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1 New perspectives on the formation of the Boulder Bed of the western

2 Bushveld Complex, South Africa

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Highlights

- Microtextural characterisation of the Boulder Bed of the Bushveld Complex
- Strongly reverse zoned plagioclase beneath boulders
- Little evidence for microscopic deformation
- A new petrogenetic model for the formation of the Boulder Bed

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Abstract

sub-circular dm-scale 'boulders' of pyroxenite, harzburgite, or norite. To better understand this unit, we have generated high-resolution element maps (µXRF and SEM-EDS) as well as compositional and microtextural data of plagioclase crystals from the boulders and their host anorthosite. Several key features pertinent to understanding the formation of this unit have been described, including: (i) anhedral olivine, orthopyroxene, and negligible base metal sulfides are concentrated at the base of boulders, whereas clinopyroxene is concentrated towards the tops; (ii) the upward decrease in grain size through the boulders; (iii) the occurrence of chromite along the base of boulders and seldom along the

The Boulder Bed of the western Bushveld Complex is an m-scale unit of mottled anorthosite containing

top; (iv) the presence of strongly reverse zoned cumulus plagioclase (An_{75-95}) in the so-called marginal zone underlying boulders; (v) the absence of deformation in the anorthosite but the prevalence of intracrystalline deformation in intercumulus pyroxene; (vi) that amphibole (\pm apatite \pm phlogopite) partially line the bases of some boulders; and (vii) traces of pyrrhotite (\pm pentlandite \pm chalcopyrite) occur within the lower halves of boulders. We propose that the boulders formed in response to the disaggregation of a locally PGE-rich pyroxenite, triggered by heat- and (or) volatile-induced partial melting of the noritic host rocks. Several of the petrologic features arose from the reaction between the boulders and the noritic partial melt, prior to late-stage viscous compaction.

Keywords: Bushveld Complex, Boulder Bed, PGE, Merensky Reef, South Africa, EPMA, EBSD

1. Introduction

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The ~ 2.056 Ga Bushveld Complex of South Africa is the largest known layered intrusion on Earth (Fig. 1A; Eales and Cawthorn 1996; Kinnaird 2005; Smith and Maier 2021). It comprises a ~ 6-8-kmthick sequence of mafic and ultramafic cumulate rocks, named the Rustenburg Layered Suite, that hosts most of the world's platinum-group element (PGE), Cr, and V resources as well as substantial Ni and Cu resources (Cawthorn 2015). The stratigraphy of the Rustenburg Layered Suite is divided into the Marginal, Lower, Critical, Main, and Upper zones (Hall 1924). Stratiform PGE mineralised reefs mostly occur at the base of so-called cyclic units in the upper Critical zone, including the Merensky Reef and the UG2 chromitite. Elevated PGE concentrations (mostly 50-70 ppb, but reaching several ppm in places) are also reported from the enigmatic Boulder Bed (Vermaak 1976; Maier and Barnes 2003; Naldrett et al. 2009), which exclusively occurs in the western lobe of the Bushveld Complex (Fig. 1A). The Boulder Bed is an unusual unit of mottled anorthosite located 10s of metres below the Merensky Reef that contains sub-circular pyroxenitic-harzburgitic aggregates known as 'boulders' (Jones 1976; Maier and Eales 1997). At the Impala and Rustenburg mines, the unit is generally 1 to 5 m in thickness. The boulders are not randomly distributed but rather occur along planar horizons in the host anorthosite. The boulders typically range from 5 to 20 cm in diameter and consistently display a 'right-way up' architecture, i.e., olivine is concentrated at the base and clinopyroxene is concentrated at the top (Jones 1976; Lee and Sharpe 1980). Chromite crystals irregularly line the lower and upper surfaces of boulders, and in places, extend laterally from boulders into the host rocks as stringer veins (Maier and Barnes 2003). In many cases, the boulders are partially rimmed by a thin corona of pure plagioclase, which in turn is rimmed by a \sim 1-cm-thick noritic to gabbronoritic margin. Several authors have likened the boulders to the central pyroxenite of the Merensky Reef, citing

Several authors have likened the boulders to the central pyroxenite of the Merensky Reef, citing their coarse-grain size relative to the host rocks, their association with chromite and anorthosite, and their comparable whole-rock and mineral compositions (Vermaak 1976; Lee and Sharpe 1980; Maier and Barnes 2003). As such, several models have been proposed for the formation of the boulders, including crystallisation from trapped intercumulus melts (Ferguson & Botha 1963; Vermaak 1976) or recrystallisation following the break-up of a pre-existing mafic-ultramafic unit (Jones 1976; Maier and

Barnes 2003). In the present study, we present high-resolution element maps of some boulders together with plagioclase compositions and electron back-scattered diffraction (EBSD) maps to test the proposed petrogenetic models for the Boulder Bed and reveal insights into the formation of other enigmatic units in the upper Critical zone, namely the Merensky Reef and Pseudoreefs.

2. Summary of previous work on the Boulder Bed

Lithological and petrological descriptions of the Boulder Bed and its host rocks have been given by several authors (Ferguson and Botha 1963; Cousins 1969; Van Reysen 1971; Vermaak 1976; Jones 1976; Lee and Sharpe 1980; Viljoen and Hieber 1986; Naldrett *et al.* 1986; Leeb-du Toit 1986; Maier and Eales 1997; Maier and Barnes 2003). At the Impala and Rustenburg mines, where the boulders are most comment, the Boulder Bed is a 1-5-m-thick interval of mottled anorthosite that occurs 20-60 m below the Merensky Reef (Fig. 1B; Maier and Barnes 2003). The unit contains sub-circular 'boulders' of (olivine)-pyroxenite, harzburgite, or norite. Olivine-pyroxenitic boulders are the focus of this study, and they contain subhedral to anhedral orthopyroxene (Mg₇₃₋₈₃), as well as anhedral olivine (Fo₈₂₋₉₁), plagioclase (An₇₁₋₇₉), and clinopyroxene (Mg₇₆₋₈₄), and minor amounts of chromite, quartz, apatite, and sulfide (primarily pyrrhotite; Lee and Sharpe 1980; Maier and Eales 1997; Maier and Barnes 2003).

Olivine and accessory sulfides are concentrated in the lower portion of boulders, whereas plagioclase and clinopyroxene are concentrated in the upper portion (Vermaak 1967; Maier and Barnes 2003). Approximately two-thirds of boulders possess thin (< 1 cm) chromite selvages along their basal surface, which in some cases extend laterally into the host rock (see Fig. 2C of Maier and Barnes 2003; Leeb Du-Toit 1986; Van Reysen 1971). Boulders seldom display chromite selvages along their upper margin. Amphibole, chlorite, and Mg-mica are found along the base of some 20% of boulders, and more rarely, along their upper contacts (Vermaak 1976; Maier and Barnes 2003).

Orthopyroxene and plagioclase within boulders are less compositionally evolved than those in the host anorthosite, yet comparable in composition to those of the Merensky footwall norite (Jones 1976; Maier and Eales 1997). Jones (1976) recorded reverse zoning in cumulus plagioclase crystals in the

host anorthosite (An₇₆₋₈₀), a feature that has been observed in mottled anorthosite elsewhere in the Upper Critical Zone (Maier and Eales, 1997; Maier *et al.* 2021). The composition of chromite crystals in and around the boulders (Cr/Fe = 0.63-1.23, Cr/Al = 1.35-2.8, and Mg# = 0.26-0.29; Maier and Eales 1997) is comparable to those in the bracketing noritic rocks but more evolved than those in Critical Zone chromitite seams.

The boulders are broadly circular in plan (average diameter of 225 \pm 40 mm; *e.g.*, Cousins 1969) and flattened in section (average thickness of 105 mm; Lee and Sharpe 1980). Their long axis is oriented mostly sub-parallel to the lithologic layering (Cousins 1969; Jones 1976). Their lower surface tends to be more convex than their upper surface (Lee and Sharpe 1980). The boulders are coarse-grained (Lee and Sharpe 1980; Maier and Barnes 2003), yet their grain size generally decreases upward and towards their margins (Vermaak 1976). Contacts between boulders and the host anorthosite can be knife-sharp (Cousins 1969) or diffuse (Fig. 2A-B). Some boulders are surrounded by so-called "bleached zones" of anorthosite that can range from several mm to \geq 1 cm in thickness (Ferguson and Botha 1963; Jones 1976). Somewhat analogous features have been described in anorthosite and leuconorite within the Merensky-UG2 interval, whereby "micro-boulders" of monomineralic anorthosite are cored by intercumulus pyroxene and sometimes olivine (Maier 1995; Maier and Eales 1997; Maier *et al.* 2021). Viljoen and Hieber (1986) have reported evidence for macroscopic deformation of the Boulder Bed, showing boulders slumping transgressively into their noritic footwall, producing a pothole (Fig. 2C).

There are relatively few published whole-rock compositional data for the Boulder Bed (Jones 1976; Lee and Sharpe 1980; Naldrett *et al.* 1986; Maier and Barnes 2003). The major element composition of the boulders is comparable to that of the Merensky coarse-grained pyroxenite (18.7-22.2 wt.% MgO and 5.2-8.1 wt.% CaO), yet with relatively lower, albeit highly variable S (~ 230-18,000 ppm) and chalcophile element (~ 50-70,000 ppb PGE, ~ 12-3,000 ppm Cu, and ~ 400-8,500 ppm Ni) concentrations (Jones 1976; Lee and Sharpe 1980; Maier and Barnes 2003). The trace element concentrations (including rare earth elements) are comparable to those of the Merensky footwall norite, where slight positive Eu anomalies indicate the presence of cumulus plagioclase (Maier and Barnes

2003). Naldrett *et al.* (2009) reported elevated PGE contents in the entire Boulder Bed from a drill core intersecting much of the interval between the Merensky Reef and the UG2 unit at Rustenburg mine (Fig. 3).

While the 'Boulder Bed' *sensu stricto* is only recognised in the southern portion of the western Bushveld Complex (mainly at Impala and Rustenburg mines, with rare boulders found further east; Fig. 1), 'boulders' associated with anorthosite have also been reported in the Bastard Reef of the northern lobe (Fig. 2D) and the hanging wall of the UG2 pyroxenite in an open pit south of Lonmin's Karee mine (Fig. 2E; Cawthorn *et al.* 2018). At Lonmin's Karee mine, boulders occur immediately above the UG2 in a ~ 2-m-thick, largely monomineralic anorthosite containing dm-sized domains of mottled anorthosite. Notably, the boulders often occur along sublayer contacts in the anorthosite (Fig. 2E; Cawthorn *et al.* 2018)

3. Samples & Methods

3.1. Optical microscopy and element mapping

Polished thick sections of the Boulder Bed (Tab. 1) were scanned and photographed in reflected light using a Leica MZ12s microscope. High-spatial resolution element maps were produced at Cardiff University using a Carl Zeiss Sigma HD Analytical Field Emission Gun Scanning Electron Microscope (FEGSEM) equipped with two Oxford Instruments 150 mm² energy dispersive X-ray spectrometers. All X-ray element maps were produced using an accelerating voltage of 20 kV, a 120 μm final aperture in high current mode, with a nominal beam current of 8.5 nA and a dwell time of 5,000-20,000 μs, at a working distance of 8.9 mm. Although the electron beam stays stable within 1% relative over periods of hours, beam current drift was optimised using a pure cobalt standard between element maps. Magnification and pixel dwell time were selected depending on the analyte (*i.e.*, entire sections or fine-scale oscillatory zoning). An additional polished block (~ 10 x 5 cm) was mapped using a Bruker Tornado micro-XRF at CSIRO (Melbourne, Australia) at a 40 μm spatial resolution (see Barnes *et al.* 2020). Modal mineralogy was calculated from element maps using ImageJTM software.

3.2. Electron backscatter diffraction (EBSD)

Electron backscatter diffraction (EBSD) is an SEM-based technique that collects data on the crystallographic orientation of any crystalline material. The studied sections were polished mechanically using a 0.3 µm suspension before being cleaned with deionised water prior to chemomechanically polishing with colloidal silica for 20 minutes. The samples were again cleaned with deionised water before being dried in air and coated with a 5 nm thick layer of carbon using an Agar Turbo carbon coater. The EBSD analysis was performed using the Zeiss Sigma HD FEGSEM with a 20 kV beam energy, and a 120 µm final aperture with the high current option activated resulting in a nominal beam current of ~8.5 nA. The sample was mounted at 70° to the incident electron beam at a working distance of 20 mm and electron backscatter patterns were recorded using an Oxford Instruments Nordlys EBSD detector inserted to 191 mm and AZtec version 5.0. Phases identified included anorthite and albite, augite, chromite, hypersthene and forsterite from the American Mineralogist and HKL databases. The EBSD maps were collected with a step size of 18 µm, using 2x2 binning with an exposure time of ~ 60 ms with a Hough resolution of 60 and a minimum number of bands of 8. Mean angular deviations (MAD) of studied phases were below 1.0 apart from augite (1.3% modal fraction) which had a MAD of 1.06. Approximately, 80.6% of pixels were indexed with 19.4% of pixels being zero solutions.

Data processing was performed using AZtec Crystal and the open-source MTex toolbox package (Bachmann *et al.* 2010). Grain reconstruction was performed using 10° thresholds on crystals greater than or equal to 10 pixels. The two-dimensional physical properties of well-constrained crystals from the host anorthosite and marginal zones were exported from AZtec Crystal and used to determine crystal size distributions (CSD). The *CSDslice* spreadsheet (Morgan and Jerram 2006) was used to estimate three-dimensional crystal habits, which were input into *CSDcorrections* software (Higgins 200) together with the plagioclase major and minor fitted ellipse length, the ellipse angle, and the grain area to produce CSD profiles. The average roundness, modal abundance, and fabric quality (or *shape preferred orientation* which is quantified by the *alignment factor*) were also used when producing the CSD profiles.

The crystallographic preferred orientation (CPO) of plagioclase is visualised using an orientation density function (ODF) that was calculated using a 10° halfwidth. The CPO strength is quantified by the pole figure J-indices (pfJ) for each axis, as well as the J- and M-indices on the orientation distribution functions (Bachmann *et al.* 2010; Mainprice *et al.* 2015; Jenkins *et al.* 2021). Theoretically, the pfJ and J-index (Bunge 1982) range from 1 (completely random) to infinity (single crystal), whereas the M-index (or *misorientation index*; Skemer *et al.* 2005) ranges from 0 (completely random) to 1 (single crystal). Lastly, the method of Cheadle and Gee (2017) was used to describe the three-dimensional orientation of crystals by calculating the *foliation* (F#) and *lineation* (L#) numbers. The F# parameter is defined as the ratio of the maximum to intermediate eigenvalues for [010] and the L# parameter is defined as the ratio of the maximum to intermediate eigenvalues for the [100] axes.

3.3. Electron probe microanalysis (EPMA)

Quantitative chemical analysis of plagioclase crystals was conducted at Camborne School of Mines (University of Exeter) using a JEOL JXA-8200 electron-probe microanalyser. Analyses were performed using a 30 nA electron beam accelerated to 15 kV with a beam diameter between 1 and 10 μm. Analyses were made using wavelength dispersive spectrometers and were calibrated to natural mineral standards supplied by P&H Developments and Astimex Scientific and quantified using the CITZAF φρZ method of Armstrong (1995) implemented for JEOL. Standards analyses and results are provided in Electronic Supplementary Materials 1 and 2 (ESM 1-2).

4. Results

4.1. Petrography & element mapping

The anorthosite hosting the boulders is composed of ~ 92-95 mod.% cumulus plagioclase, with patches of interstitial clinopyroxene (~ 4-5 mod.%) and orthopyroxene (~ 1-4 mod.%), as well as traces of phlogopite, apatite, and quartz ($\ll 1 \text{ mod.\%}$; Figs. 4-7). While section thickness precludes typical transmitted light microscopy, no undulose extinction, deformation to plagioclase twin lamellae, or

neoblasts were observed in our samples (ESM SF1). The petrographic observations of the boulders are summarised in Table 1.

Samples BBBC-BB-BBTC: These sections sample the base, middle, and top of a boulder from the Brakspruit shaft of the Rustenburg platinum mine (Fig. 4). On the scale of the whole boulder, anhedral olivine (~ 7-8 mod.%) is concentrated near the base, oikocrystic clinopyroxene (~10-12 mod.%) is concentrated near the top, and chromite (~ 0.5-1 mod.%) occurs both near the base and, to a somewhat lesser degree, along the upper margin. Orthopyroxene occurs throughout the boulder, showing an overall upward decrease in grain size from > 5 mm to ~ 0.5-3 mm in diameter.

Relative to the bulk of the anorthosite hosting the boulder, the immediate floor rock to the boulder contains an elevated proportion of interstitial orthopyroxene (~ 5-10 mod.%) for ~ 1-2 cm as well as subordinate outer patches of clinopyroxene. Hereafter, we refer to the zone comprising intercumulus orthopyroxene as the *marginal zone* (bound by dashed lines in Figs. 4-7). Subhedral plagioclase (~ 90-95 mod.%) constitutes the remainder of the marginal zone mineralogy, together with traces of very fine-grained sulfide, chromite, and phlogopite.

Anhedral and partially serpentinised olivine (~ 20-25 mod.%) containing thin magnetite veins (~ 0.5 mod.%) occur at the base of the boulder where it has been partially replaced by relatively coarse-grained orthopyroxene (~ 50-55 mod.%). The grain size of the composite olivine-orthopyroxene crystals is < 2.4 cm in diameter. Variably-sized aggregates and blocky crystals of chromite (< 1-2 mm in diameter) occur at the margins of olivine and orthopyroxene crystals. Coarse intercumulus plagioclase (~ 15-20 mod.%), fine-grained phlogopite (~ 0.5-1 mod.%) and very fine-grained sulfides (Po > Pn » Ccp; ~ 0.2-0.5 mod.%) generally occur within and at the margin of orthopyroxene crystals. Only traces of very fine-grained quartz, apatite, and secondary magnetite were identified.

The centre of the boulder is characterised by variably sized (0.2-1.8 mm in diameter), subhedral orthopyroxene crystals (~ 70-75 mod.%), with interstitial plagioclase (~ 10-15 mod.%). Clinopyroxene (~ 10-15 mod.%) sometimes occurs along the grain boundaries between plagioclase and orthopyroxene. It also forms relatively large (< 1.4 cm in diameter) interstitial crystals. Only very finely disseminated

sulfides were identified. Magnetite (~ 0.6 mod.%), chlorapatite (~ 1-2 mod.%), and phlogopite (< 1 cm in diameter, ~ 2-3 mod.%) rarely occur with partially uralitized clinopyroxene.

In the upper portion of the boulder, orthopyroxene (~ 50-55 mod.%) forms subhedral crystals (< 1 cm in diameter). The modal abundance of oikocrystic plagioclase is comparable to that of subhedral plagioclase at the base of the boulder (~ 25-30 mod.%). Partially uralitized clinopyroxene (~ 20-25 mod.%) occurs as large (< 1.9 mm in diameter) interstitial crystals that sometimes enclose orthopyroxene crystals. Traces of very fine-grained phlogopite and chromite are generally spatially associated with clinopyroxene. No sulfides or apatite were identified. There is only limited exposure of the upper margin of the boulder, which is relatively sharp, displaying a cluster of several blocky chromite crystals.

Sample BV-BB1: This sample is from the base of a boulder from Impala mine and was mapped using the micro-XRF (Fig. 5). The base of the boulder is partially circumscribed by a zone of pure plagioclase (~ 5 mm wide) as well as plagioclase-rich zones with intercumulus pyroxenes. Moreover, the size and abundance of intercumulus pyroxene gradually decrease with distance from the boulder. Anhedral olivine (< 1 cm in diameter) is concentrated near the base of the boulder. It shows irregular, strongly rounded morphologies and is enclosed within coarse-grained orthopyroxene. The olivine is partially replaced by secondary magnetite formed during serpentinization. Coarse-grained orthopyroxene and interstitial plagioclase comprise the bulk of the boulder in this sample. Mediumgrained chromite occurs at the base of the boulder, notably around the margins of olivine, whereas finergrained chromite crystals are observed within the boulder (Fig. 5). Traces of calcite and amphibole were also identified.

Samples Bou-2 and TF-Bou: These sections sample the bases of boulders from Turffontein section of Rustenburg Platinum Mine (Figs. 6-7). Their marginal zones comprise cumulus plagioclase (\sim 75-85 mod.%) and intercumulus orthopyroxene (\sim 10-15 mod.%). Directly below the marginal zones, clinopyroxene (\sim 3-5 mod.%) becomes the dominant intercumulus phase. Traces of fine- to medium-grained amphibole and chromite occur at the margins of intercumulus orthopyroxene. Chromite crystals sometimes form clusters of \geq 2 crystals (\sim 3-5 mod.%).

The basal portions of the boulders comprise relatively coarse-grained and partially serpentinised (< 3 mm in diameter; ~ 40-50 mod.%) olivine that is partially rimmed by orthopyroxene (~ 10-35 mod.%). The base of sample Bou-2 (Fig. 6) is lined by a ~ 0.5 cm rim of amphibole (~ 10 mod.%), together with traces of very fine-grained mica, sulfide, and apatite – this is not observed in sample TF-Bou (Fig. 7). Plagioclase (~ 15-30 mod.%) in the marginal zone occurs as fine- to medium-grained cumulus crystals with intercumulus orthopyroxene. Fine-grained chromite crystals (< 3 mod.%) form clusters of two or more crystals that are typically rimmed by orthopyroxene (Figs 6B and 7B). Fine-grained phlogopite and sulfides (Po > Pn and no Ccp; < 0.5 mod.%) occur at the margins of olivine-orthopyroxene crystals (within plagioclase-rich domains), yet sulfides and phlogopite are not spatially associated.

Sample BB-B2C: This samples the middle of a boulder from Impala mine (Sup. Fig. 1). The section has several similarities with the central portion of the boulder from Brakspruit section (Fig. 4) including: (1) the grain sizes of plagioclase and orthopyroxene decrease upward through the boulder; (2) olivine and orthopyroxene host fine grained chromite; (3) the mode of uralized clinopyroxene increases upward through the boulder; (4) fine-grained magnetite is spatially associated with clinopyroxene; (5) interstitial phlogopite and Cl-rich apatite are spatially associated with each other; and (6) only very fine-grained sulfides are present.

4.2. Plagioclase composition

Cumulus plagioclase in the host anorthosite: Six plagioclase crystals from the underlying anorthosite (samples BBBC, Bou-2, and TF-Bou) and two crystals from the overlying anorthosite (sample BBTC; Fig. 8A; ESM 1) were chemically analysed rim-to-rim. The crystals are ≤ 1 -2 mm in diameter and possess An contents mostly ranging from ~ 80 -75 mol.% with seemingly no systematic variation indicative of compositional zoning. There is, however, one exception (transect AE in Figure 7) occurring just beneath the marginal zone, where the outer ~ 0.2 mm is up to 10 mol.% more anorthitic than the corresponding transect centre.

Cumulus plagioclase in the marginal zone: Fifteen crystals present in the marginal zones were analysed (Fig. 8B; ESM 1). The crystals have broadly similar morphologies as those in the host anorthosite (~ 1-2 mm in diameter and subhedral), yet those present in the marginal zones record subtle to strong reverse zoning, where the outermost analyses can be up to 15 mol.% more anorthitic than the corresponding central analyses (see also Fig. 8D). The centres of crystal transects have An contents of ~ 83-76 mol.% and the corresponding transect margins (*i.e.*, the outer 0.5-0.3 mm) have An contents of ~ 90-95 mol.%.

Plagioclase within boulders: Fifteen crystals present within the defined outline of the boulders were analysed (Fig. 8C; ESM 1), this includes: (i) two cumulus grains (Fig. 6); (ii) seven intercumulus grains from the lower portions of boulders (Figs 4, 6, and 7; ESM Figure S2); and (iii) six intercumulus grains from the upper portions of boulders (Fig. 4). The two cumulus grains in section Bou-2 (AA and AB; Fig. 6) are the smallest (< 1 mm in diameter) and show pronounced reverse zoning comparable to crystals sampled in the marginal zone (*i.e.*, An contents of transect edges at > 10 mol.% greater than corresponding centres). Intercumulus plagioclase crystals sampled from the lower halves of boulders have relatively constant An contents (~ 78-74 mol.%), yet with an outer ~ 0.1-0.2 mm that is up to 10 mol.% more anorthitic. In contrast, intercumulus plagioclase sampled from the upper halves of boulders have markedly lower constant An contents (~ 71-67 mol.%) with no clear systematic variation. However, two exceptions to this (transects M and N; Fig. 4) have An contents of ~ 78-79 mol.% with slightly more anorthitic rims; these grains occur amongst the chromite-bearing upper surface of a boulder.

Microstructural analysis of plagioclase crystals

Plagioclase crystals from the host anorthosite and marginal zone of section TF-Bou (Fig. 7) were analysed by EBSD (Figs. 9, 10 and 11). The physical characteristics of plagioclase crystals are illustrated in Figures 9A-C, summarised in Table 2, and fully reported in ESM 3. These data show that crystals in the host anorthosite and marginal zone have near-identical physical properties (Table 2).

The crystal size distribution (CSD) of plagioclase crystals was determined using the CSDcorrections software (Higgins 2002) and the crystal shape was determined using CSDslice (see Tab. 2; Morgan & Jerram 2006). The CSD patterns of plagioclase crystals in the host anorthosite and marginal zone have similar profiles, in that they display convex-up patterns in crystals ranging from ~0.2 to 0.5 mm and broadly flat to shallow concave-up patterns for crystals > 0.4 mm (Fig. 9D). Our CSD profiles are shallower than those recorded by Upper Critical Zone crystals sampled from the Jagdlust section of the eastern lobe of the Bushveld Complex (Boorman *et al.* 2004; Fig. 9D), but are steeper, with less-defined convex-up patterns at smaller crystal sizes, than those from the JM reef package rocks of the Stillwater Complex (Jenkins *et al.* 2022).

Rose diagrams were also produced using CSDcorrections, which show that the plagioclase crystals are generally aligned parallel to rock layering in the Critical Zone, and thus, parallel with the long axes of boulders (Jones 1976). The strength of the mineral alignment (*i.e.*, shape preferred orientation; SPO) is quantified using the alignment factor, whereby a value of 1 represents perfect alignment and a value of 0 represents perfectly random orientation (see Holness *et al.* 2020). Plagioclase in the host anorthosite have an alignment factor of 0.28 and those in the marginal zone have an alignment factor of 0.11 (Tab. 2)

Electron backscatter diffraction analysis was conducted at the base of the boulder TF-Bou (Fig. 7). Our aim was to identify potential evidence for viscous deformation (such as dislocation creep or diffusion-controlled processes such as dissolution-reprecipitation; Holness *et al.* 2017; Vukmanovic *et al.* 2019) in cumulus plagioclase crystals that host the boulders. Typically, magmatic samples that have undergone significant viscous deformation through compaction have a fabric defined either by crystal shapes or preferred orientations (Holness et al. 2017). Features such as dislocation creep (as evidenced by undulose extinction or mechanical twins) and dissolution-reprecipitation (as evidenced by truncation of crystals, the interpenetration of crystals, or suture contacts) may also be expected.

Figure 10 represents equal-area, lower hemisphere pole figures of the [100], [010], and [001] axes distribution for plagioclase crystals in the host anorthosite, marginal zone, and entire sample. Because of the ubiquitous presence of twins in the plagioclase crystals, we calculated the orientation distribution

function (ODF) using every measurement available, using a 10° halfwidth for the calculation. The pole figures display the orientation of the crystallographic axis of plagioclase crystals in three dimensions, where the strength of axis distribution is quantitatively assessed using the pfJ, J-, and M- indices (Table 2) and their relative symmetry is reported using the F# and L# values (Methods section; Fig. 10; ESM 4). If the crystallographic axes are distributed in a way that is non-random, the subject phase has a crystallographic preferred orientation (CPO). A non-random distribution may manifest as a point maximum if the axes are clustered along a specific direction or as a girdle if the axes are randomly distributed along a plane.

Pole figures of plagioclase crystals in the host anorthosite (Fig. 10B) show a cluster of [010] axes, *i.e.*, a direction that is perpendicular to the bedding plane of the host rock. The [100] axes are found to cluster on the bedding plane. While the fabric strength is low (AF = 0.28; J-index = 4.21; M-index = 0.03), our data indicate a non-random orientation of plagioclase in the host anorthosite, one that is comparable to B-type axis fabrics that are known to develop in tabular cumulus minerals of layered intrusion (Cheadle and Gee 2017; Holness *et al.* 2017). Conversely, pole figures of plagioclase crystals in the marginal zone show no clearly defined CPO (Fig 10C), as indicated by their relatively weaker fabric indices (AF = 0.11; J-index = 2.53; M-index = 0.00) and their lower maximum mud reached.

Figure 11A is a map of grain reference orientation deviation (GROD) angle, which reports the angular deviation of each pixel within a given grain relative to the mean orientation of the same grain. Figure 11B is a grain orientation spread (GOS) map of plagioclase, where individual grains are coloured by the average of misorientation angles recorded within each pixel of a given grain relative to the average orientation of the same grain. In GROD and GOS maps, higher values (Fig. 11) correspond to higher degrees of intragranular deformation (Brewer *et al.* 2009; Allain-Bonasso *et al.* 2012). Plagioclase in both the host anorthosite and marginal zone display negligible degrees of misorientation, inconsistent with significant plastic deformation (Brewer *et al.* 2009). This determination is further supported by the absence of neoblasts and the preservation of magmatic twins. Only localised misorientation is seemingly randomly distributed throughout plagioclase crystals of the marginal zone.

Microstructural analysis of ferromagnesian crystals

Hypersthene, augite, and forsterite were also indexed during the acquisition of EBSD data. While there are too few of these crystals to illustrate and quantify meaningful fabric data, misorientation within these crystals can still be visualised. Figure 11C is a GROD angle map of forsterite, where while a subgrain is present in the upper left portion of the map, no systematic intra-crystalline deformation is present. Furthermore, the textural relationship olivine shares with cumulus plagioclase indicates that it crystallised after plagioclase. Figures 11D-F illustrate GROD angle and grain maps for hypersthene. Hypersthene oikocrysts in the marginal zone display variable degrees of intra-crystalline misorientation possibly consistent with a limited degree of plastic deformation. Our data illustrates that the hypersthene present in the marginal zone, as well as augite present in the host anorthosite, crystallised after plagioclase and that no further episode of deformation, plastic or magmatic, occurred after that.

5. Discussion

- 5.1. Summary of key characteristics
- Several key features of the Boulder Bed from this and past studies are summarised in Figure 12 and listed below:
 - Orthopyroxene (En₈₂₋₈₁; Jones 1976) and interstitial plagioclase crystals decrease in size from
 the base to the top, whereas interstitial clinopyroxene crystals increase in size and mode from
 the base to the top
 - The marginal zone consists of an intercumulus orthopyroxene-dominated zone along the contact to the host anorthosite transitioning to an intercumulus clinopyroxene-dominated zone outwards.
 - 3. Chromite crystals commonly occur at the base of boulders and in the marginal zone but never in the host anorthosite (see also Van Reysen 1971; Jones 1976; Leeb du Toi 1986; Maier and Barnes 2003)

4. Partially serpentinised olivine (Fo₈₅₋₈₂; see also Van Reyson 1971; Jones 1976; Maier and Eales

1997) and very fine-grained sulfides (pyrrhotite ± pentlandite > chalcopyrite) predominantly

occur at the base of the boulders and seldom in the marginal zone

- 5. In ~ 20% of boulders (Maier and Barnes 2003) the lower surface (and less commonly the upper surface) is lined with intercumulus amphibole and subordinate biotite (Fig. 6)
- 6. Phlogopite occurs both at the margins of olivine-orthopyroxene crystals at the base of boulders and as coarse crystals in the centre of boulders. There is no spatial relationship between phlogopite and sulfides as described for the Merensky Reef (Smith *et al.* 2021)
- 7. Cumulus plagioclase crystals in the marginal zone and at the bases of boulders show strong reverse compositional zoning, where rims are up to 10 mol.% more anorthositic than the corresponding transect centres. In contrast, cumulus plagioclase of the host anorthosite lacks systematic reverse compositional zoning (Fig. 8) or evidence for deformation (Fig. 10)
- 8. The plagioclase crystals in the host anorthosite (Fig. 10C) show evidence of a weak planar fabric parallel to rock layering in the Critical Zone, consistent with B-type fabrics exhibited by elongated cumulus phases in layered intrusions (Cheadle and Gee 2017; Holness *et al.* 2017). The long axes of boulders are also aligned parallel to this. Plagioclase crystals in the marginal zone display no discernible fabric.
- 9. Interstitial pyroxenes in the marginal zones record evidence for intra-crystalline deformation whereas adjacent cumulus plagioclase record little-to-no evidence for deformation. This suggests that either pyroxenes are rheologically weaker than plagioclase at magmatic temperatures, and hence more readily accommodated stress from viscous compaction, or that whilst intercumulus melts were crystallising, the stress from viscous compaction was sufficient to shear these crystals as they formed.
- 10. Naldrett *et al.* (2009) analysed a drill core that sampled the entire ~ 10 m anorthosite unit hosting the boulders at Rustenburg. The anorthosite unit has low S, Ni, Cu, and Au concentrations and high PGE concentrations relative to the bracketing norite units;
- 11. Figure 2C (from Viljoen and Hieber 1986) shows localised evidence for slumping of the Boulder Bed into its noritic footwall (*i.e.*, producing a pothole). Notable features outside of the

pothole, include: (i) the Boulder Bed consists of crudely banded anorthosite containing boulders as well as pyroxene mottles that appear to be preferentially concentrated in specific horizons; and (ii) the pyroxene mottles show increasing elongation towards the centre of the slump structure. Notable features within the pothole, include: (i) boulders are rotated while remaining largely of similar size and shape, except for sharpening of their margins; and (ii) the anorthosite is almost monomineralic, except for a few streaks of pyroxene.

5.2. Previous models for the formation of the Boulder Bed

The origin of the Boulder Bed has remained controversial, due to its highly unusual features including the circular, flattened shape and sharply defined margins of many boulders, their relatively uniform size, and their location within an anorthosite layer. A number of contrasting petrogenetic models addressing the formation of the Boulder Bed have been proposed.

- (i) *In situ* models advocate the crystallisation of pyroxene and plagioclase from residual melt trapped in a relatively impermeable anorthosite matte (Ferguson and Botha 1963; Vermaak 1976; Lee and Sharpe 1980). The model could explain the spherical shape and 'up-right' crystal sequence of the boulders as well as the occurrence of amphibole (and mica) in some boulders relative to the remainder of the Critical Zone. However, the model is inconsistent with the lateral alignment of boulders, an abundance of relatively unevolved olivine, orthopyroxene, and plagioclase, the presence of chromite, and the negligible proportions of quartz in the boulders. A model of essentially *in situ* fractionation also has difficulty accounting for the high PGE concentrations (including the relatively immobile IPGE) of boulders relative to the host anorthosite (Fig. 3; Maier and Barnes 2003; Naldrett *et al.* 2009).
- (ii) *Ex situ* models include the sinking of boulders formed from magma 'fingers' generated during the influx of new magma (Campbell *et al.* 1983) and the sinking of fragments of a structurally-disrupted pyroxenitic layer (Jones 1976), which may originally have formed part of the Merensky Reef or correlate to the Pseudoreefs of the northern portion of the western Bushveld lobe (Van Reysen 1971; Jones 1976; Maier and Eales 1997). The latter model can explain the relatively unevolved composition

of the main rock-forming minerals, the PGE enrichments in some boulders, and the fact that boulders and the Pseudoreefs never occur at the same locality. However, the break-up of a 'Merensky Reef' or Pseudoreef would be expected to result in fragments of variable size, shape, and orientation, and chromite would not be expected to line the surfaces of boulders. More importantly, the model cannot explain the occurrence of boulders in anorthosite overlying the UG2 pyroxenite (Fig. 2E).

(iii) Maier and Barnes (2003) proposed a modification of the *ex situ* model whereby a broken-up pyroxenite layer and its noritic host were modified by late-magmatic reactive porous flow involving an ascending Si-undersaturated volatile phase. This model could explain the association of unevolved pyroxene, olivine and plagioclase with amphibole, the lack of quartz, the chromite selvages, the reversely zoned plagioclase and the presence of anorthositic bleach zones around some of the boulders, and the formation of the anorthosite layer hosting the boulders. However, as in the case of other *ex situ* models (*e.g.*, Jones 1976), the relatively uniform size and shape of the boulders remains enigmatic.

(iv) Maier (1995) and Maier et al. (2021) described 'microboulders' or 'mottles' in the Upper Critical Zone sampled at the Wildebeestfontein North Section at Impala Platinum Mines. The mottles contain anhedral olivine rimmed by intercumulus orthopyroxene, surrounded by a halo of strongly reverse zoned plagioclase and phlogopite. They proposed that these features formed in response to a proto-norite being fluxed by acidic fluids migrating along layer contacts. The fluids triggered the incongruent dissolution of pyroxenes as well as the leaching of alkalis from plagioclase to form a calcic anorthositic restite of reversely zoned plagioclase. Secondary olivine and pyroxene locally precipitated in fluid channels, where mixing occurred between relatively cold Si-undersaturated and hot Si-saturated fluids.

5.3. Towards an internally consistent petrogenetic model

The data presented in this study suggest that the Boulder Bed formed through both primary magmatic and secondary hydromagmatic processes. We distinguish the following petrogenetic stages:

Disintegration of proto-cumulates: We hypothesise that a (leuco)noritic cumulate pile, representing the temporary chamber floor, was overlain by sulfide-bearing pyroxenitic cumulates upon chamber replenishment following Nicholson and Mathez (1991; Fig. 13A). The heat from the replenishment event triggered partial melting of the (leuco)norite, generating an anorthosite restite and a reaction front between the ultramafic cumulates and upwelling partial melts (Fig. 13B-C), as suggested previously at the Basistoppen Sill (Naslund 1986), Rum (O'Driscoll et al. 2009) and the Bushveld Complex (Eales et al. 1986; Scoon and Mitchell 2012; Scoon and Costin 2018). This process could explain the relatively thicker marginal zone at the bases of boulders (this study; Fig. 4), the precipitation of chromite by Cr₂O₃ liberated from the partially molten anorthosite (Scoon and Costin 2018; Figs. 4, 6, and 7), and the occurrence of unevolved anhedral olivine via incongruent dissolution of orthopyroxene (Shaw and Dingwell 2008; Maier et al. 2021; Marsh et al., 2021; Figs. 4-7). If the replenishing magma intruded the noritic cumulates as a sill (e.g., Mungall et al. 2016), both the noritic floor and roof rocks may partially melt to form bracketing anorthosite. In either case, the pyroxenitic cumulates could sink and dismember prior to complete solidification to form fragments within the feldspathic mush that would later become boulders. Alternatively, fluxing of volatiles from cooler rocks at deeper levels of the cumulates could have triggered the partial dissolution of noritic proto-cumulates to form anorthosite (Meurer et al. 1997; Maier et al. 2021).

Deformation of boulders and bracketing anorthosite: The hypothetically partially molten and (or) dissolved pyroxenite layer was rheologically weakened and underwent hot subsimple shearing triggered by the sliding of hanging-wall rocks towards the centre of the progressively subsiding intrusion (Maier et al. 2013; Vukmanovic et al. 2019). While there is evidence for localised macroscopic deformation (Fig. 2C), crystal-scale deformation was only recorded within intercumulus pyroxenes of the marginal zone (Fig. 11). We interpret this as a result of strain partitioning during late-magmatic pure to subsimple shear in response to viscous compaction of the cumulate pile (Fig. 13D), whereby strain was accommodated within the less-component melt generated during the reaction between the boulders and (leuco)noritic partial melt (Vigneresse and Tikoff 1999). This process can explain the weakly flattened appearance of boulders (Fig. 2E; Holness et al. 2017). If some form of shearing had occurred, one might

expect to see evidence for dynamic recrystallisation (e.g., bent twins, undulose extinction, sub-grains, serrated or lobate grain boundaries; Holness et al. 2017) recorded in bracketing cumulus plagioclase. Vukmanovic et al. (2019) and Maier et al. (2021) reported microtextural evidence for dynamic crystallisation of cumulus plagioclase within Upper and Critical Zone anorthosites, respectively, and ascribed these features to slumping and subsidence of crystal mush. However, no such features were consistently observed in our samples (Fig. 11; ESM Figure S1). One could argue that static annealing of cumulus plagioclase under super-solidus conditions (> 500°C) diluted or overprinted evidence for internal strain and CPO, while preserving any SPO (Hunter 1996; Heilbronner & Tullis 2002; Piazolo et al. 2006; Holness et al. 2017). The degree of static annealing is likely to increase with intrusion size and depth (Holness et al. 2017) and given the size of the Bushveld Complex (Smith & Maier 2021) and the prolonged cooling history of the cumulates (~ 20-80 kyr; Cawthorn and Walraven 1998), supersolidus conditions could have been sustained. However, static recrystallisation would eradicate the observed reverse zoning (Holness et al. 2017; Robb and Mungall 2020), unless reverse zoning of plagioclase formed after grain annealing. Further microstructural and along-plane investigations are required to understand how these processes gave rise to planar layers of seemingly isolated boulders of comparable size and composition. Reverse zoning of plagioclase: Several studies report the reverse zoning of cumulus plagioclase in Upper Critical Zone anorthosite (Maier and Eales 1997; Robb & Mungall 2020; Smith et al. 2021; Maier et al. 2021). In this study, cumulus plagioclase within the host anorthosite contain no systematic compositional zoning, whereas cumulus plagioclase within the marginal zone become increasingly reversely zoned with proximity to the boulder bases (i.e., increasing in intensity with increasing volumes of intercumulus pyroxenes; Figs. 8 and 12). Reverse zoning of plagioclase may arise from dissolution-reprecipitation (Humphreys 2009; Bennett et al. 2019) or the preferential leaching of Si and Na by Si-undersaturated fluids and (or) melts (Baker and Boudreau 2019; Maier et al. 2021; Marsh et al. 2021). In the absence of core-rim microtextural evidence and the presence of relatively disrupted plagioclase grains (e.g., lower textural indices) with highly anorthitic rims in the marginal zone, we favour Ca-rich rim formation by the preferential removal of alkalis during interaction with residual (leuco)noritic partial melt, which coincided with the precipitation of olivine and spinel at the base of

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515 boulders. A late-magmatic origin for plagioclase reverse zoning is also preferred since under prolonged 516 super-solidus conditions, reverse zoning may not be preserved (Robb and Mungall 2020). 517 Coarsening of boulders: The relatively coarser grain size of the boulders may result from crystal ageing 518 (Ostwald Ripening), whereby larger crystals grow at the expense of smaller crystals (Voorhees 1985). 519 Such a model has been proposed to explain pegmatoidal rocks associated with the UG2 chromitite 520 (Cawthorn and Barry 2007), Merensky Reef (Cawthorn and Boerst 2006), and JM Reef (Jenkins et al. 521 2022). The lower portion of boulders is relatively coarser than the upper portions, which may arise from 522 different cooling rates facilitated by the concentration of interstitial melts, particularly if incongruent 523 dissolution of pyroxene was pronounced at the base of boulders. In CSD profiles, Ostwald Ripening is 524 evidenced by convex-up profiles at smaller grains sizes and fanning profiles at larger grains sizes (Fig. 9). While our plagioclase CSD profiles (Fig. 9) are consistent with that expected from ripened grains, 525 526 uncertainty, particularly at lower grain sizes, means that the operation of Ostwald Ripening of 527 plagioclase in the marginal zone cannot be empirically concluded. Occurrence of olivine and chromite: Chromite crystals occur within and along the bases of boulders, 528 along sides and upper margins of boulders, and may extend laterally as chromitite seams into the host 529 rocks (Maier and Barnes 2003). Chromite crystals may occur as isolated grains (Figs. 4 and 5) or as 530 531 chains surrounded by orthopyroxene, spatially associated with anhedral olivine (Fo91-82; cf. Yudovskaya et al. 2022). Nicholson and Mathez (1991) reported noritic inclusions within the Merensky 532 533 pegmatoidal pyroxenite that were surrounded in turn by a ~ 1 cm thick anorthosite and a < 5 mm thick chromitite; similar sequences occur at the base of the Merensky cyclic unit (Smith et al. 2021) as well 534 as within the Rum (O'Driscoll et al. 2009) and Stillwater (Marsh et al. 2021) complexes. Several models 535 536 for the precipitation of anorthosite-associated chromitite seams have been proposed, many of which 537 involve partial melting (O'Driscoll et al 2009; Mathez and Kinzler 2017; Scoon and Costin 2018; 538 Veksler and Hou 2020) or dissolution (Nicholson and Mathez 1991; Meurer et al 1997; Baker and 539 Boudreau 2019; Marsh et al. 2021) of ultramafic proto-cumulates. In either case, the partial incongruent 540 melting (or dissolution) of orthopyroxene catalysed by evolved partial melts could form the observed 541 forsteritic olivine and Cr-rich spinel concentrated at the bases of boulders (Shaw et al. 1998; Shaw and 542 Dingwell 2008; Marsh et al. 2021). Such a process can account for other features reported in the

marginal zone, including: (i) the reverse zoning of marginal zone plagioclase at boulder bases, particularly the increase in intensity with proximity to the boulder-anorthosite reaction front (Schiffries 1982) and the preservation of compositional zoning (Robb and Mungall 2020); (ii) the relatively coarser grain size of the boulder bases; (iii) olivine-bearing boulders are associated with more calcic plagioclase (Jones 1976); and (iii) the relative increase in amphibole, apatite, and phlogopite abundance (Fig. 6-7) Sulfides and PGE contents of boulders: Models of in situ crystallisation cannot explain the relatively high PGE concentrations of the Boulder Bed (Cu/Pd values < 1000; Maier and Barnes 2003). The PGE contents may instead have been inherited from a pre-existing PGE-rich cumulate layer, of which there are many reported in the western lobe of the Bushveld Complex (Maier et al. 2013). Maier and Barnes (2003) noted that PGE and Au possess positive inter-element correlations (R² values > 0.7). Sulfur relatively poorly correlates with PGE contents (IPGE R² value = 0.43; PPGE R² value = 0.68), yet strongly positively correlates with Ni ($R^2 = 0.97$) and Cu ($R^2 = 0.98$). This suggests that PGE may be concentrated in discrete platinum-group minerals and spinel (Pagé et al. 2012), while Ni and Cu are controlled by sulfides. Sulfur and Cu may be preferentially removed during incongruent dissolution of sulfides catalysed by an S-undersaturated phase, whereby Ni could be retained in mss-derived phases (pyrrhotite and pentlandite) or relocated to olivine (Peregoedova et al. 2004; Maier et al. 2021). During such a process the PGE may concentrate in the residual sulfide (Kerr and Leitch 2005) and (or) undergo desulfurisation to produce platinum-group minerals and alloys (Li and Ripley 2006). Such a model can explain why pyrrhotite (± pentlandite) are the most common sulfides in our olivine-bearing samples, yet the deportment of chalcophile elements amongst boulders warrants further investigation.

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6. Conclusion

The Boulder Bed of the western Bushveld Complex is an m-scale unit of mottled anorthosite containing sub-circular dm-scale 'boulders' of pyroxenite, harzburgite, and norite, occurring 10s of metres below the Merensky Reef. Several petrogenetic models have been proposed for this enigmatic unit, which can be categorised as *in situ* and *ex situ* models. We propose that the boulders are the remains of a PGE-rich ultramafic layer that became dismembered during the heat- and (or) volatile-induced

partial melting of the underlying (leuco)noritic cumulate rocks, which created an anorthosite restite. The upwelling partial melt reacted with the bases of boulders, which led to: (i) the incongruent dissolution of cumulus orthopyroxene and crystallisation of unevolved anhedral olivine; (ii) the coarsening of the lower portions of boulders relative to the upper portions; (iii) precipitation of chromite; (iv) the increase in the intensity of reverse zoning in cumulus plagioclase with proximity to the boulder bases; (v) the presence of amphibole and late-stage silicates along the lower margins of some boulders; and (vi) the dissolution of pre-existing base metal sulfides. Late-stage viscous compaction caused the partial flattening of boulders, where strain partitioned into intercumulus pyroxenes underlying the boulders. Given that magma replenishment, partial melting and (or) dissolution of proto-cumulates, and viscous compaction are likely common processes operating during the formation of layered intrusions, it remains unclear as to why boulders have not yet been described at other layered intrusions.

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777 Figure Captions

- 778 **Figure 1. A.** Geological map of the western lobe of the Bushveld Complex (modified from Yao and
- Mungall 2021), highlighting locations of samples acquired for this study. RPM = Rustenburg Platinum
- 780 Mine. B. Stratigraphic section of the Bushveld Complex (modified from Yao and Mungall 2021) and
- the Upper Critical Zone from drill hole H3 at Wolhuterskop (modified from Maier and Eales 1997).
- Note the location of the Boulder Bed (BB) below the Merensky Reef (MR). Abbreviations: NR = norite,
- 783 TC = troctolite, AN = anorthosite, and PX = pyroxenite.
- Figure 2. A-B. Photographs of the Boulder Bed (Footwall 6) in drill core sampled from the Impala 16#
- shaft. C. Figure 16 from Viljoen and Hieber (1986) showing an example of slumping of the Boulder
- Bed (host anorthosite and pyroxenitic boulder) into the footwall. **D.** Sulfide-bearing pyroxenitic
- boulders hosted in anorthosite of the Bastard Reef in the northern limb of the Bushveld Complex. E.
- 788 The occurrence of pyroxenitic 'boulders' in mottled anorthosite bracketed by norite at the Lonmin
- Marikana UG2 pit. The mottled anorthosite unit has been partially intruded by an iron-rich ultramafic
- 790 pegmatite (pictures provided by D. Reid).
- 791 **Figure 3.** Compositional and modal data from the Merensky footwall (drill hole H3, Wolhuterskop) in
- the western Bushveld Complex (modified from Maier and Eales 1997) combined with S and chalcophile
- metal contents of the Merensky footwall from Rustenburg (drill hole PDL 117-3; Naldrett *et al.* 2009).
- Note the higher PGE concentrations and lower S, Ni, and Cu concentrations in the Boulder Bed, relative
- 795 to the bracketing rock units.
- 796 **Figure 4. A.** PPL scans of sections BBBC, BB, and BBTC, which sample the base, middle, and top of
- a boulder, respectively. **B.** Mg-Ca-Cr-log[K] SEM-EDS element map. **C.** Si-log[K]-P-S-Fe SEM-EDS
- 798 element map. Dashed lines outline the marginal zone (i.e., extent of intercumulus orthopyroxene) and
- 799 red arrows represent EPMA transects (see Figure 8; ESM 1). Abbreviations: ol = olivine, cpx =
- clinopyroxene, opx = orthopyroxene, pl = plagioclase, chr = chromite, sul = sulfides, ap = apatite, phl
- = phlogopite, and mag = magnetite.
- **Figure 5. A.** Photograph of sample BV-BB1 showing a pyroxenitic boulder and its marginal zone. **B.**
- 803 Cr-Fe-Ca XRF map of sample BV-BB1. C. Cr-Fe-Ni XRF map of sample BV-BB1. ol = olivine, opx
- = orthopyroxene, plg = plagioclase, chr = chromite, amp = amphibole, and cal = calcite.

- Figure 6. A. PPL scan of section Bou-2, which samples the base of a boulder. B. Mg-Ca-Cr-log[K]
- 806 SEM-EDS element map. C. Si-log[K]-P-S-Fe SEM-EDS element map. Dashed lines outline the
- 807 marginal zone (i.e., extent of intercumulus orthopyroxene) and red arrows represent EPMA transects
- 808 (see Figure 8; ESM 1). Abbreviations: ol = olivine, cpx = clinopyroxene, opx = orthopyroxene, pl =
- plagioclase, chr = chromite, sul = sulfides, ap = apatite, phl = phlogopite, amp = amphiboles, and mag
- 810 = magnetite.
- Figure 7. A. PPL scan of section TF-Bou, which samples the base of a boulder. B. Mg-Ca-Cr-log[K]
- 812 SEM-EDS element map. C. Si-log[K]-P-S-Fe SEM-EDS element map. Dashed lines outline the
- marginal zone (*i.e.*, extent of intercumulus orthopyroxene) and red arrows represent EPMA transects
- 814 (see Figure 8; ESM 1). Abbreviations: ol = olivine, cpx = clinopyroxene, opx = orthopyroxene, pl =
- plagioclase, chr = chromite, ap = apatite, phl = phlogopite, amp = amphiboles, sul = sulfides, and mag
- 816 = magnetite.
- Figure 8. Rim-core-rim EPMA transects of plagioclase crystals measured in the host anorthosite (A),
- 818 marginal zone (B), and within boulders (C). The profiles have been smoothed by polynomial
- approximation. The corresponding EPMA transects are annotated in Figures 4, 6, 7, and ESM Figure
- S2 as well as reported in ESM 1.
- **Figure 9.** Characteristics of plagioclase crystals at the base of a boulder in section TF-Bou (see Figure
- 7 for corresponding element maps). A-C. Maps and stacked histograms of equivalent circle diameter
- 823 (ECD; A), aspect ratio (B), and roundness (C) exported from AZtec Crystal software. **D.** Plagioclase
- 824 CSD plot and rose diagrams exported from CSDcorrections (Higgins 2002). Underlain are CSD profiles
- of plagioclase crystals from the Upper Critical Zone (Fig. 8 and sample 58 from Boorman *et al.* 2004¹)
- and from the JM reef package of the Stillwater Complex (Jenkins *et al.* 2022²). Additional plots display
- theoretical CSD profiles (Marsh 1988; Higgins 2002; Boorman *et al.* 2004).
- **Figure 10.** Lower hemisphere, equal-area pole figures of the [100], [010], and [001] axes of plagic lase
- crystals in the host anorthosite (A) and marginal zone (B). The pfJ, M-index, and J-index values together
- with the L# ($[100]e_1/e_2$) and F# ($[010]e_1/e_2$) are given for each grain group (see Section 3.2).
- **Figure 11. A.** Grain reference orientation deviation (GROD) angle maps of plagioclase, whereby pixels
- within a given grain are coloured by the difference in orientation angle relative to the grain average. **B.**

Grain orientation spread (GOS) map of plagioclase. Grains are coloured by the average misorientation angle of each pixel within a given grain relative to the average orientation of the same grain. **C.** GROD angle map of forsterite. Note the subgrain in the upper left as well as the downward percolative textures of olivine into cumulus plagioclase of the host anorthosite. **D.** GROD angle map of hypersthene. Note the high degrees of misorientation as indicated by the yellow colours. **E.** Mean orientation map of hypersthene where similar colours indicate similar orientations for the grains. One can observe regions where hypersthene crystals preserve similar orientations, which suggests that they are oikocrysts. **F.** Enhanced view of internal misorientation within two hypersthene oikocrysts. Note that the internal misorientation is relative to each exposed portion of the oikocryst itself.

Figure 12. Schematic diagram summarising the key features of the Boulder Bed. The image of the mottled anorthosite is from Maier *et al.* (2021).

Figure 13. A. Deposition of pyroxenitic cumulates atop (leuco)noritic cumulates during magma chamber replenishment. B. The influx of heat triggers partial melting of the (leuco)noritic floor rocks, which facilitates the disaggregation and sinking of the pyroxenitic cumulates. C. The upwelling (leuco)noritic partial melts react with the bases of boulders further promoting their disaggregation and rounding. This reaction causes: (i) incongruent dissolution of orthopyroxene to form unevolved anhedral olivine; (ii) coarsening of the boulder bases; (iii) plagioclase resorption and generation of reverse zoning; (iv) dissolution of base metal sulfides; (v) precipitation of chromite along the base and sides of boulders; and (vi) the precipitation of amphibole and other late-stage silicates in some cases. The upwelling of melts also explains the lack of comparable features on the upper margin of boulders.

D. Late-stage viscous compaction causes partial flattening of boulders and intra-crystalline deformation

Tables

of intercumulus pyroxenes.

- **Table 1.** Summary of analysed sections from the Boulder Bed.
- Table 2. Summary of the physical properties and fabric indices of plagioclase crystals from the host anorthosite and marginal zone of the Boulder Bed.