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Reply to discussion of 'Crustal fluid contamination in the Bushveld Complex, South Africa: an analogue for subduction zone fluid migration' by Roger Scoon and Andrew Mitchell (2020)

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ABSTRACT
In their discussion of our recent publication, Scoon and Mitchell (2020) put forward a number of arguments against the hydromagmatic model of Bushveld Complex formation that we present. Their criticisms of our model focus primarily on the formation mechanisms of the discordant bodies present at Bushveld, namely the iron-rich ultramafic pegmatites and the dunite pipes. While this was a minor portion of our paper, we here review evidence in favor of a fluid-related origin for these discordant bodies, in contrast to the primarily magmatic origin that Scoon and Mitchell present.

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In their discussion of our recent publication, Scoon and Mitchell (2020) (hereafter just Scoon and Mitchell) express a number of criticisms and reservations regarding some details of our model. We welcome the opportunity to discuss their concerns. Briefly, we suggested that extensive dehydration of the underlying country rock of the Bushveld Complex generated large volumes of volatile fluid that rapidly penetrated the lower section of the Bushveld stratigraphy, channelled both by rapid porous flow and local formation of diatreme pipes and that this fluid contamination can account for some isotopic anomalies of the Main Zone. These country fluids were locally channelled by synchronous diapir formation of the floor rocks. More broadly, we suggested that the Bushveld system is an excellent analogue for understanding fluid migration in subduction zones. We also suggested that our results had consequences for other discordant, pipe-like bodies and iron-rich ultramafic pegmatite (IRUP) formation models, but did not elaborate as it was outside the scope of the paper. The comment by Scoon and Mitchell allows us to follow up on this aspect of our work: indeed, we think this is yet another area where the Bushveld system can enlighten on mantle processes and vice-versa.

Scoon and Mitchell pointed out a number of minor errors in our stratigraphic section and some imprecise terminology which we welcome; other errata are noted at the end of this reply. Without much comment on the main points of the paper noted above, they instead put forward a number of critiques focused on our suggestion that our results have implications for the formation of other discordant bodies in the Bushveld Complex. We recognize that the origin of the dunite pipes and IRUP are controversial; for this reason, our discussion of these features focused primarily on the importance of a breccia pipe, located in the eastern limb of the Bushveld Complex, as the stronger evidence for large-scale fluid involvement. We called attention to its similarity with km-scale blowout pipes developed in sedimentary basins (recently, deep blowout pipes are now appearing in tundra regions of Siberia, presumably driven by climate change and accelerated methane production in the permafrost and illustrating that gas blowouts can occur on land as well; Chuvilin *et al.* 2020). In their discussion of our paper, Scoon and Mitchell agree that this breccia pipe could have acted as a pathway for country fluid into the Bushveld but disagree on its potential importance.

Scoon and Mitchell suggest that, if the Bushveld was affected by country fluid infiltration, that there should be evidence in the Lower Zone and Critical Zone. We note that, except for the Upper Zone, the mineral assemblages of the Bushveld complex do indeed show extensive evidence for isotopic disequilibrium (e.g., Mathez and Waight 2003; Prevec *et al.* 2005; Chutas *et al.* 2012; Roelofse and Ashwal 2012; Yang *et al.* 2013; Roelofse *et al.* 2015). A more important question is if the fluid can retain its crustal signature as it traverses the lower

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portions of the complex. Our model for fluid migration draws on a channelized flow, whereby the fluid is able to 'skip' over the Lower and Critical Zones and directly invade the Main Zone magma. This is most evident in the rapid flow required to form a 10 m diameter breccia pipe.

In regards to porous flow, Boudreau (2019) discussed two mechanisms that can limit isotopic equilibration with the country fluids in the lower, cooler sections of the Bushveld Complex. First, grain-scale interaction of fluids based on isotopic re-equilibration is on the order of 10^3–10^5 years for amphibolite-granulite grade metamorphic rocks (e.g., Van Haren *et al*. 1996; Graham *et al*. 1998), the longer times approaching the cooling time for the Bushveld complex (e.g., Cawthorn and Webb 2013). Le Roux *et al*. (2009) summarized slow diffusion rates in mantle minerals that imply that isotopic equilibration may not be reached during partial melting at hot mantle temperature and relatively slow melting rate of an isotopically heterogeneous assemblage. Second, some isotopes will not have high bulk rock abundances of the element in question to affect the isotopic character of the infiltrating fluid, particularly if the fluid is otherwise in chemical equilibrium with the host minerals. For example, both Sr and Nd are strongly incompatible in olivine and low-Ca pyroxene. Hence, the ultramafic rocks that make up the lower portions of the Bushveld and Stillwater complexes do not present a large reservoir of Sr and Nd to exchange with a rapidly moving fluid, a point made in our paper. As in a subduction system, it does appear that crustal fluids can traverse thick sections of hot ultramafic Bushveld rock and still retain a crustal isotopic signature. The observation that IRUP is found mainly above the Lower Critical Zone is consistent with the infiltrating fluids not becoming particularity reactive until they encountered hotter rocks above the Lower Zone.

Another implication noted in our paper is that the vertical pipe-like structures will channel both country fluid as it flows upward and also pulls in igneous fluids by horizontal flow component from the solidifying Bushveld host use the same channels to migrate out of the crystal pile. This has implications for ore components that some pipes can contain, and can complicate the isotopic signature of the pipes as the two fluid sources mix and react in the fluid channels (see below).

Scoon and Mitchel also disagree with our initial magma thickness in our summary model, suggesting it is too thick. In our Figure 11, we had combined our modelled fluid migration results with that of Cawthorn and Webb (2013) for the crystallization history of the Bushveld magma and Gerya *et al*. (2003) for the footwall diapir formation. In terms of the thermal history of the complex, we note that crystallization of a sill is largely controlled by heat loss out the top by direct contact of the magma with the roof rock. In regards to how fast the crystal pile grows, the thickness of the magma chamber is not important as long as some magma is always present, either as a single fractionating thick sill or a continuously replenished inflating sill undergoing mixing and fractional crystallization. Another minor point could be made of the original Cawthorn and Webb model. It is not clear how one can form an 8 km thick intrusion that initially intruded only 2 km below the surface without forming a large blister on the surface of the earth. The shallow intrusion model of Cawthorn and Webb also leads to perhaps a too short cooling time and it does not reproduce the hotter footwall country-rock geotherms of Harris *et al*. (2003). These problems could be minimized by placing the intrusion deeper and are the focus of ongoing study.

Iron-rich ultramafic pegmatite (IRUP) formation

As for the IRUP and dunite pipe formation, there is debate if they represent a distinctive process as Scoon and Mitchell suggest or if the IRUP-dunite represent a continuum of replacement by a reactive fluid/melt or a fractionating intrusive magma crystallizing in eroded melt-filled channels. Strong evidence for replacement is seen in the relic chromitite layers in the host rock that continue through the dunite pipes (Scoon and Mitchell's Figure 2).

In regard to IRUP, there have been a number of models and contrasting interpretations. As noted by Scoon and Mitchell, the IRUP appears to have replaced the original lithology (ignoring those examples that may have a fault or fracture control); the debate is if this is a volume for volume metasomatic replacement or if the host rock was first removed (physically or by melting) prior to crystallization of a distinctive IRUP magma. In mantle sections of ophiolites, discordant bodies are common and are generally viewed as important for rapid flow of mantle partial melts. Most workers interpret these bodies as metasomatic replacement bodies that involve a focused reactive flow (e.g., Kelemen *et al*. 1995). For the Bushveld metasomatic replacement favoured by Scoon and Mitchell, they suggest that the metasomatic agent was an Fe-rich silicate liquid derived from the Upper Zone that percolated downward through the Main Zone and into the Critical Zone, based on the Fe-rich mineralogy and Sr isotopic similarity between the two. Others have suggested that the metasomatic agent was a Fe-, PGE- and Cl-rich fluid (e.g., Schiffries 1982; Boudreau 2019). In contrast, others have suggested that the IRUP formed by the intrusion of a mantle magma that first removed portions of the host rock before crystallizing (e.g., Cawthorn *et al*. 2000, 2018; Günther *et al*. 2018).

In regard to the Scoon and Mitchell interpretation, we note the following counter evidence for Upper Zone liquid involvement in IRUP formation, at least for those in the Critical Zone. Besides Fe-rich minerals, the Upper Zone magmas are also saturated in plagioclase and, in the more evolved liquids, apatite. As Scoon & Mitchell note, the IRUPs do not contain significant plagioclase and bulk rock IRUP compositions reported by Cawthorn *et al*. (2018) contain <0.05 wt. % P_2O_5 with one notable exception. As noted by Cawthorn *et al*. (2018) and Boudreau (2019) the IRUP clinopyroxene is enriched in the wollastonite (wo) component and lack exsolution lamellae as compared with typical Bushveld host rock clinopyroxene of similar mg#. This is again inconsistent with the clinopyroxene having precipitated from a UZ magma but it is consistent with a lower temperature (secondary) origin.

A point generally ignored is that the hydrous mineral assemblage of the Upper Zone rocks contains very little Cl, in contrast to the Lower Zone/Critical Zone rocks for which these minerals, particularly the apatite, are unusually Cl-rich as compared with other layered intrusions (Willmore *et al*. 2000). Although extensive study of the hydrous minerals of the IRUP and dunite pipes is lacking, Schiffries (1982) reports that they are also Cl-enriched. This is particularly important, as high-temperature Cl-rich fluids will be solute-rich, particularly for Fe (e.g., Yardley 2005), with obvious implications for the formation of Fe-rich pegmatite and Fe-rich hortonolite pipes. Finally, as is evident in their name, IRUPs have a very coarse-grained to pegmatoidal texture whereas the UZ rocks are not pegmatoidal. This and the abundance of biotite and amphibole suggests that they grew in a fluid-saturated system. In short, a connection between the IRUP and the Upper Zone is, in many ways, difficult to justify.

A broader point that Scoon and Mitchell and some other studies overlook is that the IRUPs tend to be associated with leucocratic to anorthositic host rocks (see Figure 1 of the Scoon and Mitchell comment, also Cawthorn *et al*. 2018). Analogous ultramafic/anorthosite segregations occur in other intrusions as well. For example, beneath the Basistoppen sill in the Skaergaard Complex, Naslund (1986) noted podiform anorthosite-ultramafic segregations thought to be caused by the sill's intrusion into the hot Skaergaard ferrogabbros. Similar segregations are also found in the Lower Zone and Marginal Border Series of Skaergaard, where there is no source of reheating. Sonnenthal (1992) suggested these segregations were due to reaction with a Cl-rich fluid, although they did not present details of the mechanism involved. Discordant troctolite-anorthositic bodies in the Stillwater Complex have also been attributed to volatile fluids becoming undersaturated in the mafic component as they rise into hotter rocks and remove the pyroxene component by incongruent reaction, producing olivine as an intermediate reaction product (Meurer *et al*. 1997). Applied to the IRUP problem, the model of Boudreau (2019) simply suggests that, while the scale is different in the Bushveld case, the IRUP is simply the mafic component lost from the host anorthosite.

While the above look at the role of fluids, it is also possible that fluid interaction can be mediated by melt produced by hydration melting of the host rock. For example, Keller and Katz (2016) and Keller *et al*. (2017) summarize the ways in which volatiles are important in lowering the mantle solidus and promoting channelled, pipe-like transport of mantle melts, despite the low concentration of volatiles in the mantle. In regards to the Bushveld system, Cawthorn *et al*. (2018) have suggested that IRUP mineral assemblages crystallized from a fractionating magma that has affinities with volatile-rich meimechite or ankaramite mantle magmas that are characterized by relatively high concentrations of alkalis, the REE and high CaO/Al_2O_3 wt. ratios. Green *et al*. (2004) have suggested that ankaramite liquids form by flux melting of a refractory lherzolitic mantle. Similarly, a reasonable hypothesis is that a Cl-rich volatile fluid containing significant Ca, alkalis, and REE could induce partial melting as it moves into hotter Bushveld rocks and produces a meimechite- or ankaramite-like liquid. This would explain the paucity of evidence for the external derivation of these magmas in the underlying country rock, but explain the local occurrence of what appear to be examples of magmatic intrusion within the Bushveld. The local formation of a volatile-rich hydration melt would explain the coarse texture of the IRUPs and the volatile- and Ca-enriched liquid would favour the crystallization of olivine and clinopyroxene over orthopyroxene and plagioclase. As noted by Boudreau (2019), 'Why go into the mantle when you can make everything in your back yard?'

Dunite pipes

We appreciate the detailed work Scoon and Mitchell have done to outline the orthomagmatic model of dunite pipe formation. As referenced in Benson *et al*. (2020), Viljoen and Scoon (1985) noted that the dunite pipes of the eastern Bushveld are commonly located adjacent to downwarped igneous layering in the host rock. Regionally, they are commonly found in the vicinity of faults and structural upwarps in the floor rock (our Figure 3). We noted that the latter observation was consistent with a localized origin for these pipe structures (this correlation would not be expected for mantle magmas that could pop through anywhere). Scoon and Mitchell note that IRUPs and other discordant bodies are not always associated with upwarps in the floor rocks. However, our numerical model shows that channelized, pipe-like porous flow can develop spontaneously from minor porosity perturbations even when the contact with the underlying country rock is horizontal. The presence of floor diapirs only enhances fluid focusing of both country and Bushveld fluids and explains why the more impressive examples (such as the dunite pipe Scoon and Mitchell show as their Figure 2) are seen in the eastern Bushveld where floor rock diapirs are best developed.

Günther *et al*. (2018) suggest that the oxygen isotopes of the dunite pipes are indicative of a recycled crustal component in a mantle magma, but again this could be introduced by fluids from the underlying country rock. Roger and Scoon also discredit the work of Schiffries (1982), who proposed a high-temperature chloride solution could cause a number of metasomatic reactions. Thus, olivine can be produced via the incongruent reaction of orthopyroxene with a fluid that becomes progressively silica-undersaturated as it moves into hotter rocks:

$$\underset{orthopyroxene}{2(Mg,Fe)SiO_3} \rightarrow \underset{olivine}{(Mg,Fe)_2SiO_4} + \underset{fluid}{SiO_{2,aq}} \quad (1)$$

Experimental work by Newton and Manning (2000) suggests that modest amounts of NaCl in a hydrous fluid ($X_{NaCl} \cong 0.1$) can cause a roughly 50% increase in SiO_2 solubility compared to NaCl-absent aqueous fluids at 700° C and 0.2 GPa. This and other fluid-driven reactions can produce olivine, loss of plagioclase and introduction of the PGE. This reaction would be accompanied by a 67% volume loss, which can create the collapse structures around the Driekop pipe noted by Scoon and Mitchell. They use the presence of Cr-spinel in the pipes as an argument against the hydrothermal model. However, as noted in Boudreau (2019), pyroxenes can contain significant Cr that cannot be accommodated by the secondary olivine and reaction shown in Equation 1 can be generalized to produce the Cr-spinel component of the pipes:

$$\underset{orthopyroxene}{\left(Mg^{2+},Fe^{2+},Cr^{3+}\right)\left(Si^{4+},Cr^{3+}\right)O_3} \rightarrow \underset{olivine}{(Mg,Fe)_2SiO_4} + \underset{Cr\text{-}spinel}{(Mg,Fe)(Cr)_2O_4} + \underset{fluid}{SiO_{2,aq}} \quad (2)$$

In regard to the PGE-rich cores seen in some of these pipes, Peyerl (1982) noted that the platinum-group minerals of the UG2 chromitite have been affected by the crosscutting PGE-bearing Driekop pipe out to a distance of 1–2 km, with the PGE-base metal sulphide assemblage replaced by PtFe alloy and various Pd-Pt-Ab-Sb minerals towards the pipe. Peyerl suggested this change was driven by fluids moving *outward* from the pipe and removing sulphur. The results of our numeric model show that the development of vertical channelled fluid flow induces a lateral flow of fluid in the host rock *towards* the regions of the channelized porous flow. In detail, as a porosity wave moves up, at the top of the wave fluid is pumped into the matrix on a small scale relative to the compaction length. This fluid, and any other background fluid present, is then drawn back in at the bottom of the wave on a scale comparable to the compaction length. Excluding the chromitites, the Lower Critical Zone and Upper Critical Zone rocks, average 40 and 119 ppb Pt, respectively (summary of Barnes and Maier 2002). Assuming that as little as 5 ppb Pt was, on average, scavenged from a 1 to 2 km diameter region in the host rock and concentrated into a 10 m pipe core, the core would contain from 50 to 200 ppm Pt, well above what is seen in the PGE-bearing reefs (Boudreau 2019).

Conclusions

Scoon and Mitchell present a number of criticisms of our paper (Benson et al., 2020). These critiques are primarily focused on our presentation of Bushveld discordant bodies, particularly IRUP and dunite pipes, as hydromagmatic in nature; they prefer an orthomagmatic model for the formation of these discordant bodies. We reiterate that the discussion of discordant bodies and their relationship to large-scale Bushveld fluid flow was a minor line of evidence for this hypothesis. The bulk of our paper revolved around the role of fluids as a source of contamination and called on less controversial structures such as the breccia pipe as evidence of rapid fluid transport. This idea of country fluid mixing into the Bushveld magma (and driving chromitite formation) also has been recently suggested by Veksler and Hou (2020). As we briefly attempted to suggest here, the origin of the IRUP and dunite pipes is not so clear-cut and neatly orthomagmatic as Scoon and Mitchell (2020) describe. They overlook a number of studies which present evidence contradictory to their findings and which support a hydromagmatic model for the formation of these bodies, and indeed discount evidence from other igneous and mantle settings where similar features have been attributed to reactive fluid flow. While one can appreciate the conservatism in interpretation that they espouse, we believe layered intrusions present a much richer variety of physical and chemical processes than conventionally proposed.

Errata in original text

In the discussion of assumptions made regarding the rheology in the model of dehydration of the footwall, a reference was inadvertently attributed to Schiffries and Rye (1990). It should have been attributed to Paterson and Luan (1990).

In the final paragraph of page 11, there is a sentence that reads, 'This has been suggested to be the result of alterations to Pb isotopic composition around the time of crystallization, but after certain minerals (such as plagioclase) had closed to isotopic alteration.' This sentence should instead read, 'This has been suggested to be the result of alterations to Pb isotopic composition of some minerals (such as sulphides) around the time of crystallization, but after other minerals (such as plagioclase) had closed to isotopic alteration.'

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