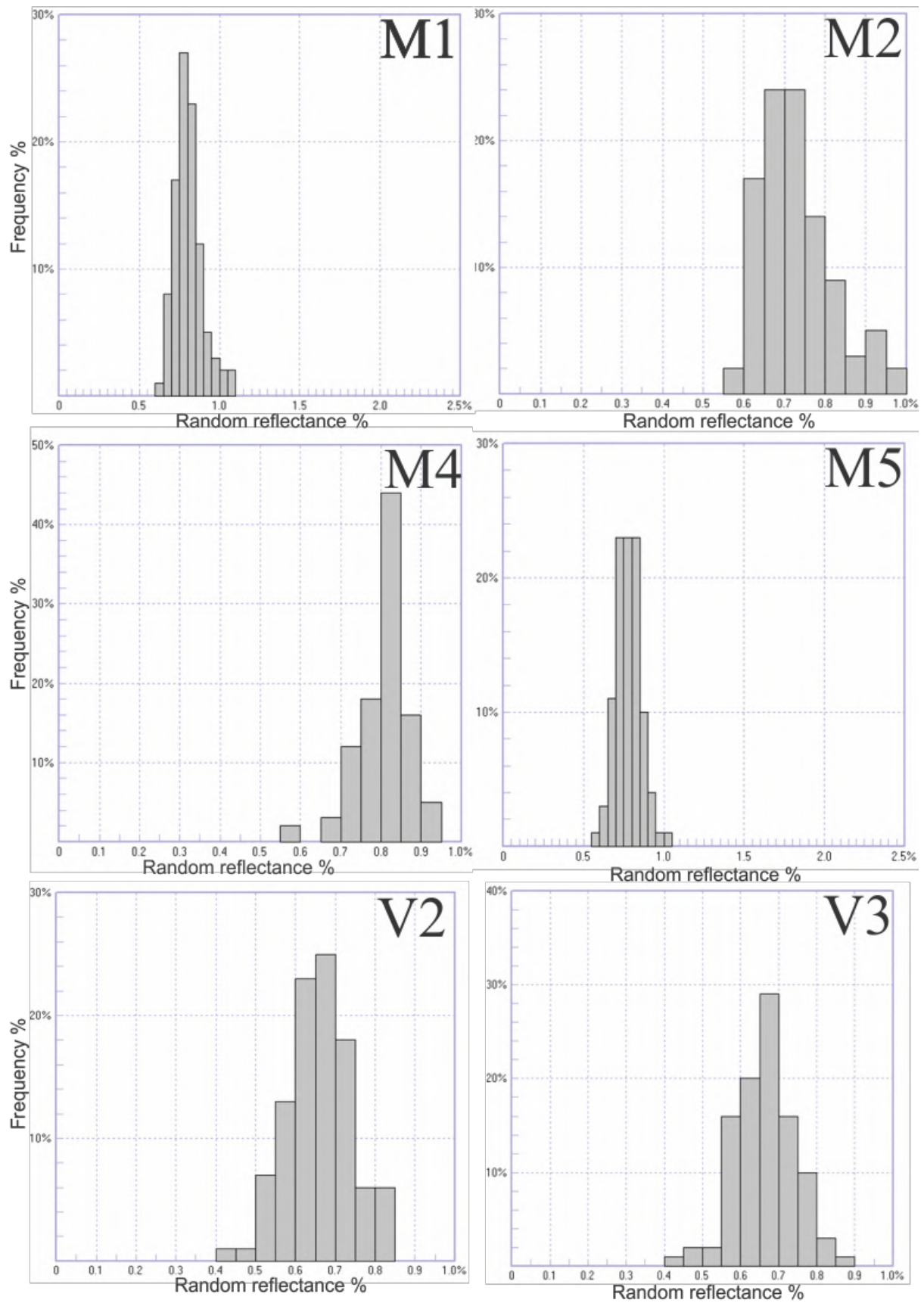
Appendix 1. Sample inventory of coal collected from Makhado Project and Vele Colliery

CoAL Property	Sample ID.	Sample description	Year since exposure	Date sampled
Makhado Project	M1	Stockpile 1: weathered surface coal layer 1	2010	30/03/2015
	M2	Stockpile 1: "fresh" coal layer 2	2010	30/03/2015
	M3	Stockpile 5: ROM bright coal	2010	30/03/2015
	M4	Stockpile 5: ROM weathered surface coal	2010	30/03/2015
	M5	Stockpile 5: ROM "fresh" coal	2010	30/03/2015
Vele Colliery	V1	Stockpile fridge coal	2010	31/03/2015
	V2	Stockpile weathered surface coal layer 1	2013	31/03/2015
	V3	Stockpile "fresh" coal layer 2	2013	31/03/2015
	V4	Borehole OV125157A TM Seam Coal	2014	31/03/2015
	V5	Borehole OV125157A TL Seam Coal	2014	31/03/2015
	V6	Borehole OV125157A MS Seam Coal	2014	31/03/2015
	V7	Borehole OV125157A BM Seam Coal	2014	31/03/2015

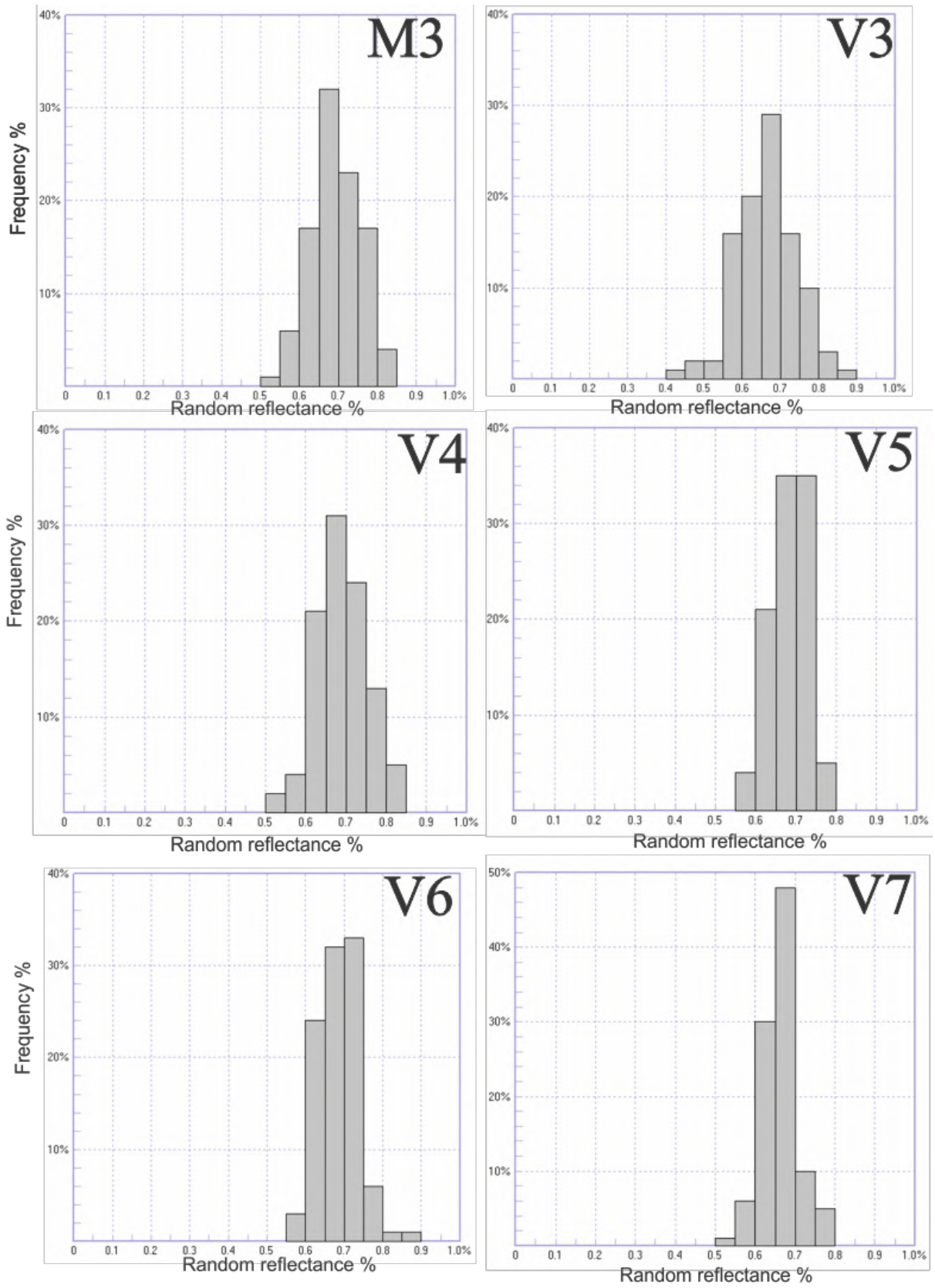
Appendix 2. Maceral point count results for Makhado Project and Vele Colliery coal samples

CoAL property	Sample ID	Maceral Group (inc. mm vol%)			MINERAL MATTER
		VITRINITE	INERTINITE	LIPTINITE	
Makhado Project	M1	80.10	9.80	0.00	9.40
	M2	80.50	12.80	0.00	6.10
	M3	71.40	16.70	0.50	10.70
	M4	59.00	6.00	0.00	34.30
	M5	75.20	15.40	0.00	8.70
Vele Colliery	V1	60.20	5.41	14.74	18.90
	V2	48.00	13.90	4.00	33.30
	V3	43.67	3.47	11.91	40.20
	V4	56.08	7.94	10.17	25.10
	V5	53.98	11.19	8.71	25.30
	V6	70.22	5.71	7.44	15.80
	V7	62.95	2.42	17.43	16.40

Appendix 3. Vitrinite reflectograms for Makhado Project and Vele Colliery coal samples



Appendix 3 continued



Appendix 4. ACA values by vol% for Makhado Project and Vele Colliery coal samples

Sample ID	Petrographic weathering features							
	Fresh organic	Uneven edges	Cracked edges	Prevalent cracks	Oxidation rims	Colour change	Altered minerals	Fresh minerals
M1	89.40	0.00	0.00	0.30	0.00	3.80	1.00	5.50
M2	74.50	3.20	7.00	0.50	2.80	6.30	3.20	2.50
M3	93.00	0.00	0.00	0.00	0.00	0.00	4.70	2.30
M4	73.50	0.00	0.00	0.50	0.00	1.00	18.00	7.00
M5	94.50	0.00	0.00	0.30	0.00	0.00	2.20	3.00
V1	81.50	0.00	0.00	0.50	0.00	0.00	1.00	17.00
V2	71.00	0.00	0.00	0.30	0.00	0.00	0.70	28.00
V3	62.35	0.00	0.00	0.50	0.00	0.25	2.20	34.70
V4	76.00	0.00	0.00	1.00	0.00	0.00	0.00	23.00
V5	80.50	0.00	0.00	0.50	0.00	0.00	0.00	19.00
V6	86.00	0.00	0.00	1.00	0.00	0.00	0.00	13.00
V7	74.90	0.00	0.00	5.40	0.00	0.00	0.00	19.70

Appendix 5. Proximate results for Makhado Project and Vele Colliery coal samples (Air dry basis)

CoAL property	Sample ID	Initial mass (g)	Moisture %	Ash %	Fixed Carbon %	Volatile matter %
Makhado Project	M1	1.0021	2.11	22.72	50.18	25.00
	M2	1.0056	1.39	17.68	54.41	26.52
	M3	1.0033	1.32	18.15	51.43	29.10
	M4	1.0007	2.29	44.05	19.93	33.73
	M5	1.0026	1.21	19.60	50.16	29.03
Vele Colliery	V1	1.0166	1.57	34.01	35.87	28.55
	V2	1.0009	1.52	45.89	29.01	23.58
	V3	1.0187	1.73	47.45	27.73	23.09
	V4	1.0143	1.42	32.23	34.18	32.16
	V5	1.0080	1.62	27.94	38.63	31.81
	V6	1.0142	1.67	26.85	39.94	31.54
	V7	1.0055	1.64	23.50	44.19	30.67

Appendix 6. Ultimate results (including calculated oxygen) for Makhado Project and Vele Colliery coal samples (Air dry basis)

Sample ID	Sample description	% C	% O	%H	% S	% N
M1	Stockpile 1: weathered surface coal layer 1	66.60	27.05	4.08	0.74	1.53
M2	Stockpile 1: "fresh" coal layer 2	71.40	21.79	4.31	0.84	1.66
M3	Stockpile 5: ROM bright coal	70.40	22.95	4.31	0.64	1.70
M4	Stockpile 5: ROM weathered surface coal	42.00	54.17	2.58	0.47	0.78
M5	Stockpile 5: ROM "fresh" coal	69.20	24.25	4.28	0.60	1.67
V1	Stockpile fridge coal	53.60	40.92	3.58	0.85	1.05
V2	Stockpile weathered surface coal layer 1	42.80	52.42	3.02	0.98	0.78
V3	Stockpile "fresh" coal layer 2	41.10	54.09	2.95	1.12	0.74
V4	Borehole OV125157A TM Seam Coal	53.00	33.58	3.46	9.14	0.82
V5	Borehole OV125157A TL Seam Coal	58.40	31.59	3.96	5.08	0.97
V6	Borehole OV125157A MS Seam Coal	58.50	30.72	3.86	5.78	1.14
V7	Borehole OV125157A BM Seam Coal	64.00	28.99	4.13	1.50	1.38

Appendix 7. Calorific values of Makhado Project and Vele Colliery coal samples

CoAL property	Sample ID	Sample description	Calorific Value (MJ/kg)
Makhado Project	M1	Stockpile 1: weathered surface coal layer 1	26.44
	M2	Stockpile 1: "fresh" coal layer 2	27.12
	M3	Stockpile 5: ROM bright coal	28.09
	M4	Stockpile 5: ROM weathered surface coal	14.81
	M5	Stockpile 5: ROM "fresh" coal	27.12
Vele Colliery	V1	Stockpile fridge coal	20.91
	V2	Stockpile weathered surface coal layer 1	16.31
	V3	Stockpile "fresh" coal layer 2	18.00
	V4	Borehole OV125157A TM Seam Coal	24.86
	V5	Borehole OV125157A TL Seam Coal	24.17
	V6	Borehole OV125157A MS Seam Coal	20.05
	V7	Borehole OV125157A BM Seam Coal	25.52

Appendix 8. Free Swelling Index results for Makhado Project and Vele Colliery coal samples (Air dry basis)

CoAL Project	Sample ID	Sample Description	FSI INDEX
Makhado Project	M1	Stockpile 1: weathered surface coal layer 1	0.0
	M2	Stockpile 1: "fresh" coal layer 2	7.5
	M3	Stockpile 5: ROM bright coal	8.0
	M4	Stockpile 5: ROM weathered surface coal	1.5
	M5	Stockpile 5: ROM "fresh" coal	7.5
Vele Colliery	V1	Stockpile fridge coal	1.0
	V2	Stockpile weathered surface coal	7.0
	V3	Stockpile "fresh" coal	0.0
	V4	Borehole OV125157A TM Seam Coal	2.5
	V5	Borehole OV125157A TL Seam Coal	4.0
	V6	Borehole OV125157A MS Seam Coal	3.5
	V7	Borehole OV125157A BM Seam Coal	3.0

Appendix 9. Eskom quality specifications for South African thermal coal products after Steyn and Minnitt, (2010)

Parameter	Basis	Eskom	Domestic	Export
Calorific value	NAR	21-24MJ/kg	24.5-27.5MJ/kg	5850-6000kcal/kg 27.5MJ/kg
Total moisture	% AR	8-12	8-12	8-12
Ash	% AR	25-34	15-21	15
Volatile matter	% AR	20-30	23-30	22-20
Sulphur	% AR	0-2	0-1.5	0-1.0

Appendix 10. CoAL's coking coal classification scheme (Sparrow et al., 2013)

**BASED ON COKE MARKETING NOMENCLATURE,  
NORMAL VELE COAL IS CLASSIFIED AS A SOFT COKING COAL**



**Vele**

Other Limpopo coals: Jutland \ Chapudi \ Makhado  
Wildebeestehoeck \ Generaal \ Mount Stuart

Product Type	Volatile Matter	Indicative VM content (ad), %	Max vitrinite reflectance (Ro), %	CSN (FSD)	General Use
Hard (HCC)	LV	<22	1.3 – 1.7	7-9	Coking
	MV	22 - 28	1.1 – 1.5		Coking
	HV	>28	0.95 – 1.1		Coking
Semi-Hard (SHCC)	As above, but typically higher ash hard coking coals			6-8	Coking
Soft (or weak) (SCC)		Predominantly HV	< 1.0	4-8	Coking
Semi-soft (SSCC)		Predominantly HV	< 1.0	1-4	Coking/PCI
LV, semi anthracites and anthracites		<17	> 1.5	0	PCI

LV = low volatile. MV = medium volatile. HV = high volatile

**OTHER LIMPOPO COALS ARE CLASSIFIED AS HARD COKING COALS ON THIS CONVENTIONAL CLASSIFICATION – WHEN NOT NEAR FAULTS**