
A new look at sulphide mineralisation of the northern limb, Bushveld Complex: a stable isotope study

E. R. Sharman-Harris, J. A. Kinnaird, C. Harris and U. E. Horstmann

The Platreef is the main platinum-group element (PGE)-bearing horizon in the northern limb of the Bushveld Complex, South Africa. It is considered to be richer in sulphides than other similar horizons within the Bushveld Complex, in particular the Merensky Reef. Previous work has indicated that assimilation of dolomite may be a mechanism to add sulphur to the magma from which the Platreef formed. Sulphur isotope data presented in this study indicates an additional sulphur source contributing to the Platreef. In the southern Platreef, the Duitschland Formation of the Transvaal Supergroup forms part of the direct footwall. Within this sequence are pyrite-rich shales, which we suggest contributed to the sulphur budget of the Platreef. Local variations in sulphur isotopes, as well as a decrease in crustal sulphur in the Platreef further away from the footwall, also indicate local rather than regional contamination processes. Platreef samples have $\delta^{18}\text{O}$ values that are higher than expected in a mantle-derived magma, but there is no apparent systematic variation in $\delta^{18}\text{O}$ with distance from the footwall. This may possibly indicate contamination of the Bushveld magma pre-intrusion, probably in a

staging chamber. However, when compared to data from the central sector of the Platreef itself, analysis from this study have lower $\delta^{18}\text{O}$ values indicating changes in the degree of contamination with varying footwall lithology. Also, differences between plagioclase and pyroxene $\delta^{18}\text{O}$ values indicate exchange with fluids. It is possible that late-stage deuteric fluids may have caused this exchange.

E. R. Sharman-Harris (E-mail: sharmane@science.pg.wits.ac.za) and J. A. Kinnaird (for correspondence E-mail: kinnairdj@geosciences.wits.ac.za) are in the School of Geosciences, University of the Witwatersrand, Private Bag 3, Wits, 2050, South Africa; C. Harris (E-mail: charris@geology.uct.ac.za) is in the Department of Geological Sciences, University of Cape Town, Rondebosch 7700, South Africa; and U. E. Horstmann (E-mail: horstmann@src.wits.ac.za) is at iThemba Labs Gauteng, Private Bag 11, Wits, 2050, South Africa.

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INTRODUCTION

The Platreef is the main platinum-group element (PGE) bearing lithology of the northern limb of the Bushveld Complex. It has been the site of platinum prospecting and mining since the 1920s. The term Platreef as used in this paper refers to a series of pyroxenites and norites, containing xenoliths/rafts of footwall rock. The Platreef is irregularly mineralised with PGE, Cu and Ni, and a greater modal percentage of sulphides is present than in the Merensky Reef. There is an extremely varied PGE mineralogy, occurring mainly as tellurides, bismuthotellurides, antimonides and arsenides of Pd, Pt, and Rh.^{1,20} The main base-metal sulphides within the Platreef are pyrrhotite, pyrite, pentlandite, and chalcopyrite.

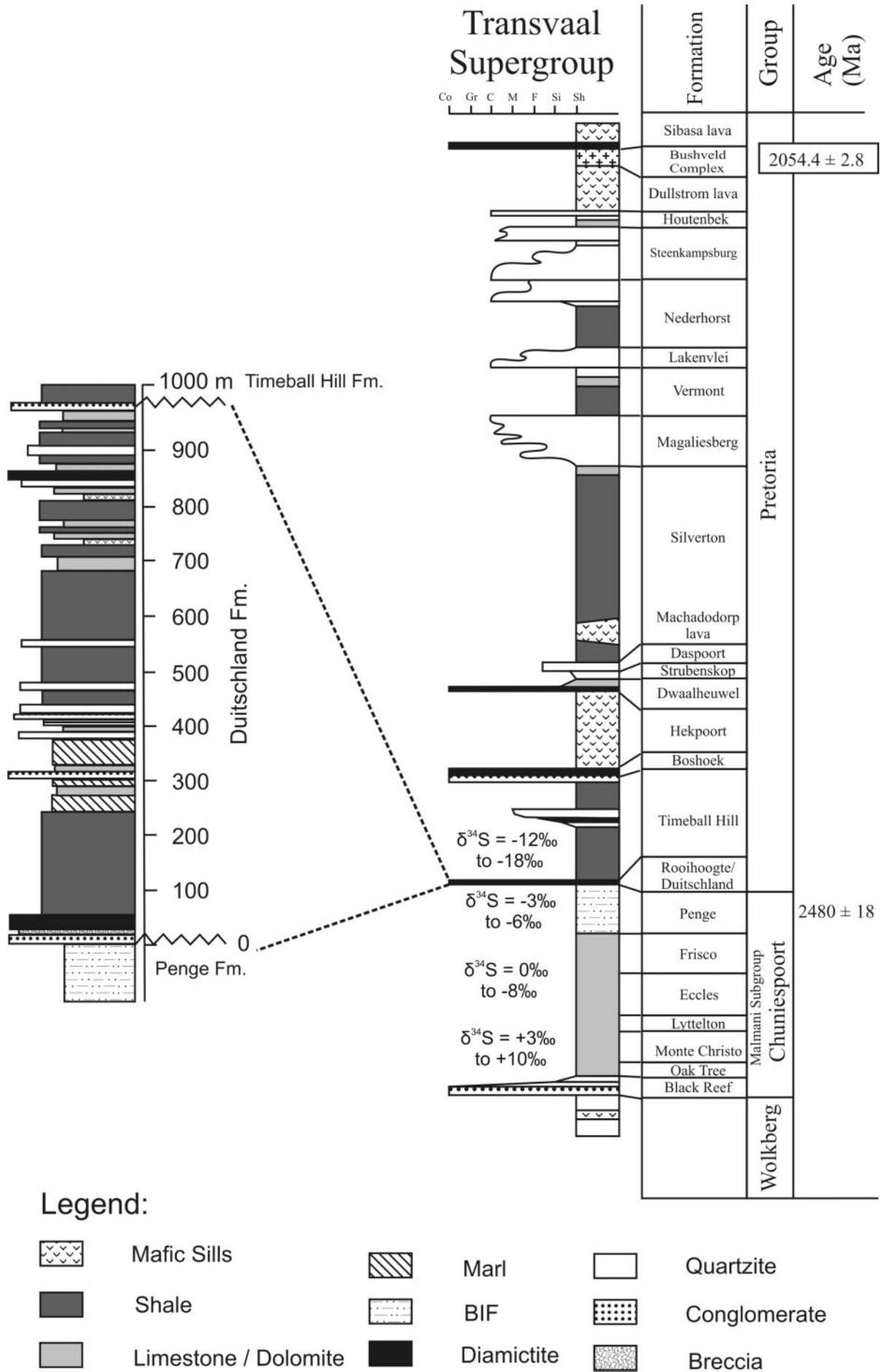
In this paper, we aim to examine sulphur and oxygen isotope data from the southern sector of the Platreef to gain a better understanding of the role that contamination played and in order to compare these data to those from earlier work conducted in the central and northern sectors of the Platreef. We conducted a detailed isotopic study through 400 m of the Platreef and also investigated isotopic variations in

different metasedimentary footwall lithologies of the Platreef itself as well as from unaltered sedimentary lithologies of the Duitschland Formation.

REGIONAL GEOGRAPHY

Transvaal Supergroup

The Palaeoproterozoic Transvaal Supergroup (Fig. 1) was deposited between 2.67 and 2.07 Ga^{8,10} in the Transvaal and the Griqualand West Basins which are separated by the Vryburg Rise.³ The Supergroup attains a maximum thickness of 12 km within the Transvaal Basin.¹⁰ The basement rocks of the Transvaal Supergroup are predominantly Archaean granites, gneisses and greenstones, as well as a Witwatersrand sedimentary succession and Ventersdorp lavas.^{8,10} The Transvaal Supergroup comprises the Wolkberg, Chuinespoort, and Pretoria Groups (Fig. 1); however, as there is an erosional contact between the Wolkberg and Chuinespoort Groups, Coetzee⁸ does not consider the Wolkberg group to be part of the Transvaal Basin *sensu stricto*.



1 Stratigraphic column of the Transvaal Supergroup, showing detail of the Duitschland Formation^{3,8}

In the southern sector of the Platreef, where this study is focused, the footwall is often comprised of Deutschland Formation which marks the base of the Pretoria Group and disconformably overlies the Chuniespoort Group. It comprises diamictite, an alternating sequence of fine-grained laminated shales; occasionally containing pyrite and dolomite, along with some minor sequences of quartzite and mudrock (Fig. 1). The formation is exposed in two areas near Mokopane and has a maximum thickness of 1000 m.²⁸ The Deutschland Formation is thought to have been deposited in a relatively shallow marine environment,^{3,28} which is supported by the presence of stromatolites. Carbonaceous shales represent the deepest depositional environment within the formation.²⁸ The base of the formation is marked by a diamictite, and the top of the formation is characterised by an ivory-white dolomite containing bornite and chalcopyrite. It is bounded at its base in the north by the Penge Iron Formation which is composed predominantly of quartz, magnetite and hematite⁸ and also contains subordinate carbonaceous shale beds. The top of the sequence in the study area is marked by Timeball Hill Formation which is composed of shale, quartzite, and diamictite.

Bushveld Complex

The Bushveld Complex (BC), which intruded into the Transvaal Supergroup, comprises the Rustenburg Layered Suite (RLS) – a thick sequence of layered mafic and ultramafic rocks ranging from dunite and pyroxenite, to anorthosite and pure oxide layers⁹ – as well as the Bushveld Granites. The Bushveld Complex covers an area of approximately 65 000 km², with a thickness of 7–9 km. The rocks of the RLS occur in extensive layers of varying thickness, and despite their age of 2.05 Ga¹⁴ have experienced no metamorphism, and only mild or local alteration.⁹ The RLS comprises five major limbs – the western limb, the far western limb, the northern limb, the eastern limb (all of which outcrop at surface) and the south-eastern or Bethal limb, which is covered by younger sediments. The western and eastern limbs are the best known, being the most extensive, with average lengths of ~200 km. The northern limb is partially concealed beneath younger rocks of the Waterberg Supergroup, the far western limb is an eroded remnant, and the Bethal limb is only identifiable through a gravity high, with its location confirmed by borehole data.⁹ The stratigraphy of the RLS is divided into five zones – the Marginal Zone, Lower Zone (LZ), Critical Zone (CZ), Main Zone (MZ), and Upper Zone (UZ).⁹

Northern limb

The northern limb occurs as a slightly sinuous, north-west striking sequence with a length of 110 km and a maximum width of 15 km,^{1,34} and differs in several ways from the eastern and western limbs. Previously, not all of the zones of the Bushveld Complex were thought to be represented in the northern limb, where the existence of units from the Critical Zone were yet to be identified north of Mokopane (formerly

Potgietersus) and units from the Lower Zone were thought to only be present south of Mokopane^{4,16} and as satellite bodies. Current work, however, indicates that a Lower Zone component comprising pyroxenites and harzburgites occurs at the base of the succession south of Farm Tweefontein, as well as in satellite bodies.²² Marginal Zone norite also occurs sporadically within the succession on farms Macalacaskop 243KR and Turfspruit 241KR. These sequences are followed by the Platreef itself, and it appears that the entire magmatic sequence has an overlapping relationship with the floor rocks, with each subsequent magmatic event being intruded over a wider area than the previous sequences.

The hanging-wall rocks of the Platreef are generally PGE-poor Main Zone comprising predominantly gabbro-norite and are up to 2000 m in thickness.³⁴ The thickness of the MZ in the northern limb is much less than in the eastern and western limbs of the Bushveld Complex. This Main Zone is followed by Upper Zone cyclic units of magnetite, magnetite gabbro, gabbro and anorthosite, which are ~1500 m thick.² In the west, the hanging wall is a variety of Bushveld granites and metasedimentary rocks.²²

An important feature of the northern limb is that the layered rocks have a transgressive relationship with the country rocks of the Transvaal Supergroup (*i.e.* the intrusion rests on progressively older rocks towards the north). To the south of the town of Mokopane, the floor rocks comprise Magaliesburg quartzites. Northwards, the floor rocks first comprise the Pretoria Group, the banded iron-formations and dolomites of the Chuniespoort Group, and finally the Archaean granites.¹⁶ The entire layered sequence pinches out to the north.^{4,16,34}

General geology of the Platreef

The Platreef, which is the major PGE-bearing layer of the northern limb, occurs at the base of the layered rocks. The Platreef comprises a 'complex series of medium-to-coarse-grained pyroxenites and norites that contains xenoliths of the floor rocks'.¹² It is observed as being a highly inhomogeneous body, comprising different rock types including pyroxenites, para-pyroxenites, feldspathic pyroxenites (pyroxenites with > 10% interstitial plagioclase), serpentinites, gabbro-norites and norites. The Platreef itself has been equated with the Critical Zone and, specifically, the Merensky Reef, of the Bushveld Complex proper,^{35,36} although other authors have regarded the Platreef as the base of the Main Zone.³³

The Platreef also contains xenoliths of altered footwall, which vary depending on the footwall; however, the composition of these rafts is not always the same as the directly underlying footwall. For example, the hornfels rafts within the Platreef; that have a Deutschland Formation footwall of dolomitic marble; have a mineral composition of aluminous cordierite-spinel hornfels. These rafts comprise mainly cordierite, with disseminated green spinel, and accessory corundum, sillimanite and andalusite, that indicates a high-grade of metamorphism of a shale

protolith. Another example is the occurrence of calc–silicate rafts in the northern sector of the Platreef on the Farm Drenthe, where the direct footwall is Archaean granite.^{12,22}

The base- and precious-metal mineralisation of the Platreef is unevenly distributed, and occurs over a zone that is up to 400 m thick.²² The sulphides in the Platreef usually occur as a combination of pyrrhotite, pentlandite, chalcopyrite and, occasionally, pyrite and bornite, which exist as several different styles or phases of mineralisation. Massive sulphides are recorded in core up to several metres in thickness, and often contain a complex assemblage of sulphide minerals. Typically, these massive sulphides are dominated by pyrrhotite. Chalcopyrite usually occurs around the rims of the pyrrhotite, and has pentlandite associated with it. Pentlandite also occurs as veins, and as exsolution lamellae within the pyrrhotite. More rarely, these massive sulphides are dominated by chalcopyrite, which have pyrrhotite veins running through them. Associated with massive sulphides, and often within serpentinites are ‘net-textured’ sulphides that occur as a matrix to the silicate minerals, with samples having 50–80 modal percent sulphides. ‘Net-textured’ sulphides are dominated by pyrrhotite, with minor chalcopyrite and pentlandite.

Fine-to-medium-grained disseminated sulphides are common in the finer grained lithologies of the Platreef, and are often interstitial. These sulphides have a different modal mineralogy than that of the massive or ‘net-textured’ sulphides. They usually comprise approximately 50% pentlandite, 40% chalcopyrite, as well as pyrrhotite and occasional bornite. ‘Blebbly’ sulphides also occur, and contain a similar combination of sulphide minerals to that of the interstitial sulphides. These blebbly sulphides are often observed as pyrrhotite with chalcopyrite rims.

Sulphides also occur within the metamorphosed footwall of the Platreef, particularly within calc–silicates, but also occasionally within cordierite spinel hornfels. Typically, these sulphides are cubic pyrite, but pyrrhotite has also been observed, sometimes occurring in a cubic form. This is probably due to the conversion of pyrite to pyrrhotite during the metamorphism of the footwall. It should also be noted that mineralisation occurs around calc–silicate rafts within the Main Zone of the northern limb, especially in the northern sector of the Platreef, and shows similarities to that in the Platreef (*sensu stricto*).^{12,22}

The sulphide mineralisation was thought to be of primarily magmatic origin by some authors;³³ however, evidence for post-magmatic fluid interaction¹⁶ is also present. Buchanan *et al.*,⁴ Harris and Chaumba,¹⁶ and Armitage *et al.*¹ suggested that assimilation, or contamination, by sedimentary floor rocks, especially those rich in sulphur, may have contributed to the sulphide mineralisation of the Platreef. Petrographic studies indicate an association between sulphides and biotite, as well as the alteration of silicates in samples with high sulphide content. The degree of alteration associated with sulphides, as well as the presence of biotite, is probably due to an

interaction between pre-existing silicate minerals, and an immiscible sulphide liquid or later fluid-rich phase.

It has previously been assumed that the Pt and Pd mineralisation within the Platreef was associated with sulphide mineralisation. Armitage *et al.*¹ noted that the occurrence of PGE mineralisation associated with alteration zones indicates ‘syn- to post-magmatic crystallization or redistribution of PGE by hydrothermal fluids’. The authors also noted that the PGE mineralisation in the Platreef in the central sector is more complex than previously thought, and that several different mineralisation processes have been involved in its formation. A new detailed study using laser ablation-inductively coupled plasma–mass spectrometry has revealed that, while Pd, Ru, Rh and some Os and Ir are commonly hosted by sulphides, Pt generally is not.²¹ In addition, a number of Pt-bearing minerals, including geversite (PtSb₂) and sperrylite (PtAs₂) and various Pt–Pd–Bi–Te-antimonide, Pd–Pt–Sb–Bi-telluride and Pd–Pt–Bi-telluride phases have been seen to occur in the silicates external to the sulphides. This may be linked to contamination. Hutchinson and McDonald²¹ noted that where the degree of contamination was high, the Pt formed As and Sb complexes, which were subsequently expelled from the sulphide liquids. As a result of this, Pt was fractionated from Pd and the other platinum-group elements which remained within the sulphide minerals.

Previous stable isotope studies

The first sulphur isotope analyses conducted on the northern limb were by Liebenberg.²⁴ He recorded two $\delta^{34}\text{S}$ values for the farm Zwartfontein 818LR, both of which had approximate magmatic values of +0.7‰ (for a calc–silicate) and +1.9‰ (from a pegmatoidal norite; Fig. 3). Hulbert¹⁹ presented data from Lower Zone pyroxenites and chromitites south of Mokopane, as well as pyroxenites, chromitites and anorthosites from the Critical Zone. These yielded $\delta^{34}\text{S}$ values from +0.96‰ to +7.54‰ (Fig. 3). He also analysed two samples of Upper Zone material from the northern sector of the Platreef on farm Molendraai, which also had $\delta^{34}\text{S}$ values of approximately +2‰ (Fig. 3). Hulbert interpreted these data as indicating that the majority of sulphur associated with sulphides is of mantle origin. Elevated $\delta^{34}\text{S}$ values were ascribed to an increased f_{O_2} which the author proposed was due to the release of volatiles from sedimentary material.¹⁹

The majority of the sulphur isotope data available for the Platreef itself prior to this work was presented by Buchanan *et al.*⁴ and Buchanan and Rouse⁵ and was for a variety of sulphide samples taken from Tweefontein and Turfspruit farms (Fig. 3). These samples indicated $\delta^{34}\text{S}$ values in pyrrhotite of +6‰ to +9‰, which they suggested might have originated from contamination by anhydrite within the Malmani Dolomite.⁴ Sulphides from graphite layers on Farm Turfspruit yielded values of –2.42‰ to +2.0‰.⁵

Manyeruke and Maier²⁷ examined samples taken from a core on farm Townlands (Fig. 3), and identified three different units separated by hornfels

rafts, which they termed the Upper, Middle and Lower Platreef. Sulphur isotope values from this core indicated that the Upper and Lower Platreefs had elevated $\delta^{34}\text{S}$ values of about +8‰, whereas the Middle Platreef had $\delta^{34}\text{S}$ values of about +4‰. These elevated values were also attributed to the addition of sulphur of crustal origin.

Cameron⁶ investigated sulphur isotope variations for the Transvaal Supergroup, specifically for the Timeball Hill shale, Penge Iron Formation, the Malmani Subgroup, and the Black Reef Quartzite in the eastern Bushveld. It is recorded that the majority of these lithologies have significantly negative $\delta^{34}\text{S}$ values. The Timeball Hill Formation yielded $\delta^{34}\text{S}$ values of -12‰ to -18‰, and the Penge Formation has $\delta^{34}\text{S}$ values of -3‰ to -6‰ (Fig. 1). Pyrite-bearing shales from the Malmani Subgroup showed some variations, with the upper three-quarters yielding $\delta^{34}\text{S}$ values of 0‰ to -8‰, and the lower quarter yielding values of +3‰ to +10‰. Pyrite in quartzites of the Black Reef Formation gave $\delta^{34}\text{S}$ values of about +3‰ (Fig. 1).

Li *et al.*²³ examined sulphur isotope variations for the Uitkomst Complex, which is considered to be a satellite body of the eastern Bushveld Complex. In the unmineralised lithologies of the complex, as well as the basal gabbro, $\delta^{34}\text{S}$ values are recorded as being close to magmatic (-0.9‰ to +2.6‰). However, in the sulphide-bearing harzburgites, the $\delta^{34}\text{S}$ values indicated a contribution from crustal sulphur (-2.6‰ to -7.1‰), which the authors proposed was probably from either the Malmani Subgroup or the Timeball Hill Formation.

Oxygen isotope work has been conducted both on the northern limb,^{16,17} and on the eastern and western limbs of the Bushveld.^{30,31} Schiffries and Rye³¹ recorded $\delta^{18}\text{O}$ values from mineral separates within the RLS at approximately 1‰ higher than values expected for a basaltic magma. No systematic variation with stratigraphic height was observed. This was suggested to indicate RLS magmas were contaminated and were well mixed before emplacement. The slightly elevated $\delta^{18}\text{O}$ values could be attributed to the assimilation of Transvaal Supergroup rocks into the magma, *i.e.* up to 10–29% contamination.

This observation was supported by Harris *et al.*¹⁷ who also found slight elevated $\delta^{18}\text{O}$ values, and no stratigraphic variation. This is thought to indicate that BC magmas were contaminated before emplacement, supporting the staging chamber hypothesis. However, the most likely contaminant is suggested to be the middle-to-lower crust, and not Transvaal rocks as previously proposed. This is also supported by a Sr–Nd isotope study conducted by Maier *et al.*²⁵

Reid *et al.*³⁰ examined samples taken the footwall of the Merensky Reef as it occurs in a pothole formed in the Merensky Reef of the western limb of the Bushveld Complex. They found that their $\delta^{18}\text{O}$ data agreed well with that of Schiffries and Rye,³¹ with $\delta^{18}\text{O}_{\text{plag}}$ values of approximately +7.1‰, and $\delta^{18}\text{O}_{\text{opx}}$ values of approximately +6.6‰. No variation with depth was observed by the authors. They also

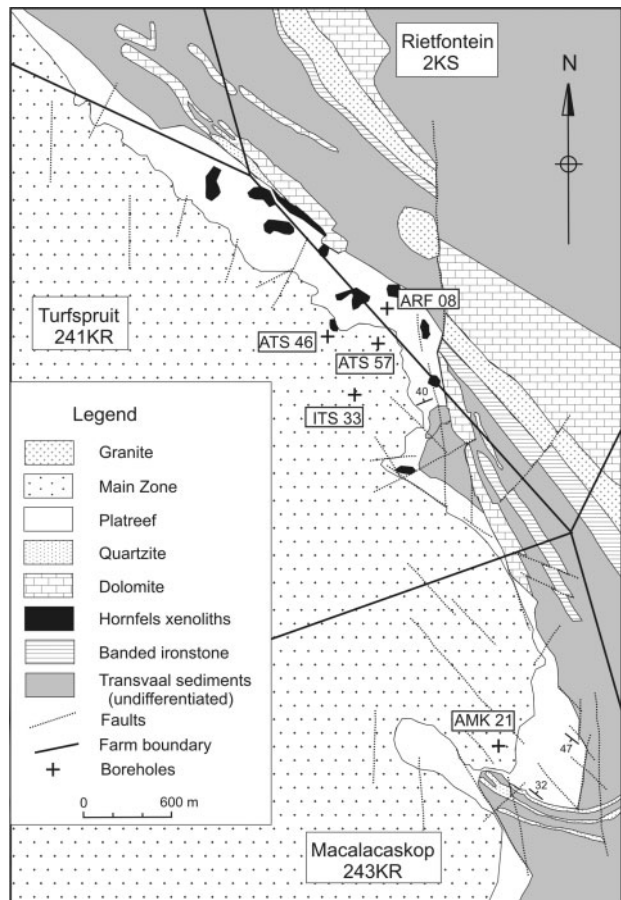
recorded a $\Delta_{\text{plagioclase-pyroxene}} = 0.5\text{‰}$, indicating that magmatic processes were responsible for silicate formation. This is also in agreement with Schiffries and Rye.³¹

Harris and Chaumba¹⁷ conducted stable isotope studies on plagioclase and pyroxenes taken from feldspathic pyroxenites of the Platreef at Sandsloot Mine in the northern limb. These samples (which were genuine magmatic feldspathic pyroxenites) indicated assimilation of Malmani Dolomite footwall.

METHODOLOGY

Sample selection

Sulphides for this study were collected through a vertical Platreef profile of 450 m and from sulphide-bearing footwall lithologies. Samples were chosen from selected cores on Farms Macalacaskop (AMK-21), Turfspruit (ATS-46, ATS-57, ITS-33) and Rietfontein (ARF-08) (Fig. 2). The cores selected had varying footwall lithologies including pyritic shales, sulphide-bearing dolomite of the Duitschland Formation and anhydrite-bearing dolomite of the Malmani Subgroup. Sulphides from the Platreef were collected systematically from top to bottom and included disseminated, blebby, net-textured and massive ore together with sulphides from hornfels xenoliths. Chosen samples were either crushed or



2 Geological map of the northern limb of the Bushveld Complex showing the location of boreholes used in this study

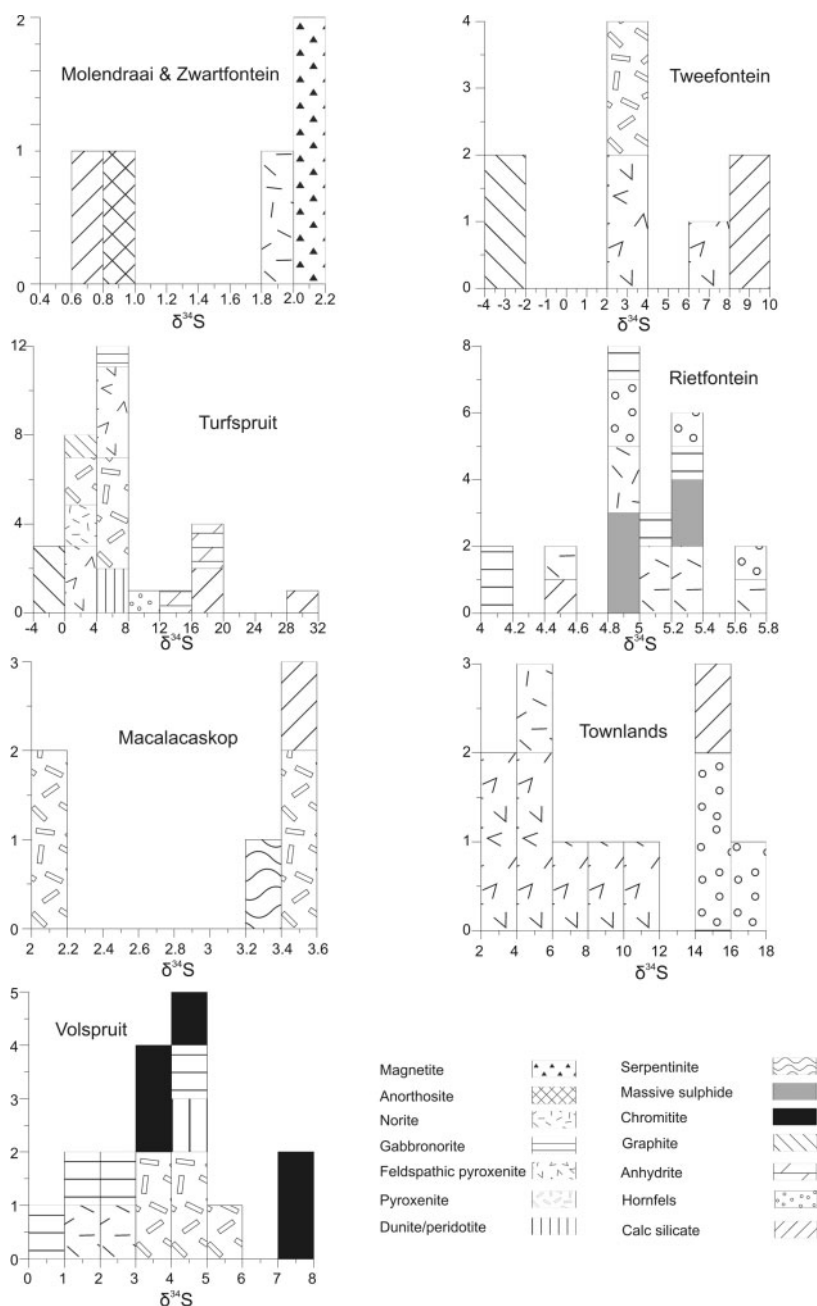
prepared into polished blocks. An oxygen isotope study was also conducted to compare any effects of crustal contamination on host rock silicates with those of sulphide minerals.

Sample preparation

Samples that were crushed were then milled, and sieved to $-125+90\ \mu\text{m}$ in size. These fractions were then washed and dried to remove any very fine material. Samples were also cleaned of any heavily magnetic minerals (*i.e.* magnetite or pyrrhotite) using a hand magnet, then separated according to magnetic susceptibility on a Frantz Isodynamic Separator using various amperages. Separates were then further cleaned to remove any remaining silicates using heavy liquid separation (bromoform) resulting in pure

sulphide fractions. For samples where it was not possible to separate out pure sulphides, mineral separates were extracted by microdrill from polished blocks at University College London. This drill allows for the extraction of mineral separates on a small scale as its tip is approximately $50\ \mu\text{m}$ in diameter.

Sulphur isotope analyses were conducted on pyrrhotite, pentlandite and chalcopyrite mineral separates from the Platreef, pyrrhotite and pyrite from hornfels within cores, as well as on pyrite and bornite from footwall material of the Deutschland Formation. Anhydrite samples from a dolomite, which occurs in the Central Sector of the Platreef, were also analysed for comparative purposes. Sulphur isotope analyses were conducted at the Environmental Isotope Laboratory of the iThemba Laboratories in South

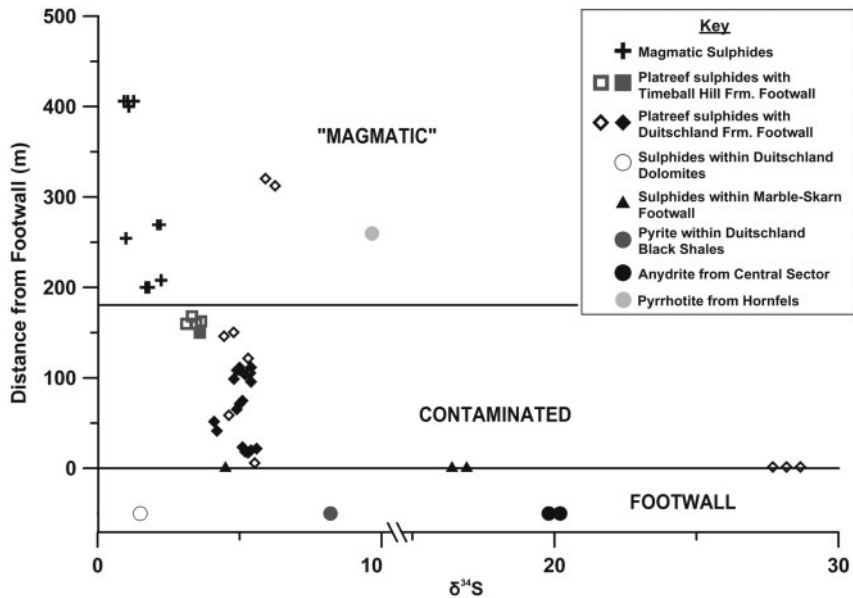


3 Histograms showing all of the sulphur isotope analyses for the northern limb of the Bushveld Complex. Data for Zwartfontein from Liebenberg;²⁴ data for Macalacaskop, Turfspruit and Rietfontein from this study; data for Tweefontein from Buchanan *et al.*⁴ and Buchanan and Rouse;⁵ data for Townlands from Manyeruke;²⁶ and data for Molendraai and Volspruit from Hulbert¹⁹

Table 1 Sulphur isotope data in per mil relative to CDT

Sample number	Rock type	Mineral	Sulphide/sulphate texture	$\delta^{34}\text{S}\text{‰ CDT}$
ARF 08/36.42	Cordierite-Spinel Hornfels	Pyrrhotite	Fine-grained disseminated	5.3
ARF 08/36.42	Cordierite-Spinel Hornfels	Chalcopyrite	Fine-grained disseminated	5.6
ARF 08/36.53	Cordierite-Spinel Hornfels	Pyrrhotite	Fine-grained disseminated	5.0
ARF 08/44.65	Norite	Pyrrhotite	Medium-grained disseminated	5.2
ARF 08/49.26	Norite	Pyrrhotite	Fine-grained disseminated	4.5
ARF 08/49.26	Norite	Chalcopyrite	Fine-grained disseminated	4.8
ARF 08/49.26	Norite	Pyrrhotite	Fine-grained disseminated	5.2
ARF 08/52.42	Gabbro	Pyrrhotite	Fine-grained disseminated	5.4
ARF 08/73.24	Gabbro	Pyrrhotite	Fine-grained disseminated	5.1
ARF 08/76.80	Cordierite-Spinel Hornfels	Pyrrhotite	Fine-to-coarse grained blebs	5.0
ARF 08/82.74	Gabbro	Pyrrhotite	Medium-grained disseminated	4.9
ARF 08/96.32	Gabbro	Pyrrhotite	Medium-grained disseminated	4.1
ARF 08/106.67	Gabbro	Pyrrhotite	Coarse-grained blebs	4.2
ARF 08/124.64	Massive Sulphide	Pyrrhotite	Massive sulphide	4.9
ARF 08/124.64	Massive Sulphide	Pyrrhotite	Massive sulphide	4.9
ARF 08/124.64	Massive Sulphide	Pyrrhotite	Massive sulphide	4.9
ARF 08/124.64	Massive Sulphide	Pyrrhotite	Massive sulphide	5.4
ARF 08/124.64	Massive Sulphide	Pyrrhotite	Massive sulphide	5.4
ARF 08/126.27	Norite	Pyrrhotite	Fine-to-coarse grained interstitial	5.6
ARF 08/129.78	Norite	Pyrrhotite	Medium-to-coarse grained blebs	5.0
ARF 08/129.78	Norite	Chalcopyrite	Medium-to-coarse grained blebs	5.4
ARF 08/130.89	Norite	Pyrrhotite	Very coarse grained bleb	5.3
ARF 08/153.46	Calc-Silicate	Pyrrhotite	Fine-grained disseminated	4.5
ATS 46/60.34	Pegmatoidal Norite	Pyrrhotite	Coarse-grained bleb	1.1
ATS 46/60.34	Pegmatoidal Norite	Chalcopyrite	Coarse-grained bleb	1.1
ATS 46/61.12	Feldspathic Pyroxenite	Pyrrhotite	Coarse-grained bleb	1.1
ATS 46/61.12	Feldspathic Pyroxenite	Chalcopyrite	Coarse-grained bleb	1.3
ATS 46/61.12	Feldspathic Pyroxenite	Pentlandite	Coarse-grained bleb	0.9
ATS 46/146.71	Feldspathic Pyroxenite	Pyrrhotite	Fine-to-medium grained disseminated	5.9
ATS 46/154.67	Gabbro Norite	Pyrrhotite	Net-textured vein	6.3
ATS 46/207.50	Hornfels	Pyrrhotite	Medium-grained bleb	9.7
ATS 46/316.62	Peridotite	Pyrrhotite	Medium-to-coarse grained interstitial	4.8
ATS 46/345.60	Dunite/Peridotite	Pyrrhotite	Net-textured	5.3
ATS 46/457.49	Pyroxenite	Pyrrhotite	Fine-grained interstitial	5.4
ATS 46/461.21	Feldspathic Pyroxenite	Chalcopyrite	Medium-grained interstitial	5.5
ATS 46/469.25	Calc-silicate	Pyrrhotite	Fine-grained disseminated	16.4
ATS 57/72.48	Pyroxenite	Chalcopyrite	Coarse-grained bleb	1.0
ATS 57/118.78	Pyroxenite	Pyrrhotite	Fine-to-coarse grained interstitial	2.2
ATS 57/180.64	Feldspathic Pyroxenite	Pyrrhotite	Fine-to-medium grained disseminated	4.5
ATS 57/218.39	Pyroxenite	Pyrrhotite	Net-textured	5.0
ATS 57/218.39	Pyroxenite	Chalcopyrite	Net-textured	4.9
ATS 57/221.39	Massive Sulphide/Pyroxenite	Pyrrhotite	Massive sulphides	5.3
ATS 57/221.39	Massive Sulphide/Pyroxenite	Pentlandite	Massive sulphides	5.4
ATS 57/268.15	Feldspathic Pyroxenite	Pyrrhotite	Fine-grained disseminated	4.6
ATS 57/325.32	Calc-silicate	Pyrrhotite	Coarse-grained bleb	28.2
ATS 57/325.32	Calc-silicate	Pyrrhotite	Coarse-grained bleb	27.7
ATS 57/325.32	Calc-silicate	Pyrrhotite	Coarse-grained bleb	28.7
ATS 57/335.07	Calc-silicate	Pyrite	Fine-grained disseminated	16.9
AMK 21/125.76	Pyroxenite	Pyrrhotite	Coarse-grained bleb	2.2
AMK 21/125.76	Pyroxenite	Pyrrhotite	Very coarse grained bleb	2.1
AMK 21/227.15	Serpentinite	Pyrrhotite	Interstitial	3.3
AMK 21/232.62	Pyroxenite	Pyrrhotite	Interstitial	3.6
AMK 21/232.62	Pyroxenite	Chalcopyrite	Interstitial	3.6
AMK 21/235.31	Calc-silicate	Pyrrhotite	Vein within chalcopyrite bleb	3.5
ITS 33/241.16	Feldspathic Pyroxenite	Pyrrhotite	Coarse grained interstitial	1.8
ITS 33/241.16	Feldspathic Pyroxenite	Chalcopyrite	Coarse grained interstitial	1.7
DL1	Dolomite	Bornite	Alteration vein	1.5
DL2	Shale	Pyrite	Medium-grained nodules along bedding planes	8.2
TN75	Anhydrite	Anhydrite	Vein	20.2
VK75-305	Anhydrite	Anhydrite	Vein	19.8

Sample prefixes ARF, ATS and AMK refer to Rietfontein, Turfspruit and Macalacaskop, respectively.

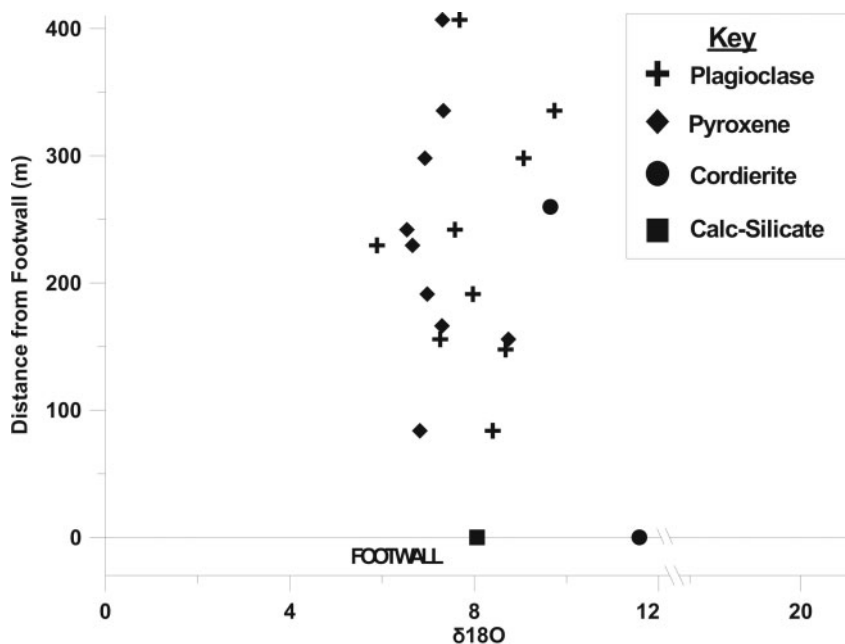


4 Diagram showing variation of $\delta^{34}\text{S}$ values with depth and increasing distance from the footwall, as well as $\delta^{34}\text{S}$ values from various sulphur-bearing footwalls of the northern limb

Africa, and microdrilled samples at Royal Holloway, University of London in the UK. Samples for sulphur isotope analysis in South Africa were prepared using the procedure outlined by Horstmann and Malinga¹⁸ and analysed using a continuous-flow isotope ratio mass-spectrometer (GEO 20-20). Sulphur isotope analyses conducted in the UK were done using procedures outlined in Grassineau *et al.*¹³ All sulphur isotope data are reported relative to Cañon Diablo Troilite (CDT).

Oxygen isotope analyses were undertaken on plagioclase and orthopyroxene separates from various lithologies of the Platreef, as well as on cordierite from footwall xenoliths within the Platreef. These minerals

were hand-picked under a binocular microscope using samples that had been crushed and sieved to $-250+125\ \mu\text{m}$. These silicate separates were crushed to a fine powder, and placed in an oven at approximately 50°C to dry. Once dry, the sample was weighed out ($\sim 10\ \text{mg}$) and placed in a nickel reaction vessel (inside diameter $\sim 12\ \text{mm}$) which was over-pressurised above atmosphere with dry nitrogen.³⁴ The extraction line consisted of ten reaction vessels, of which eight were used for samples, and two for standards.¹⁵ The analysis thereafter follows the procedure outlined in Harris *et al.*¹⁷ The analytical precision is estimated to be ± 0.1 per million based on the long-term average difference between duplicates of the MQ quartz standard.¹⁷



5 Diagram showing variation of $\delta^{18}\text{O}$ with depth and distance from footwall, as well as $\delta^{18}\text{O}$ values for footwall material that was analysed

Table 2 Details of all oxygen isotope analyses for this study (‰, V-SMOW), as well as the difference between $\delta^{18}\text{O}_{\text{plag}}$ and $\delta^{18}\text{O}_{\text{opx}}$ analyses ($\Delta_{\text{plag-opx}}$)

Sample number	Rock type	$\delta^{18}\text{O}_{\text{plag}}$	$\delta^{18}\text{O}_{\text{opx}}$	$\delta^{18}\text{O}_{\text{cord}}$	$\delta^{18}\text{O}_{\text{calc-sil}}$	$\Delta_{\text{plag-opx}}$
AMK021/90.25	Spotted Anorthosite	9.1	6.9			2.2
AMK021/222.06	Serpentinite		7.3			
AMK021/232.62	Pyroxenite	7.3	8.7			-1.4
AMK021/240.62	Pyroxenite	8.7				
AMK021/393.33	Metamorphosed quartzite			11.6		
ATS046/60.19	Norite–Leuconorite	7.7	7.3			0.4
ATS046/131.64	Feldspathic Pyroxenite	9.7	7.3			2.4
ATS046/207.25	Hornfels			9.7		
ATS046/275.80	Olivine. Feldspathic Pyroxenite	8.0	7.0			1.0
ATS046/383.23	Feldspathic Pyroxenite–Pyroxenite	8.4	6.8			1.6
ATS046/461.13	Feldspathic Pyroxenite					
ATS046/496.25	Calc-silicate				8.1	
ITS033/241.16	Feldspathic Pyroxenite	7.6	6.5			1.1
ITS033/253.44	Pyroxenite	5.9	6.7			-0.8

Abbreviations: plag, plagioclase; opx, orthopyroxene; cord, cordierite; calc-sil, calc-silicate.

RESULTS

Sulphur isotopes

Data obtained from Platreef sulphides varied according to the type of footwall lithology. On the Turfspruit farm, where the footwall is Duitschland Formation, $\delta^{34}\text{S}$ values can be separated into two distinct groups, one with an average of +5.2‰, the other with an average of +1.4‰ (Table 1; Fig. 3). These two groups occur at different depths in the sampled cores, with higher values occurring towards the base (Fig. 4). On the Rietfontein farm, where the footwall is also Duitschland Formation, $\delta^{34}\text{S}$ values all grouped together, with an average of +5.0‰ (Table 1; Fig. 3). Hornfels xenoliths from the two farms also split into two groups, with hornfels from the Rietfontein farm having an average $\delta^{34}\text{S}$ of +5.2‰, and a hornfels from the Turfspruit farm yielding a $\delta^{34}\text{S}$ value of +9.7‰ (Table 1; Fig. 3). Calc-silicates from the two farms generally showed two different sets of results, with pyrite and pyrrhotite within calc-silicates from farm Turfspruit having average $\delta^{34}\text{S}$ values of +16.6‰, and a pyrrhotite from farm Rietfontein, yielding an average $\delta^{34}\text{S}$ of +4.5‰. Analysis of a coarse-grained sulphide bleb which occurs within a calc-silicate at the base of core ATS 57 yielded anomalously high $\delta^{34}\text{S}$ values of +27.69‰ to +28.17‰.

Sulphides from magmatic Platreef rocks sampled from farm Macalacaskop, which has a footwall of Timeball Hill Formation quartzite, showed slightly less variation. Analyses yielded an average $\delta^{34}\text{S}$ value of +2.9‰ (Table 1), although a slight decrease in sulphur isotope values (from +3.6‰ to +2.2‰) is seen from the bottom to the top of the sampled core. A serpentinite from the same core yielded a $\delta^{34}\text{S}$ value of +3.3‰ and a pyrrhotite from the calc-silicate footwall has a $\delta^{34}\text{S}$ value of +3.6‰ (Table 1).

Footwall sedimentary sulphides taken from Farm Duitschland gave varying sulphur isotope values. Bornite from dolomite at the top of the Duitschland sequence yielded $\delta^{34}\text{S}$ values of +1.5‰, pyrite from the

shales at the base of the sequence gave $\delta^{34}\text{S}$ values of +8.2‰, and anhydrite gave $\delta^{34}\text{S}$ values of between +19.8‰ and +20.2‰.

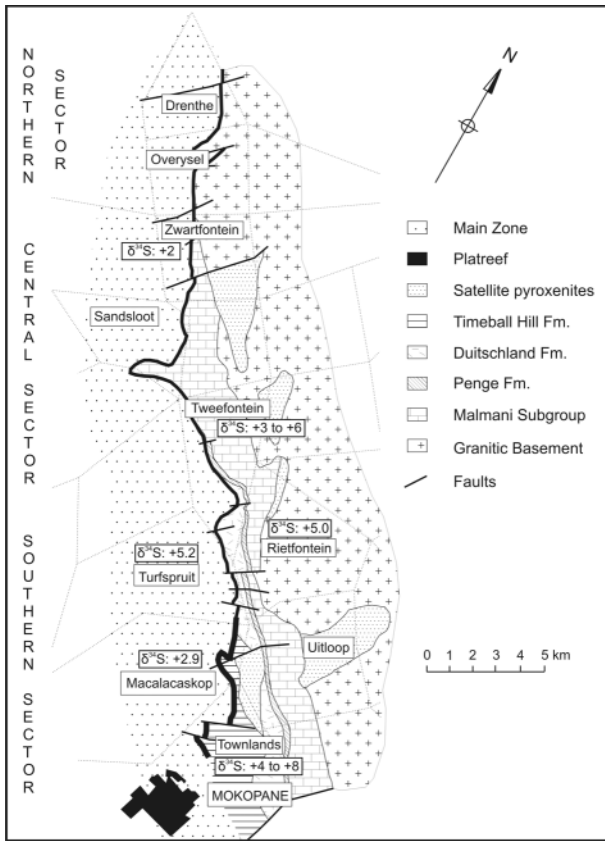
Oxygen isotopes

In the Platreef, some small variations of oxygen isotope data occur according to height above the footwall (Fig. 5). Recorded $\delta^{18}\text{O}$ values varying from +6‰ to +9‰ for plagioclase, and +6.5‰ to +8.5‰ for pyroxene (Table 2). Cordierite from hornfels rafts showed slightly higher $\delta^{18}\text{O}$ values with a maximum of +11.5‰ (AMK021/393.33, ATS046/207.25), whereas a calc-silicate from the direct footwall (ATS046/496.25) had a similar $\delta^{18}\text{O}$ as that of the Platreef itself (+8‰). The $\Delta_{\text{plag-opx}}$ values recorded by this study show a large variation from -1.48 to +2.41 (Table 2).

DISCUSSION AND CONCLUSIONS

Sulphur isotope analyses of sulphides from eclogites and associated kimberlites on the Kaapvaal Craton yield $\delta^{34}\text{S}$ values of +0.2‰ to +2.1‰.³³ This provides a viable range of $\delta^{34}\text{S}$ values for magmatic sulphides as they occur beneath Kaapvaal Craton. In this study, sulphides with values within this range were obtained from the top of cores from farms Macalacaskop and Turfspruit. Therefore, the group of $\delta^{34}\text{S}$ values that have an average of +1.4‰ can be said to be of magmatic origin and hence help to constrain a magmatic end member for the Platreef.

With increasing depth in the Platreef towards the footwall contact, these values increase from +1.4‰ to +5.2‰, which is seen clearly in Figure 4 where data are plotted against height above footwall contact. The only elevated $\delta^{34}\text{S}$ values more than 175 m above the footwall contact are related to a hornfels raft. This increase in $\delta^{34}\text{S}$ values towards the footwall contact seems likely to reflect an increasing contribution from crustal sulphur close to the floor. In the southern sector, the most likely source of crustal sulphur is from



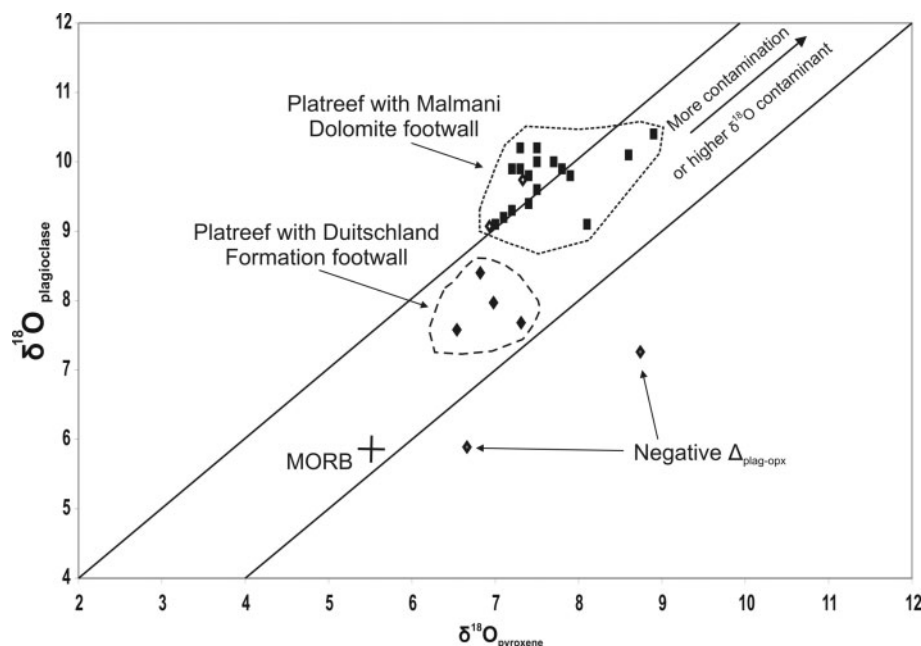
6 Diagram showing the geology of the Platreef with sulphur isotope data of the Platreef and equivalent lithologies. Map from P. Nex; data for Townlands from Manyeruke and Maier;²⁷ data for Macalacaskop, Turfspruit and Rietfontein from this study; data for Tweefontein from Buchanan *et al.*⁴ and Buchanan and Rouse;⁵ data for Zwartfontein from Liebenberg²⁴

occurred through the release of sulphur- and other volatile- and trace element-enriched fluids from the pyrite-rich shale during metamorphism to aluminous cordierite spinel hornfels which occur as rafts in the southern sector of the Platreef. The whole-rock geochemistry of these rafts is similar to that of the shale in the Deutschland Formation.

When combined with data from previous work,^{4,5,19,24,26} the data from this study indicates a variation of contaminated $\delta^{34}\text{S}$ values for the Platreef along strike (Fig. 6). When there is a quartzite footwall, sulphur isotope values are approximately +3‰, whereas with a footwall of Deutschland formation $\delta^{34}\text{S}$ values are approximately +5‰. In the central sector where the footwall is Malmani dolomites, sulphides have sulphur isotope values of +3‰ to +6‰, whereas on Archaean granites in the northern sector $\delta^{34}\text{S}$ values are +2‰. We propose that during metamorphism of footwall, volatiles, (including sulphur) are released into the magma. These volatiles travel through the system, causing sulphur saturation and the formation of an immiscible sulphide liquid which re-crystallises any pre-existing magmatic sulphides. The magmatic sulphides which occur at the top of the core have not been affected by these volatiles, and hence retain their primary sulphur isotope signature. Thus, whereas Buchanan *et al.*⁴ and Buchanan and Rouse⁵ proposed contamination of the Platreef by anhydrite, this study has shown that pyritic shales as well as anhydrite-bearing dolomites have contributed crustal sulphur to the Platreef magma.

Deutschland Formation footwall, as $\delta^{34}\text{S}$ values for other lithologies within the Transvaal Supergroup are generally negative (Fig. 1). This contamination probably

The $\delta^{18}\text{O}$ data from this study agree in general with previous studies.^{16,17,30,31} These datasets indicate the presence of crustal oxygen within the northern limb, and little variation with depth through the entire sequence, especially in contrast to the sulphur isotope data. This is to be expected, however, as oxygen is a major element, and sulphur is a trace element. Addition of a small amount of sulphur from a sedimentary source containing high $\delta^{34}\text{S}$ will be noticeable, but similar contamination of a magma



7 Diagram showing different oxygen isotope data for different sectors of the Platreef. Data for Malmani dolomite footwall from Harris and Chaumba¹⁶

with a $\delta^{18}\text{O}$ of 7‰ by a hornfels with a $\delta^{18}\text{O}$ of 10‰ will not make a noticeable difference. It is, however, important to note that the Platreef pyroxenites in contact with shale/quartzite have slightly higher $\delta^{18}\text{O}$ values than the RLS in general, and are also different to those recorded at Sandsloot (Fig. 7).²⁹ This indicates either a greater degree of crustal contamination at Sandsloot, or that contaminants at Sandsloot have higher $\delta^{18}\text{O}$ values. We believe that the latter of these two possibilities is the more likely, with $\delta^{18}\text{O}$ values for the dolomite footwall at Sandsloot being much higher than the $\delta^{18}\text{O}$ values for hornfels and quartzite in the south. This agrees with the conclusions reached by this study from the interpretation of sulphur isotope data.

Previous authors^{16,17} have recorded $\Delta_{\text{plag-px}}$ values of approximately +0.5‰, indicating that silicate minerals were magmatic. These authors suggest that this indicates that contamination of the magma occurred pre-intrusion, probably in a staging chamber. However, the $\Delta_{\text{plag-opx}}$ values recorded in this study do not completely support the view that the silicate minerals in the Platreef were formed through magmatic processes. The data presented in this study suggest that some sub-solidus re-equilibration has occurred. This may be due to deuteric fluids moving through the northern limb in the final stages of its formation and possibly causing the recrystallisation of certain silicate minerals. For example, this may be the result of a granitic component flushing through the system, which is now represented in the Platreef as quartzo-feldspathic veins.²¹ This fluid is probably related to the volatile-rich fluid that assisted in the formation of the sulphides within the Platreef.

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Authors

Elizabeth Sharman-Harris completed her BSc Honours specialising in geochemistry at the University of the Witwatersrand in 2004. She is currently completing her MSc at the same university looking at the sulphide mineralisation of the Platreef. This year, she won the Inaugural Institute of Materials, Minerals and Mining Young Persons' World Lecture Competition, held at the Institute's headquarters in London.

Judith Kinnaird is Co-Director of the Economic Geology Research Institute in the School of Geosciences, University of Witwatersrand. She was awarded an Honours BSc degree from the University of London, an MSc and PhD from the University of St Andrews in Scotland and is a chartered engineer. She has taught for the Open University in UK and University College Cork in Ireland. Prior to taking up a post in South Africa in 1999 she was an independent consultant for 6 years. She has worked on tin and columbite deposits in Nigeria, gem-bearing pegmatites in Namibia, Nigeria and Somaliland, uranium deposits in Namibia, lead–zinc deposits in Ireland, copper deposits in Ireland and Zambia, alluvial gold deposits in Mozambique and is currently working on the Platreef.

Chris Harris graduated from Oxford University with a BA in 1979 and DPhil in 1982. He is currently Associate Professor at the University of Cape Town and has been in charge of the Stable Isotope Laboratory in the Department of Geological Sciences since 1989.

Uwe Horstmann received his diploma (MSc in 1981) and doctorate (in 1986) in Geology from the University of Göttingen, Germany. His thesis involved K/Ar dating of sedimentary sequences of the Nama Group in Namibia. As a postdoctoral researcher, he worked on the geochemistry of banded iron-formation at the University of Stellenbosch. After a short stint as lecturer at the then University of Durban Westville, he joined the then Geological Survey of South Africa in 1991, later to become the Council for Geoscience. He was in charge of the stable isotope laboratory, working on a variety of isotope applications from ore-deposit research to environmental investigations. In 2005 he was appointed as head of the Environmental Isotope Group at iThemba Labs (Gauteng) for accelerator-based sciences.