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Coal quality and uranium distribution in Springbok Flats Coalfield samples

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Synopsis

The presence of coal in the Springbok Flats Coalfield (SFC) has been known since the beginning of the 1900s. However, the SFC has not been mined to any degree of economic profit, mostly due to the presence of uranium in the coal. Five boreholes were drilled in the SFC (BH1 to BH5); BH5 intersected two coal zones, the other boreholes intersected one coal zone. Coal samples were collected, selected, and characterized using proximate, ultimate, and calorific value analyses. X-ray fluorescence, instrumental neutron activation analysis, and inductively coupled plasma mass spectrometry were used to determine uranium content. The BH1 intersection and the upper coal zone in BH5 had ash contents higher than 50% and were considered to be primarily carbonaceous shale. BH2 was observed to have better coal quality, resembling typical South African bituminous coal used in local electricity generation. The highest uranium content was found in BH3 (up to 199 mg kg^{-1}, followed by BH2 and BH1. BH4, the upper coal zone in BH5, and the lower coal zone in BH5 all had uranium contents averaging less than 10 mg kg^{-1}. Uranium in the SFC samples was found both in the coal and carbonaceous shale. For all boreholes except BH5, uranium is concentrated within the uppermost 1 m of the coal zone. X-ray fluorescence was the preferred analytical technique since the analysis gave consistent results that compared well with instrumental neutron activation analysis results.

Keywords
Springbok Flats Coalfield, coal quality, uranium.

Introduction

Coal in South Africa is found in 19 coalfields located in the middle and northern sections of the country (Jeffrey, 2005). The main coal mining areas in the country at present are in the Witbank-Middelburg, Waterberg, and Sasolburg coalfields of the Mpumalanga and Limpopo provinces. The Springbok Flats Coalfield (SFC), located in the Limpopo Province, has not been mined to any degree of economic profit, predominantly due to the presence of uranium in the coal. The first coordinated exploration programme was conducted by the Council for Geoscience (CGS) between 1952 and 1957, where 27 boreholes were drilled in the northeastern portion of the SFC (Visser and Van der Merwe, 1959). Further exploration by the CGS in the western and south-central portions of the coalfield was halted in 1972 when uranium was detected in the upper Ecca coal zone (Christie, 1989). Similar to most materials in nature, coal contains small quantities of naturally occurring radionuclides such as ^{40}K, ^{238}U, ^{232}Th, and their decay products (Papastefanou, 2010). The Medium Rank C bituminous coal zones in the Springbok Flats Basin, hosted in the coal horizons of the Late Permian in the uppermost part of the Hammanskraal Formation within the SFC basin, have a significant uranium content (Cole, 1998). Further, the uranium in the SFC is believed to be disseminated throughout the coal and the associated carbonaceous shale (Christie, 1989). This is particularly noteworthy as it means that this coalfield could potentially have areas that are rich in uranium but with very little coal present. This gives metallurgists the opportunity to concentrate on extracting uranium from the carbonaceous horizons without worrying about the effects of the uranium extraction process on coal and its ability to combust post-extraction.

Overall, there is limited public domain research pertaining to the quality of coal present in the SFC as well as the uranium associated with the coal and carbonaceous horizons. This knowledge is strategically important in determining the viability of the SFC as a potential source of either coal or uranium, thus addressing two key energy markets. The Department of Mineral Resources (DMR) has seen a need for South African coal researchers and metallurgists to investigate cleaner coal processing and energy production, and has thus created intervention strategies for the optimal beneficiation of coal (Department of Mineral Resources, 2011), which, among numerous other objectives, seek to invest in metallurgical research on the

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quality of the coal associated with uranium in the SFC. Thus, the aim of this study is to assess the quality of newly acquired borehole core coal samples, and to accurately determine the uranium content in those samples. Once an understanding of the uranium content and distribution is obtained, the extraction of the uranium from specific localities can be considered; this discussion is targeted for a future publication.

Experimental procedure

The study covers five boreholes drilled in the SFC (BH1 to BH5) in 2013. BH1 to BH4 intersected a single coal zone, while BH5 intercepted two coal zones, an upper and a lower zone. Thus, six coal localities were included in this research. Figure 1 depicts the five farms where the drilling occurred. The boreholes were drilled up to a depth of 450 m, recovering a 4 cm cylindrical core. The recovered cores were placed in 1.5 m long core trays, logged, and stored at the CGS Donkerhoek core shed, Pretoria, for about a month prior to sampling. One quarter section of core from each coal horizon was removed (the rest retained for future projects), and milled using a Reutsch mill to obtain a -1 mm split (retained for coal petrography) and a subsequent -250 µm sample. A MACSALAB design rotary cascade splitter was used to obtain a representative -250 µm split for geochemical analysis (proximate, ultimate, CV, uranium determination). All samples were studied on an as-received basis. Proximate analysis was conducted at the CGS coal laboratory following ISO 1171:1981, SANS 5924:2009, and ISO 562:1981. Leco CHN and Leco S instruments were used to determine ultimate analysis using ISO 17247:2013. The CV data was obtained using a Parr 3600 bomb calorimeter where net CV (NCV) was used as the measure of CV. To determine uranium content, X-ray fluorescence (XRF) data was processed using a PANalytical wavelength-dispersive Axios X-ray fluorescence spectrometer. Inductive coupled plasma–mass spectrometry (ICP-MS) analysis was conducted using a Bruker 500 MHz NMR spectrometer. Eleven samples that revealed an uranium content higher than 10 mg kg^{-1} were subjected to instrumental neutron activation analysis (INAA) to confirm the results obtained from ICP-MS and XRF.

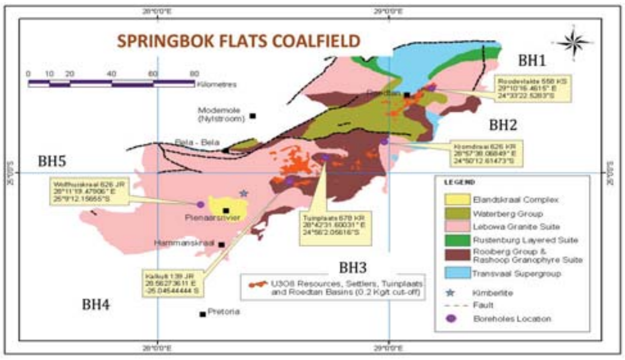

Figure 1 – Borehole localities

Coal quality results

Figure 2 shows the major coal quality parameters relative to depth for each borehole coal zone, with the actual data reported in Tables I–V for each of the coal zones sampled. Photographs of the coal zone intersections are provided in Figure 3.

BH1: The volatile matter content was highest in the samples proximal to the roof of the coal zone where the bright bands were observed. The average ash content for BH1 peaked at 88.4%, indicative of carbonaceous shale horizons or partings in the coal zone (Table I). The CV peaked at 4.2 MJ/kg, which

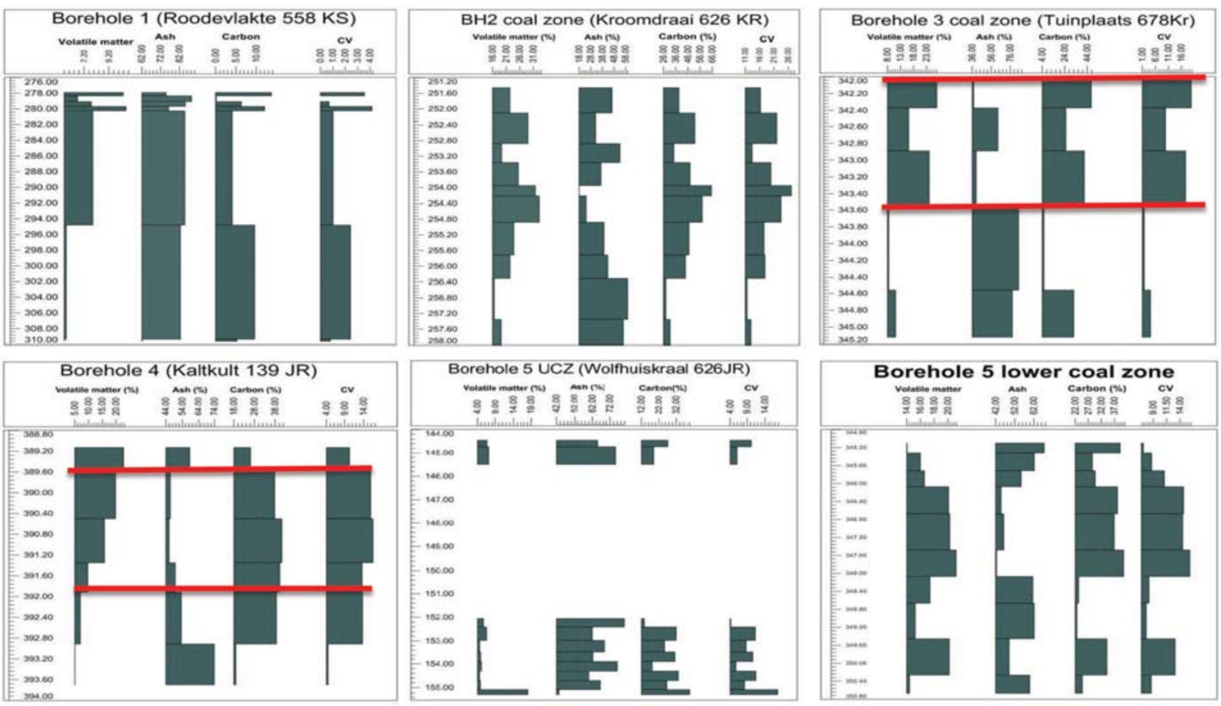

Figure 2 – Borehole logs, showing major coal quality parameters

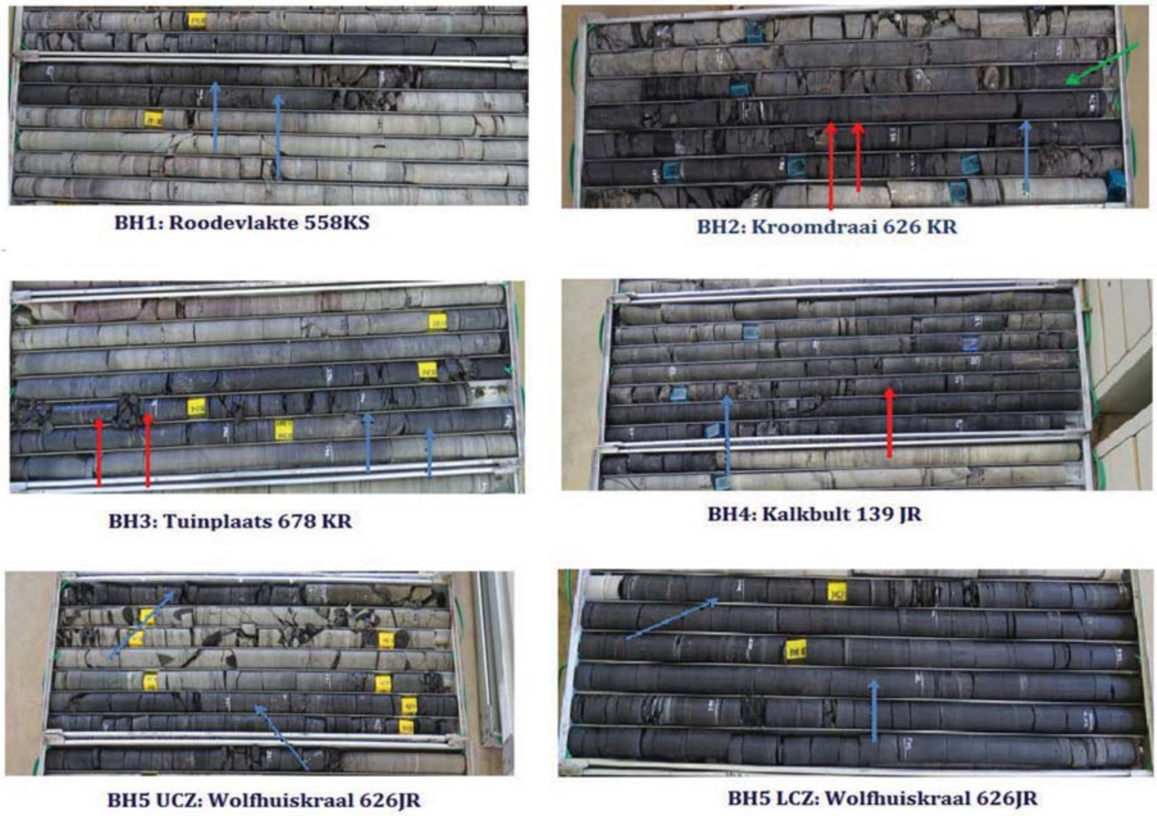

Figure 3 – Photographs of the six coal zone intersections

indicates a coal zone that is virtually incombustible. It was thus concluded that the whole coal zone sampled is predominately carbonaceous shale instead of coal as defined by SANS 10320:2004.

BH2: Relatively good CV data was obtained for samples in BH2, peaking at 27 MJ/kg (Table II). However, the sulphur content was alarmingly high for a number of coal samples, reaching a maximum of 8.9%. Sulphur was found to be abundant in the samples with high CV and volatile matter, and low ash content. The ash content was higher towards the floor of the coal zone. The majority of the coal zone registered ash contents well below 50%, indicating that the zone is made up predominately of coal.

BH3: When one considers the entire coal zone in BH3, it is can be noted that 60% of the 3.6 m coal zone contained samples with ash contents higher than 50% (Table III). The remaining 40% of the samples recorded CV and volatile matter averages of 18.7 MJ/kg and 25.5% respectively. Similar to BH2, these samples also recorded a higher average sulphur content (3.5%).

BH4: The ash content in BH4 was fairly constant in all samples (46–58.4%), except for the sample close to the floor of the seam. Table IV shows that the coal quality in BH4 is very poor, supported by the low CV that peaked at 16.5 MJ/kg. Similar to the other boreholes, the sulphur content was highest (4.2%) in the regions where the coal quality was better than the surrounding samples.

BH5 upper coal zone: 90% of the coal zone had an ash content higher than 50%. The lowermost sample was of comparatively better coal quality, with a CV and ash content

Table I

Coal quality data, BH1

Depth (m)	Sample number	Volatile matter (%)	Ash (%)	Carbon (%)	Sulphur (%)	NCV (MJ/kg)
277.0–277.9	1436	10.4	74.9	13.7	0.2	3.5
278.0–278.8	1437	6.7	88.4	0.2	0.2	0
278.8–279.4	1438	7.8	85.1	6.3	0.2	0.7
279.4–280.0	1439	10.6	76.2	12.1	0.2	4.2
280.0–280.5	1440	7.9	84.9	4.1	0.1	1.0
308.7–309.1	1441	5.8	82.5	9.6	0.1	2.4
309.1–309.6	1442	5.6	62.2	5.3	0.1	0.8
Average		**7.8**	**79.2**	**7.3**	**0.2**	**1.8**

Table II

Coal quality data, BH2

Depth (m)	Sample number	Volatile matter (%)	Ash (%)	Carbon (%)	Sulphur (%)	NCV (MJ/kg)
251.34–251.4	1426	22.4	46.2	38.6	4.4	16.3
252.30–252.7	1427	29	32.1	51.7	8.9	22
252.75–253.0	1428	19.3	53.1	34.4	1	13.4
253.12–253.7	1429	25.4	36.8	47.8	2.5	20
253.72– 254.1	1430	31.8	18.1	65.3	2.1	27
254.14–254.2	1431	33.2	24	57.7	7.2	23.5
254.6–255.5	1432	23.8	39.3	46.8	1.4	17.5
255.5–255.93	1433	22.4	42.6	44	0.5	17.9
256.33–256.7	1434	16.6	59.7	27.9	1.6	11.7
257.7–258.0	1435	19.1	56	31.4	1.9	12.9
Average		**24.3**	**40.8**	**44.6**	**3.2**	**18.2**

Table III

Coal quality data, BH3

Depth (m)	Sample number	Volatile matter (%)	Ash (%)	Carbon (%)	Sulphur (%)	CV (MJ/kg)
341.52–342.04	1421	27	37.3	47.6	3.5	19.8
342.1.0–342.7	1422	16.2	63	25	0.9	9.9
342.7–343.08	1423	24	40.1	41.4	3.5	17.6
343.56–344.0	1424	8.5	84.7	5.5	1.1	1.72
344.0–344.3	1425	11.1	78.2	32	3.6	4.1
Average		**17.4**	**60.7**	**30.3**	**2.5**	**10.6**

Table IV

Coal quality data, BH4

Depth (m)	Sample number	Volatile matter (%)	Ash (%)	Carbon (%)	Sulphur (%)	CV (MJ/kg)
387.81–389.13	1443	22.7	58.4	26.1	4.2	10.3
389.1–390.0	1444	19.8	46.7	37.8	4.1	15.8
390.0–391.0	1445	15.8	46.0	41.2	1.0	16.5
391.0—391.7	1446	9.7	49.7	40.2	0.6	13.6
391.7–392.13	1447	7.0	53.4	38.6	0.6	13.7
393.0–393.7	1449	5.0	73.9	19.0	0.3	4.4
Average		**13.3**	**54.7**	**33.8**	**1.8**	**12.4**

Table V

Coal quality data, BH5 upper coal zone

Depth (m)	Sample number	Volatile matter (%)	Ash (%)	Carbon (%)	Sulphur (%)	CV (MJ/kg)
143.90–144.45	1401	6.6	64.7	27.3	3.6	10.2
144.5–145.0	1402	7.0	75.1	18.9	2.2	5.9
151.6–152.10	1403	5.6	79.8	13.6	0.7	4.2
152.10–152.72	1404	6.5	61.9	32.0	0.8	11.3
152.72–153.23	1405	4.6	69.0	25.4	0.7	8.7
153.23–153.70	1406	4.8	62.2	31.2	0.9	10.6
153.7–154.1	1407	5.0	76.0	18.0	0.7	5.6
154.1–154.51	1408	4.6	60.0	33.1	1.0	11.4
154.51–154.9	1409	4.8	66.3	26.7	0.5	8.3
154.9–155.27	1410	18.1	43.3	40.0	12.4	17.8
Average		**6.8**	**65.8**	**26.6**	**2.4**	**9.4**

Table VI

Coal quality data, BH5 lower coal zone

Depth (m)	Sample number	Volatile matter (%)	Ash (%)	Carbon (%)	Sulphur (%)	NCV (MJ/kg)
344.67–345.10	1411	14.1	67.5	39.2	0.3	7.2
345.10–345.54	1412	16.0	62.2	28.5	0.3	8.7
345.54–345.89	1413	16.6	55.3	29.6	0.3	11.1
345.89–346.25	1414	20.1	44.8	38.2	0.4	14.6
346.25–347.10	1415	20.3	46.2	36.9	0.7	14.5
347.10–347.86	1416	21.2	42.4	40.5	0.6	15.8
347.86–348.28	1417	17.4	61.5	23.3	0.2	8.2
348.28–349.05	1418	15.2	62.2	22.6	0.2	7.7
349.05–349.86	1419	20.2	49.1	34.1	0.3	13.1
349.86–350.67	1420	14.4	59.8	24.7	1.5	8.6
Average		**17.6**	**55.1**	**31.8**	**0.5**	**11.0**

of 17.8 MJ/kg and 43.3% respectively (Table V). The proximate analysis and CV results for the upper coal zone of BH5 are below the limits of typical South African coals, and the results resemble those of carbonaceous shale instead of coal (Martins *et al.*, 2010).

BH5 lower coal zone coal: The ash content in the upper coal zone in BH5 was high, with no sample recording ash content less than 40%. The majority of the coal zone reported ash contents higher than 50%. The CV was low for all samples, peaking at a moderate 15.8 MJ/kg (Table VI). The proximate analysis and CV results for the upper coal zone of BH5 are below the limits of typical South African coals, and the results resemble those of carbonaceous shale instead of coal (Martins *et al.*, 2010).

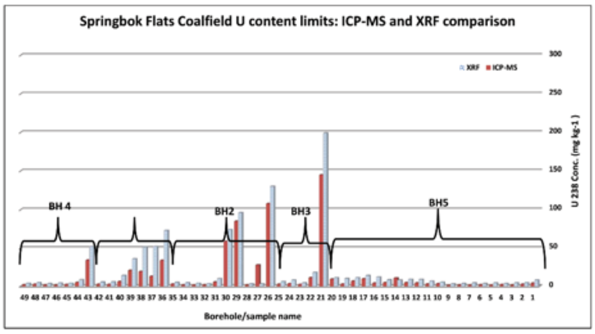

Figure 4 – Uranium assays – comparison of ICP-MS and XRF results

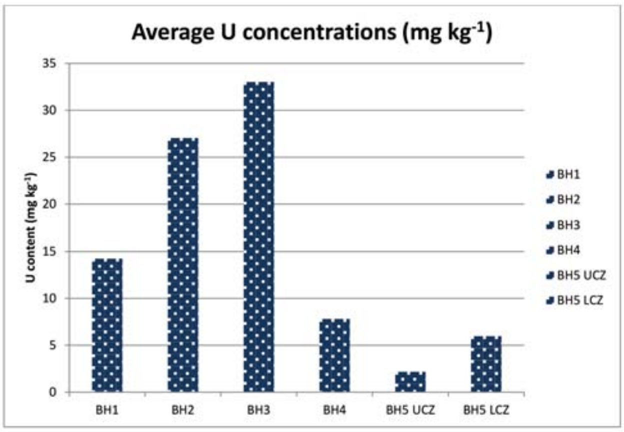

Figure 5 – Average uranium values in the six coal zone intersections

Uranium content determined in coal samples

Figure 4 provides the uranium content for each of the five borehole core samples. Figure 5 depicts the average concentrations. As the samples included the high-ash carbonaceous shales, the uranium values are not indicative of only the coal-rich zones.

ICP-MS produced the lowest uranium values of all the analytical techniques used, and XRF consistently produced the highest uranium values (Table VII). INAA (on selected samples) reported higher uranium values than ICP-MS, and the results were closer to the XRF values (Table VII). The variation in some ICP-MS results led to the conclusion that an error (either technical or human) could have occurred during analysis. Due to the consistent results provided by XRF, these results were used in the subsequent interpretations and discussion.

BH1: The uranium content in the BH1 samples ranged from 5.3 mg kg^{-1} to 73 mg kg^{-1}. The highest uranium values occurred at the roof of the coal zone. Since the BH1 sample consisted entirely of carbonaceous shale, this gives metallurgists the opportunity to concentrate on extracting uranium from these horizons without worrying about the effects of the uranium extraction process on coal and its combustion qualities post-extraction.

BH2: Samples from BH2 had uranium contents that ranged from 2.9 mg kg^{-1} to 130 mg kg^{-1}. The uranium content was highest where the ash content was < 50%, with a significant peak occurring towards the middle of the coal zone (96 mg kg^{-1}), also a coal-rich zone.

BH3: The uranium content of sample 1421 was 199 mg kg^{-1}, the highest of all samples. Relative to coal quality, both carbonaceous shale regions and coal regions contained uranium; however, the coal-rich samples yielded higher uranium values.

Table VII

Comparison of uranium assay results by XRF, INAA, and ICP-MS

Sample number	Borehole	U [XRF] (ppm)	U [INAA] (ppm)	U [ICP-MS] (ppm)
1416	BH5 LCZ	12	8.9	8.6
1417	BH5 LCZ	14	11.8	9.8
1421	BH3	199	161	145.9
1422	BH3	18	15.6	11.3
1429	BH2	96	86.9	85.9
1436	BH1	73	64.6	34.1
1437	BH1	52	43.2	13
1438	BH1	51	39.4	19.4
1439	BH1	36	29.3	20.9
1440	BH1	14	11.5	6.2
1443	BH4	52	33.5	34.4

BH4: Maximum uranium content was detected where the coal zone reported an ash content of 58.4%. Similar to previous boreholes, uranium was found to be abundant at the roof of the coal zone.

BH5 upper coal zone: Similar to BH1, the entire coal zone was made up of carbonaceous shale, and thus uranium in this coal zone occurred in the carbonaceous shale. Unlike the other intersections, where the maximum uranium content was found at the roof, the uranium was distributed fairly evenly throughout the coal zone with a difference of 5 mg kg^{-1} between the maximum and the minimum values determined.

BH5 lower coal zone coal: The lower coal zone coal in BH5 returned a peak uranium content of 14 mg kg^{-1}. Similar to BH2, other peaks of interest were found further down the coal zone. The uranium was distributed in some areas that consisted predominately of coal, and in areas that were predominately carbonaceous shale.

Discussion on coal quality

It was apparent from visual inspection that the coal zones sampled do not consist only of coal, but carbonaceous and sandstone horizons are interbedded in the coal zone, as shown in Figure 2. The darker horizons in the cores represent the coal zones, made up of interbedded coal and carbonaceous shale. BH1 was dominated by carbonaceous shale (blue arrows) with very few visible bright coal bands. BH2 had significantly more bright coal bands compared to BH1, where the coal zone appeared to be made up predominately of bright coal (red arrows to left) interbedded with carbonaceous shale (blue arrow to right). Calcite cleats (green arrow top right) were visible in some areas in the coal zone. BH3 coal zone consisted of bright coal (red arrows to left) clustered at the top of the coal zone; the bright bands of coal diminished and carbonaceous shale (blue arrows to right) dominated further down the coal zone. BH4 contained very few bright bands of coal. Carbonaceous shale dominated the top of the coal zone, with regular bright coal bands a little further down, towards the middle of the coal zone, which diminished again towards the bottom of the coal zone. The upper coal zone in BH5 had very few bright bands of coal, with large areas showing no bright bands of coal at all. Carbonaceous shale (blue arrows) dominated the entire coal zone. The lower coal zone in BH5 also had very few coal and carbonaceous shale (blue arrows) dominated the entire coal zone.

The average ash content of boreholes BH1, BH3, BH4, and the upper and lower coal zones in BH5, were far greater than the 40.3% recorded in the CGS database, and higher than the 30–55% estimated by De Jager (1983), while also being higher than the 30–35% ash inferred by the Petric Commission (1975). BH2 average ash content of 40.8% agrees with the 40.3% recorded in the CGS database for coal from the same locality, and is in line with the 30–55% ash content estimated by De Jager (1983). BH2 coal quality resembles a typical South African bituminous coal (Falcon and Ham, 1988; Pinhiero, 1999); however, the sulphur content average of 3.2% is higher than the 2.8% recorded in the CGS database for samples from the same region, while also being higher than 0.4–1.29% reported by Wagner and Hlatshwayo (2005) for Highveld coals, and 1.47% found by Roberts (2008) for samples in Mpumalanga.

Discussion on uranium content

Figure 5 shows the average uranium contents for each of the boreholes studied. All borehole coal zones studied had uranium contents averaging higher than the 2 mg kg^{-1} world average reported by Swaine (1990), and the 2.9 ppm global average for coals (Ketris and Yudovich 2009). Ren *et al.*, (1999) determined a 7.52 mg kg^{-1} arithmetic mean uranium content in Chinese coals.

The uranium content relative to selected coal quality results can be seen in Figure 6. Uranium in the SFC samples was disseminated throughout the coal and carbonaceous shale, as reported by Cole (2009) and Hancox and Gotz (2014). The uranium in the coal zones was generally restricted to a single layer, usually the highest in the local sequence, except in BH2 and the lower coal zone coal in BH5, where uranium mineralization was found in multiple locations in the coal zone. This finding is in agreement with Cole (2009), Christie (1989), and Nel (2012). BH3 had the highest average uranium content, and the highest uranium content (199 mg kg^{-1}) was determined in a sample from this coal zone. BH4 and the upper and lower coal zones in BH5 all had average uranium contents less than 10 mg kg^{-1} (which is still high in the global context).

Coal quality and uranium distribution in Springbok Flats Coalfield samples

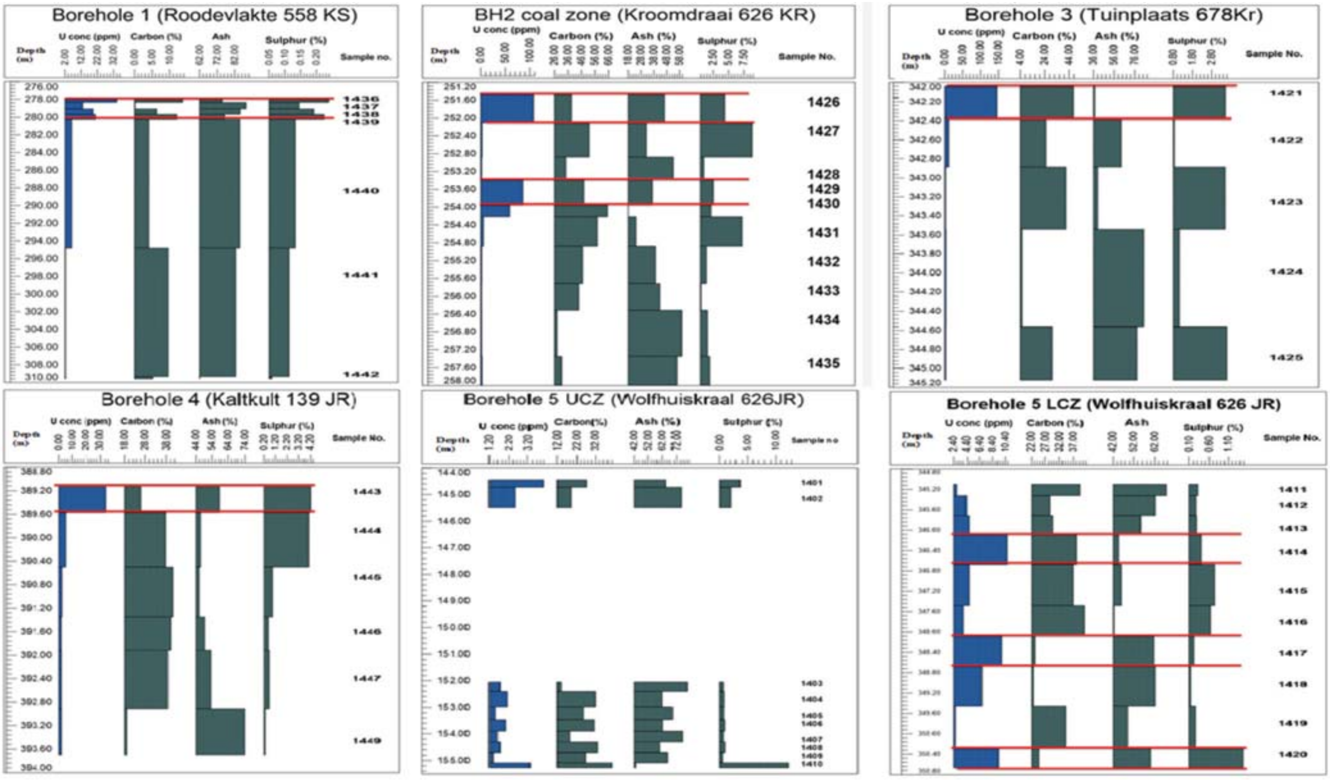

Figure 6 – Relationship between uranium content and coal quality

Conclusions on coal quality

In summary, BH1 and the upper coal zone in BH5 reported ash contents higher than 50% for all the samples collected throughout the coal zone; these coal zones are made up almost entirely of carbonaceous shale and are thus not suitable for coal exploitation. BH2 samples reported coal qualities that resemble a typical South African bituminous coal and could potentially be of economic benefit to the country, depending on the available tonnages in the area. BH3 and BH4 had coal horizons that are potentially mineable, and could be of use in the coal conversion industries.

Conclusions on uranium content

INAA reported higher uranium values than ICP-MS, with the results being closer to the XRF values; which were the highest for all the samples. All borehole coal zones studied had an average uranium content higher than the 2 mg kg^{-1} world average reported by Swaine (1990). Uranium in the coal zones sampled was generally restricted to a single layer, usually the highest in the local sequence, except in BH2 and the lower coal zone coal in BH5, where uranium mineralization occurred in multiple locations in the coal zone. This finding is in agreement with Cole (2009) and Nel (2012). Overall, uranium in the SFC samples was disseminated throughout the coal and carbonaceous shale horizons, findings supported by Hancox and Gotz (2014). BH4 and the upper and lower coal zones in BH5 all had average uranium content less than 10 mg kg^{-1} , but BH2 and BH3 had average values over 25 mg kg^{-1}. Table VII shows the samples selected for INAA analyses to confirm the high uranium values; these samples were selected for leaching experiments to extract and concentrate the uranium. This work will be reported in a future publication.

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