Geochemical Characterisation of the Witbank Coalfield Geological Strata and Assessment of Potential Metal Impact on the Receiving Environment

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Abstract

South Africa is a country that has a number of commodities, one being coal, which is one of the seventh-largest coal producers globally and has produced approximately 260.5 million tons in 2017. Coal is the main source of energy in South Africa with most of the coal being mined in the Mpumalanga Province, mainly located in the Witbank and Highveld Coalfield. Much has been written about the structural, environmental and depositional environment of the Witbank coalfields, but limited information is available on the mineralogy, petrology and geochemistry of the rocks hosting the economically-important coal beds. To augment this shortcoming, coal samples were collected and the associated non-coal strata from approximately 140 m core located in the Goedehoop Colliery of the Witbank Coalfield, for geochemical characterisation. Key to this is the identification of minerals occurring in coal to an extent of its genesis and the understanding of the distribution of minerals throughout different sedimentary units. Understanding the extent to which the geological units of the Witbank Coalfield contributes to the elevated metal concentrations in stream sediments downstream. Different analytical techniques have been used including proximate analysis, ultimate analysis, optical microscopy, X-ray diffraction and X-ray fluorescence spectrometry. The coals are sub-bituminous to high-volatile bituminous in rank typically contains of vitrinite as the most dominant maceral group followed by the inertinite and liptinite group. The mineralogy of the coal seams are mainly kaolinite, quartz with minor proportions of pyrite and carbonates and anatase and mica as accessory minerals. The minerals found in the hanging and footwalls of the coal seams are mainly clay, quartz, K-feldspar, plagioclase with minor proportions of pyrite, carbonates, mica minerals and hematite with anatase as accessory minerals. Stream sediments was used to assess the downstream metal loadings. Stream sediments was used to assess the downstream metal loadings and reflect the mineralogical composition of the source rock. This means that the coal seams were formed in anaerobic conditions due to high groundwater table levels in the peat mire, with environmental conditions returning to a lower ground-water table and oxidising conditions as seen by the presence of fusinite and semi-fusinite. Enrichment of elements like As, Pb, U, Zn, Yb, Se, W and Co in the stream sediments suggests some loss of mobile components from the source rocks during weathering and in metal transportation process. The enrichment can be attributed to the anthropogenic factor downstream of the source rock.

Introduction

South Africa is a country that has a number of commodities, one being coal which is one of the seventh-largest coal producers globally and has produced approximately 260.5 million tons in 2017. The five largest coal mining companies that supply over 80% include South32's South Africa Energy Coal, Anglo American Thermal Coal, Glencore Xstrata, Sasol Mining and Exxaro. Coal is the main source of energy in South Africa with most of the coal being mined in the Mpumalanga Province. Much has been written about the structural, environmental and depositional environment of the Witbank coalfields, but limited information is available on the mineralogy, petrology and geochemistry of the rocks hosting the economically-important coal beds. To augment this shortcoming, coal samples were collected from four coal seams and associated non-coal strata from the Goedehoop Colliery in the Witbank coalfields for characterisation.

The current study was motivated by elevated metal concentrations in stream sediments of the Witbank coalfields as a result of metal accumulation caused by acid mine drainage processes. This study of the geochemical characteristics of coals and associated geological strata from the Witbank Coalfields was undertaken to establish a relationship between mineralogy, major and trace element content and petrology of the coal beds. Understanding of mineralogy, petrology and geochemistry of coal seams in the Goedehoop area will allow an extension to the genetic processes forming the coalfields of the broader Witbank area. The downstream assessment is done by assessing and comparing the geochemistry of the stream sediments collected to represent the basin and the geochemistry of the Witbank Coalfield rock units. As indicated by Thomas (2012), coal is sediment, bio-clastic in nature, and comprises of carbon, mineral matter and macerals, which is an ignitable material. McCarthy (2005) characterises coal as a sedimentary rock which comprises mainly of carbon, formed by the accumulation of a mixture of organic and inorganic matter. Coal comprises of a mixture of organic matter (macerals), inorganic matter (minerals) and liquids. The organic matter is composed of carbon, hydrogen and oxygen, together with smaller measures of nitrogen and sulphur (Weeber, 1996).

As indicated by Weeber (1996), the Witbank Coalfield is located in the northern part of the well-known Karoo Basin where the major coal deposits in South Africa are found. The Witbank Coalfield is situated on the northern fringe of the south-eastern portion of the Transvaal Coalfield (Graham, 1931) (and in the southern part of the Olifants Catchment), and is one of the major coal mining areas in South Africa. Two borehole cores were obtained at Goedehoop colliery in the Goedehoop area which is situated in the north-east of Witbank (Fig. 1).

Geological setting

The tectonic setting of the Karoo Basin has been discussed by Cairncross (2001) and Hobday (1987). The Permian to early Triassic coals were deposited on Gondwana, in the back arc of the Gondwana orogenic belt, during a period of warming (Banks *et al.*, 2011). A period of glacial sedimentation during the Permo-carboniferous glaciation marks the commencement of numerous phases giving rise to the formation of the Karoo Supergroup in which the majority of the coal deposits of southern Africa were deposited (Thomas *et al.*, 1993). The Karoo Supergroup is a thick succession of sedimentary rocks deposited approximately 300 to 180 million years ago (Banks *et al.*, 2011). The Karoo Supergroup is subdivided into the Dwyka, Ecca, Beaufort, Stormberg and Drakensberg Groups, of which the Vryheid Formation of the Ecca Group is coal-bearing horizon (Table 1 and Fig. 2) (Banks *et al.*, 2011). According to Vermeulen and Usher (2006), the Dwyka Group consists of tillite, siltstone and sometimes a thin shale development. The sediments of the Ecca Group were deposited on an undulating Karoo floor, which influenced the distribution and thickness of the sedimentary formations as well as the associated coal seams (Pinetown, 2006). The strata of the Karoo Basin consist fundamentally of sandstone, carbonaceous shale, siltstone, minor conglomerate and several coal seams (Cairncross, 2001). The extensive variety of sedimentary environments and structural setting within which the coal seams were deposited combined with the range of ages, climates and plants give rise to the numerous differences in terms of organic and inorganic matter and the degree of maturity of the coal seams (Falcon, 1986). The Witbank Coalfield lies towards the northern extent of the Karoo Basin where the sediment thins and the Dwyka Group reposes on the Transvaal Supergroup, the Waterberg Group and volcanic rocks associated with the

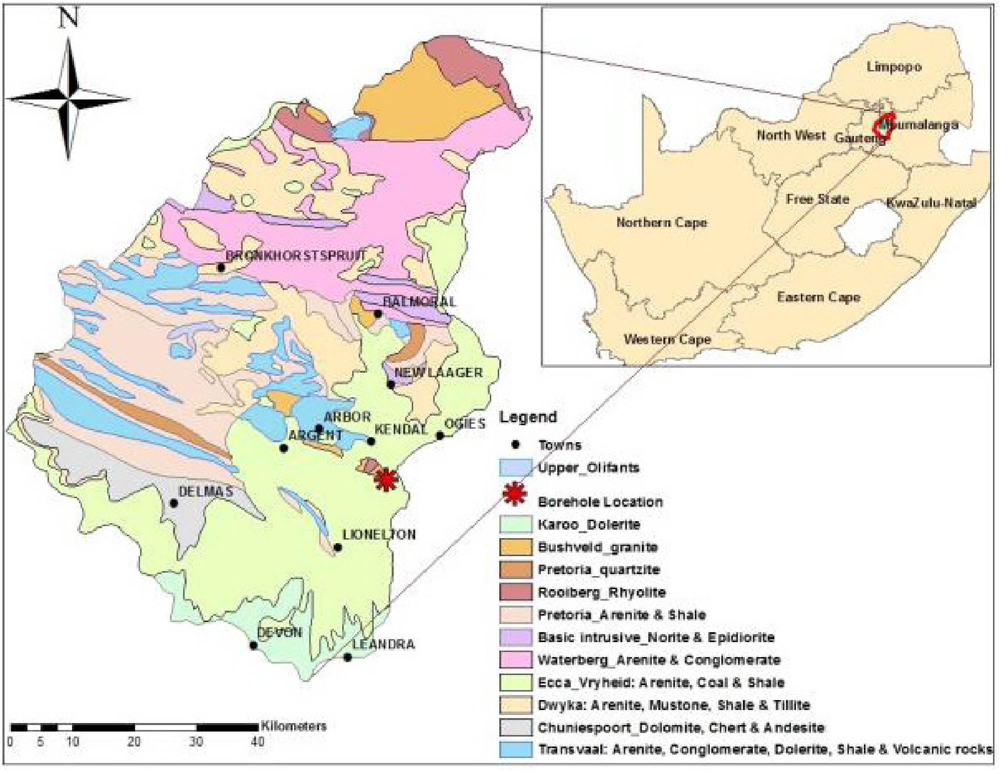

Figure 1 *Simplified geological map of the upper Olifants sub-catchment and the location of the borehole*

Table 1 *Lithostratigraphic units for Karoo Supergroup (SACS, 1980 and Catuneanu et al., 2005)*

Supergroup	Age (Ma)		Group	Formation
Karoo	140	Jurassic and Upper Triassic	Drakensberg	Draknesberg
	195			Clarens
	225	Triassic		Elliot
	230	Upper Permian and lower Triassic	Beaufort	Burgersdop
				Katberg
				Balfour
				Koonap & Middleton
	260	Middle Permian	Upper Ecca	Volksrust
			Middle Ecca	Vryheid
			Lower Ecca	Pietmaritzburg
	300	Late Carboniferous	Dwyka	

Bushveld Complex (Banks *et al*., 2011). The strata comprise of surface soils, carbonaceous cross-bedded and interlaminated sandstone or siltstone, sandstones, shale, coal seams, and intrusions of dolerite dykes or sills (Graham, 1931; Smith & Whittaker, 1986). Figure 2 shows Coal Seam No. 1 to 5 encountered in the Witbank Coalfield. Seam No. 1 is the lowest and either lays on the tillite or is separated from it by shale and sandstone bands of the Dwyka Group (Graham, 1931; Smith & Whittaker, 1986).

Methodology

As shown in Figure 2, the cores of two complete drilled boreholes were provided by Goedehoop colliery in the Goedehoop area which is situated in the north-east of Witbank town. Geographically, it is situated at approximately 29˚80' 77, 4" S and 025˚ 90' 24, 5" E, at about 1664.88 m above sea level and 29˚84' 34, 2" S and 025˚ 64' 99, 4" E, at about 1733.37 m above sea level. Four coal and 54 non-coal bed samples were collected from ZFN 1512, while for ZFN 1510 borehole, four coal and 61 non-coal bed samples were obtained to provide a mineralogical and geochemical profile. Large chips were selected at random from each coal seam and non-coal units for sample preparation of polished sections and/or thin-sections. Polished stubs and the remainder of each sample were crushed into smaller chips using a rock crusher. Samples were crushed using a 3 x 5 mm jaw crusher which reduces the size of the rock, ores and other brittle samples from >8 cm to ±4 mm. The particle size of samples from crushing and splitting needs to be reduced to <75 µm and this is accomplished by means of a swing mill. The milling pots used are made of carbon steel to ensure minimal contamination. 75 ml of sample were milled for 6 minutes in the

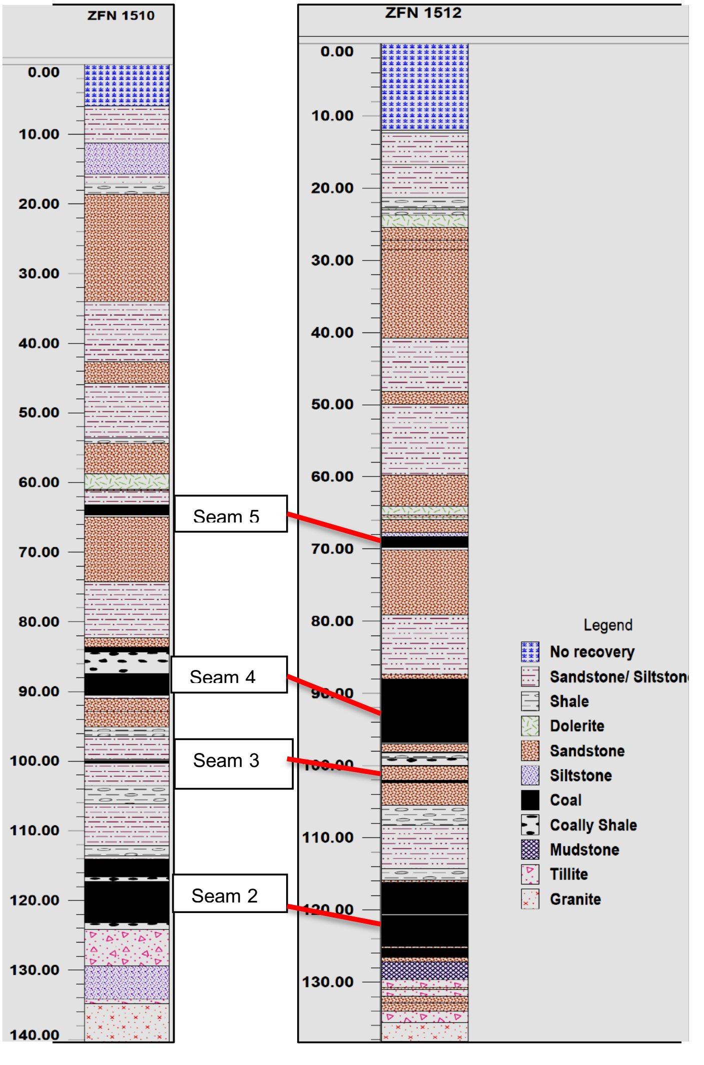

Figure 2 *Core logs of the Witbank Coalfield in the Goedehoop area*

swing mill. The samples were analysed for major and traces by XRF, mineralogy by X-ray diffraction (XRD) and petrology based on different crystallographies. The four coal seam were also analysed for proximate and ultimate analysis. The 240 stream sediment were analysed by XRF for the major and traces elements.

Results

The downstream assessment is done by assessing and comparing the geochemistry of the stream sediments collected to represent the basin and the geochemistry of the Witbank Coalfield rock units. It is important to note that the geochemistry of stream sediments generally reflects the mineralogical composition of the source rock in the drainage, upstream of the sampling sites (Rose *et al*., 1979). Table 2 indicates the geochemical variation between the bedrock and stream sediments data and briefly summarised herein.

It is evident that the downstream sediments are rich in the following major oxides: the SiO_2 with a maximum concentration of 98.10%; TiO_2 (maximum 6.50 wt.%); MgO (maximum 17.00 wt.%); Fe_2O_3 (maximum 33.60 wt.%); MnO (maximum 0.60 wt.%); Cr_2O_3 (maximum 11.50 wt.%); low contents of Al_2O_3 (maximum 24.00 wt.%); Na_2O (maximum 2.70 wt.%); K_2O (maximum 4.20 wt.%); CaO (maximum 12.70 wt.%); and P_2O_5 (maximum 1.40 wt.%). The study reveals that the elemental distribution in stream sediments is mainly guided by the bedrock geology and geochemistry. The bedrock of the area is reflected remarkably well in the elevated values of SiO_2, TiO_2, Fe_2O_3, MnO, MgO and Cr_2O_3 with a decrease in CaO. The reason for the increase of SiO_2 in the stream sediments is that much of SiO_2 is present as quartz grains which is the most resistant mineral, most stable and has low solubility. The enrichment of TiO_2 from the bedrock into the sediment is determined by the abundance of detrital oxides and silicates, such as chlorite and clay minerals. The enrichment of Fe_2O_3 in the stream sediments is due to the presence of pyrite, siderite and hematite. This means that the minerals containing this oxide have undergone weathering processes which largely are dependent on pH-Eh. The main reason for the increase of MnO in the stream sediments could be due to its association to carbonates and clay minerals. With regards to the elevated values of Cr_2O_3 is associated with the ferrochrome processing plant not the bedrock.

There is decrease in concentrations of Al_2O_3, CaO, Na_2O, K_2O and P_2O_5 in the stream sediments which suggest dissolution of some minerals from the sediments. The depletion of these elements and the enrichment of SiO_2 may be due to the sedimentary sorting and loss of small grain-sized clays. CaO K_2O and Na_2O concentration are controlled by feldspars and their depletion is associated with physical weathering and dissolution which resulted into finer grain size of clastic particles. It also suggests the alteration of plagioclase due to chemical weathering into clay minerals. Low concentrations of CaO also indicate depletion in carbonate minerals in the stream.

The stream sediments have high trace element concentrations compared to the bedrocks of the Witbank Coalfield (Table 2). The trace elements enriched in the stream sediments are As, Ba, Bi, Br, Ce, Co, Cs, Hf, La, Mo, Nb, Nd, Ni, Pb, Rb, Se, Sm, Sr, Ta, Tl, Y, Yb, U, V, W, Zn and Zr with the depleted elements being Cu, Ga, Ge, Sc and Th. The sediments have high contents of arsenic (maximum 680 mg/kg), cobalt (maximum1273 mg/kg), molybdenum (maximum 16 mg/kg), nickel (maximum 197 mg/kg), lead (maximum 98 mg/kg), uranium (maximum 1898 mg/kg) and zinc (maximum 724 mg/kg), low contents of copper (maximum 89 mg/kg), gallium (maximum 34 mg/kg), germanium (maximum 12 mg/kg), scandium (maximum 31 mg/kg), and thorium (maximum 42 mg/kg).

Conclusions and Recommendations

The main reason for high concentrations of arsenic and other elements associated with in the stream sediments can be due to the present of sulphides and organic matter. The positive correlation of the As element with S and C confirms that As is mostly concentrated in the pyrite and organic matter. There is a possibility of anthropogenic source, especially

***Table 2** Univariate statistics (min, max mean, standard deviation and median) of major (wt.%) and trace (mg/kg) elements in the bedrock and stream sediments*

Elements	Bedrock					Stream Sediments				
	Min	Max	Stdev	Mean	Median	Min	Max	Stdev	Mean	Median
SiO_2	6,29	96,63	19,12	61,96	65,74	15,40	98,10	14,60	75,60	77,70
TiO_2	0,08	1,92	0,40	0,66	0,69	0,10	6,50	0,50	0,50	0,50
Al_2O_3	1,56	31,65	6,09	13,30	13,55	0,80	24,00	3,90	7,20	6,60
Fe_2O_3	0,62	15,77	3,00	4,02	3,40	0,60	33,60	4,70	5,70	4,70
MnO	0,00	0,22	0,05	0,05	0,03	0,00	0,60	0,10	0,10	0,10
MgO	0,01	8,61	1,58	1,04	0,74	0,00	17,00	1,40	0,60	0,20
CaO	0,02	27,95	5,20	2,53	0,55	0,00	12,70	1,90	1,10	0,30
Na_2O	0,01	2,96	0,66	0,79	0,63	0,00	2,70	0,40	0,30	0,10
K_2O	0,05	4,34	1,13	2,50	2,76	0,10	4,20	0,60	1,10	0,10
P_2O_5	0,01	1,69	0,26	0,13	0,07	0,00	1,40	0,10	0,10	0,10
Cr_2O_3	0,00	0,05	0,01	0,01	0,01	0,00	11,50	0,70	0,10	0,00
As	4,00	36,00	5,58	7,41	5,30	4,00	680,00	99,40	34,40	4,00
Ba	62,00	931,00	213,47	574,17	606,00	3,00	1128,00	208,50	363,10	378,00
Bi	3,00	5,20	0,38	3,12	3,00	2,00	36,00	3,30	3,60	3,00
Br	2,00	2,00	0,00	2,00	2,00	2,00	670,00	57,60	16,20	2,10
Ce	10,00	214,00	50,69	64,89	49,00	4,50	335,00	50,20	70,10	59,00
Co	1,00	39,00	7,23	9,87	8,40	1,60	1273,00	119,80	49,50	20,00
Cs	5,00	16,00	3,07	7,06	5,00	4,60	93,00	9,60	7,50	5,00
Cu	2,00	256,00	33,01	17,63	9,20	1,60	89,00	16,70	22,90	17,00
Ga	1,80	45,00	8,66	18,74	19,00	1,00	34,00	6,00	9,00	8,50
Ge	1,00	33,00	3,97	1,60	1,00	1,00	12,00	2,30	1,80	1,00
Hf	3,00	32,00	5,98	9,05	8,00	3,00	223,00	19,40	12,30	8,30
La	10,00	87,00	23,53	32,95	24,00	2,00	105,00	16,80	23,90	21,00
Mo	2,00	6,30	0,65	2,19	2,00	2,00	16,00	2,40	2,80	2,00
Nb	5,10	40,00	7,43	16,81	17,00	1,90	358,00	30,30	14,20	9,10
Nd	10,00	78,00	20,33	30,83	23,00	6,40	379,00	31,00	28,30	22,50
Ni	2,00	65,00	17,06	21,92	18,00	4,50	197,00	22,80	30,60	24,00
Pb	2,40	38,00	8,11	21,62	24,00	2,90	98,00	16,60	23,90	19,00
Rb	2,00	154,00	41,20	94,04	106,00	3,00	181,00	31,40	47,30	43,00
Sc	3,00	64,00	10,48	13,25	12,00	1,00	31,00	6,10	8,90	8,30
Se	1,00	4,40	0,46	1,13	1,00	1,00	72,00	6,60	2,70	1,00
Sm	10,00	16,00	1,37	10,51	10,00	7,40	194,00	20,10	14,90	10,00
Sr	8,50	302,00	63,24	127,67	123,00	2,00	458,00	56,70	54,20	44,00
Ta	2,00	3,80	0,32	2,11	2,00	2,00	19,00	2,10	2,70	2,00
Th	3,00	47,00	8,91	10,58	8,40	3,00	42,00	4,60	6,90	5,70
Tl	3,00	3,00	0,00	3,00	3,00	2,00	37,00	2,30	3,20	3,00
U	2,00	11,00	2,16	3,59	2,50	2,00	1898,00	177,60	39,70	2,40
V	6,40	489,00	64,69	72,12	63,00	3,00	1028,00	107,90	94,00	76,00
W	3,00	3,00	0,00	3,00	3,00	3,00	352,00	29,70	11,30	3,00
Y	3,10	61,00	14,77	25,64	25,00	2,90	211,00	17,80	17,40	15,00
Yb	3,00	7,10	1,02	3,63	3,00	3,00	4131,00	318,10	50,30	3,00
Zn	3,00	133,00	39,96	62,18	58,00	3,00	724,00	131,90	108,20	49,50
Zr	45,00	838,00	149,19	234,68	193,00	58,00	893,00	113,30	275,00	276,00

the Bushveld Complex materials which are rich in sulphides minerals. Enrichment of elements in the stream sediments suggests some loss of mobile components from the source rocks during weathering and in metal transportation process. The enrichment of these elements in the sediments could also be caused by the weathering of the bedrocks. The elements correlate positively with Al_2O_3, which is an indication of clay minerals and Ti-bearing minerals association. The enrichment can be attributed to the anthropogenic factor downstream of the source rock.

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