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Zinc isotope evidence for volatile fluid remelting to form PGE-enriched reefs in layered intrusions

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Abstract

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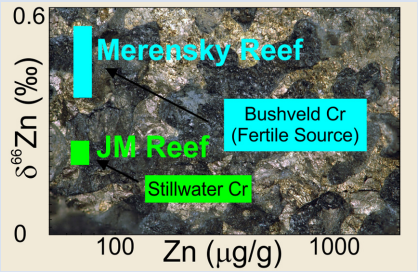

The origin of platinum group element (PGE) enriched zones in layered intrusions, and whether they require magmatic fluids to form them, is debated. Here we report Zn abundance and isotope data for chromitite seams from the 1.27 Ga Muskox, 2.05 Ga Bushveld and 2.7 Ga Stillwater layered igneous complexes, and for the PGE-enriched Merensky (Bushveld) and JM Reefs (Stillwater) and their mineral components. The >0.7 ‰ variability in δ^{66}Zn between mineral phases for both PGE-enriched reefs support their origin through volatile fluid remelting. The Merensky and JM Reefs are ~0.1 ‰ heavier than underlying chromitite seams due to Rayleigh-type distillation of Zn isotopes during igneous crystallisation. Zinc-rich chromitite seams lower in the magmatic sequences are likely to be faithful representations of the ultramafic parental melts to the intrusions. The Muskox and Bushveld intrusions originated from partial melting of fertile mantle sources, similar to the sources of plume volcanism today. In contrast, the low initial δ^{66}Zn of the Stillwater chromitite seams requires a boninite-type parental melt, suggesting fluid assisted melting processes were important in its formation 2.7 billion years ago.

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Introduction

The platinum group elements (PGE: Os, Ir, Rh, Ru, Pt, Pd) play a crucial role in many modern technologies, with more than two thirds of the world's supply sourced from PGE-enriched layers within the Bushveld and Stillwater layered intrusive complexes (*e.g.*, Mudd *et al.*, 2018). The origin of PGE mineralisation in layered intrusions remains contentious however, with models that assume formation purely through magmatic processes (*e.g.*, Eales and Cawthorn, 1996; Godel and Barnes, 2008) and those that invoke volatile fluid remelting and mineral reprecipitation to form the sulfide-rich mineralised zones (*e.g.*, Boudreau, 1999, 2008, 2016, 2026; Gupta and Boudreau, 2025). Similar controversies also exist for the potential mantle sources of partial melts feeding the layered intrusions, which have important potential implications for the tenor of PGE enrichment.

In this study, we employ Zn isotopes as a tool to explore the origin of PGE enrichment in layered intrusive complexes. Zinc is composed of five stable isotopes and Zn isotope composition is usually reported in delta notation, which for ^{66}Zn/^{64}Zn is:

$$\delta^{66}\mathrm{Zn} = \{(^{66}\mathrm{Zn}/^{64}\mathrm{Zn}_{\mathrm{sample}})/(^{66}\mathrm{Zn}/^{64}\mathrm{Zn}_{\mathrm{JMC\text{-}Lyon\ standard}}) - 1\} \times 1000$$

It has previously been demonstrated that Zn exhibits limited (typically <0.3 ‰) isotope fractionation during partial melting, magmatic crystallisation and magmatic ore processes (*e.g.*, Day *et al.*, 2022; Wilkinson, 2023). In contrast, low temperature and hydrothermal processes can lead to >1 ‰ variations in δ^{66}Zn (Moynier *et al.*, 2017; Wilkinson, 2023). To date, limited isotopic studies of Zn have been done for large mafic-ultramafic intrusive complexes. The most extensive study has been for the Sudbury Igneous Complex whose origin from a bolide impact indicates that mechanisms of evaporative loss of Zn were important (Kamber and Schoenberg, 2020). In this study we examine layered intrusive complexes formed through endogenous terrestrial magmatic processes. We examined Zn isotope variations in chromitite and PGE-enriched 'reefs' from the 1.27 Ga Muskox Intrusion (Canada), 2.05 Ga Bushveld Intrusion (South Africa) and 2.7 Ga Stillwater Intrusion (Montana) to further understand their formation.

Methods and Results

The methods for the separation and analysis of Zn abundances and isotopic composition are adapted from van Kooten and Moynier (2019) and are described in the Supplementary Information. In this study, three of the world's largest layered intrusive complexes were examined (Table S-1). Bulk rocks from three PGE-rich chromitite seams were studied from the Mesoproterozoic (1.27 Ga) Muskox Intrusion (Day and O'Driscoll, 2019). An andesite glass flow (CM19) from the Coppermine Continental Flood Basalt (CFB) and associated with chromitite seam formation in the Muskox Intrusion (Day *et al.*, 2013), was also analysed. Four bulk rock PGE-rich chromitite seams and bulk rock and mineral fractions from the PGE

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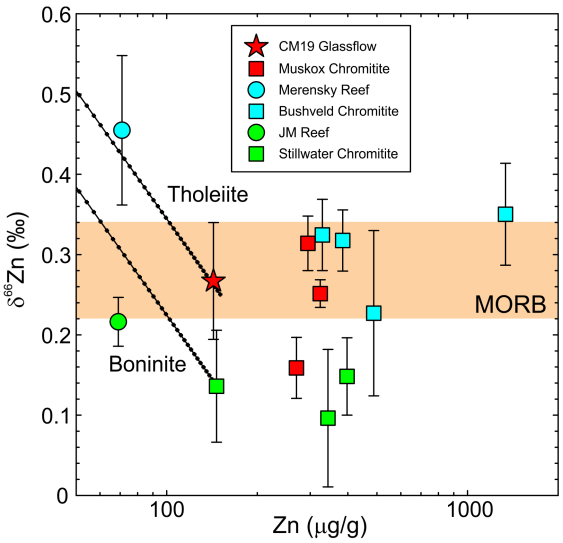

Figure 1 Zinc isotopic composition *versus* Zn abundance for bulk rock samples of PGE-enriched reefs, chromitite seams and the CM19 glass flow. MORB estimate of 0.28 ± 0.06 ‰ from Day *et al.* (2022). Shown are two Rayliegh distillation models, assuming the same initial Zn composition of melts (150 μg/g), one with a boninite-like source with low δ^{66}Zn (0.13; Boninite) and the other with a tholeiite source with higher δ^{66}Zn (0.25; Tholeiite) and assuming removal of Zn through spinel fractionation (kD = ~7) in increments of 0.3 % (dots on model curves).

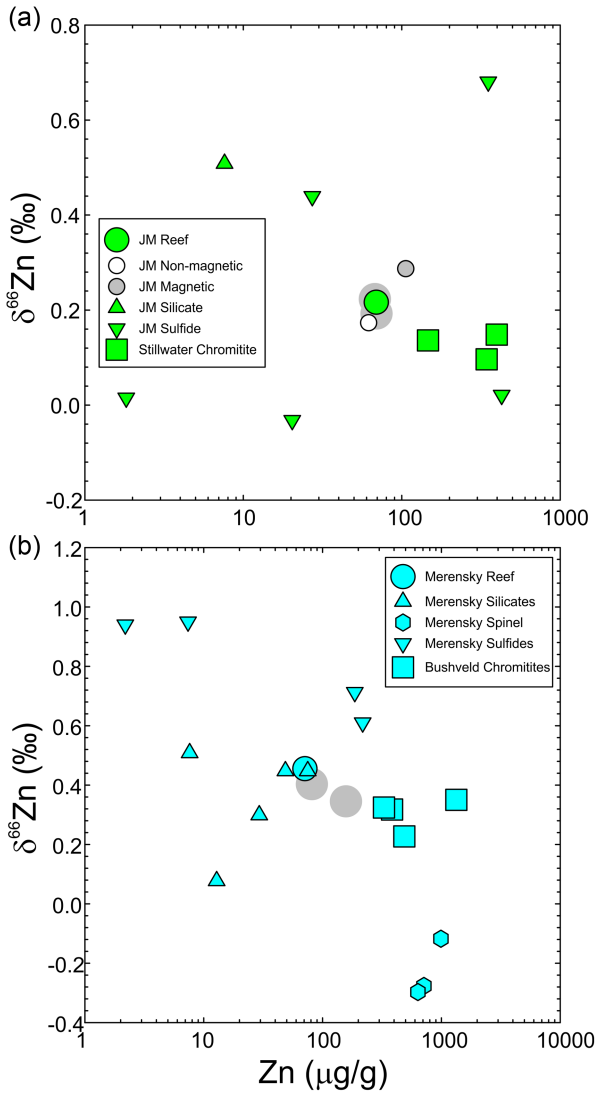

Figure 2 Zinc abundance *versus* Zn isotopic composition for **(a)** Stillwater chromitites, the JM Reef and JM Reef components and **(b)** Bushveld chromitites, the Merensky Reef and Merensky Reef components. Large grey circles denote modally recombined data using components of both reefs (Table S-2).

mineralised pegmatoid Merensky Reef of the Paleoproterozoic (2.05 Ga) Rustenberg Layered Igneous Complex in the Bushveld Intrusion were examined. Finally, three bulk rock PGE-rich chromitite seams and bulk rock and mineral fractions from the PGE mineralised Johns-Manville (JM) Reef in the Olivine-Bearing zone I of the Archean (2.7 Ga) Stillwater Igneous Complex (Day and O'Driscoll, 2019) were studied.

Zinc isotope variations follow mass dependent behaviour (Fig. S-1) and hereafter only δ^{66}Zn values are discussed. Chromitite seams from the layered intrusive complexes have broadly similar Zn contents (100's up to 1000 μg/g), with δ^{66}Zn between 0.16 and 0.31 ‰ for the Muskox Intrusion, 0.23 to 0.35 ‰ for the Bushveld Intrusion and 0.10 to 0.15 ‰ for the Stillwater Intrusion (Fig. 1). The CM19 glass flow from the Coppermine CFB, which is coeval with Muskox Intrusion, has a δ^{66}Zn value of 0.27 ‰, similar to Muskox chromitite seams. The Merensky and JM Reef samples have similar Zn abundances (~70 μg/g), with the Merensky Reef (0.45 ‰) having higher δ^{66}Zn than the JM Reef (0.22 ‰).

Mineral components in the Merensky Reef have lower δ^{66}Zn values in the order of: sulfide (pentlandite, pyrrhotite, chalcopyrite Zn = 2 to 218 μg/g; δ^{66}Zn = 0.61 to 0.95 ‰) > silicate (Zn = 8 to 75 μg/g; δ^{66}Zn = 0.08 to 0.51 ‰) > spinel (Zn = 635 to 991 μg/g; δ^{66}Zn = −0.12 to −0.30 ‰), with modal recombination using a range of measured modal percentages (Table S-2) for the Merensky Reef being within uncertainty of the bulk rock value (Fig. 2). The chalcopyrite grains have the highest Zn of all the sulfides and are ~0.3 ‰ lighter than pentlandite and pyrrhotite, consistent with equilibrium partitioning (Mason *et al.*, 2005). A single silicate mineral phase, plagioclase, was analysed from the JM Reef (7.6 μg/g; δ^{66}Zn = 0.51 ‰), and pentlandite, pyrrhotite and chalcopyrite ranged from 2 to 427 μg/g Zn with δ^{66}Zn from −0.03 to 0.68 ‰. A JM Reef aliquot was separated after crushing using a magnet, revealing that the magnetic portions (106 μg/g; δ^{66}Zn = 0.29 ‰) had higher Zn concentrations with heavier isotopic values than the non-magnetic fraction (62 μg/g; δ^{66}Zn = 0.17 ‰). Modal recombination using the silicate and sulfide or magnetic and non-magnetic fractions give results that are within uncertainty of the measured bulk rock value (Table S-2, Fig. 2).

Discussion

Evidence for volatile-induced remelting in the Merensky and JM Reefs. Mineral separate Zn isotope data for the Merensky Reef demonstrate a ~1 ‰ variation between δ^{66}Zn in spinel *versus* sulfides (Fig. 2). The ionic radius of Zn^{2+} (0.74 Å) means that it is considered to substitute for Fe^{2+} (0.78 Å) (Wang *et al.*, 2017). During purely magmatic processes and formation of the Merensky Reef as a cumulate layer (*e.g.*, Eales and Cawthorn, 1996), the typical crystallisation order would be spinel > silicate > sulfides. Merensky Reef spinel grains have nearly 20× and 7× higher Zn abundances than the silicate and sulfide grains, respectively, with $\Delta^{66}Zn_{Spl\text{-}Sil} = -0.6 \pm 0.2$ ‰ and $\Delta^{66}Zn_{Spl\text{-}Sul} = -1 \pm 0.2$ ‰. These mineral-mineral differences contrast the higher δ^{66}Zn in mantle peridotite spinel relative to silicates

and are associated with stiffer Zn-O bonds in spinel than silicate minerals (Wang *et al.*, 2017). They are also inconsistent with evidence that high temperature igneous processes are incapable of producing fractionations of δ^{66}Zn greater than about 0.2 ‰ in ore deposits (*e.g.*, Wilkinson, 2023). Zinc isotope data for Merensky Reef minerals therefore do not support its origin from purely magmatic processes.

Field observations (Nicholson and Mathez, 1991), isotopic data (Reid *et al.*, 1993; Willmore *et al.*, 2002) and modelling (Boudreau, 2008) have indicated that the Merensky Reef may be a zone of volatile-induced remelting, leading to reprecipitation of sulfide and spinel. These models are similar to those developed for the Stillwater JM Reef (see below). Hydrothermal processes have been shown to produce variations in sulfide δ^{66}Zn by up to 0.9 ‰ (*e.g.*, Mason *et al.*, 2005; John *et al.*, 2008). The causes of such fractionations are not well understood, although Rayleigh-type kinetic fractionation appears to be most favoured (Wilkinson, 2023). For the Merensky Reef, this would mean early precipitation of spinel and preferential enrichment in the lighter isotopes of Zn, followed by later precipitation of sulfide phases. These results suggest that temperature and mineral-melt chemistry may be as an important factor as coordination number and Zn isotope fractionation in spinel.

For the JM Reef, there is a ~0.7 ‰ variation in δ^{66}Zn for measured components (Fig. 2). While strictly magmatic models have been proposed for its origin (*e.g.*, Barnes and Naldrett, 1985; Godel and Barnes, 2008), the large range in Zn isotopes implies a purely magmatic process may be unlikely. Models of Cl-rich volatile fluid percolation have been well developed for the formation of the JM Reef (Boudreau, 1999, 2016), supported by Cl isotope data (Boudreau *et al.*, 1997), and involve incongruent melting of the pre-existing partially molten crystal pile. In the case of both the PGE-rich Merensky and JM Reefs, there are large Zn isotope fractionations exceeding those observed from magmatic effects (Day *et al.*, 2022; Wilkinson, 2023). The Zn isotope variations, while complex and warranting further investigation, support reprecipitation models for PGE-bearing sulfides in both reefs. While such processes have led to significant Zn isotope distribution, modal recombination of the individual mineral components reproduces the bulk rock compositions well.

Constraining mantle sources of the Muskox, Bushveld and Stillwater intrusions. A major goal in the research of layered igneous complexes that contain economically important PGE resources is to understand potential mantle sources. Chromitite horizons in the Muskox, Bushveld and Stillwater layered igneous complexes are associated with some of the most mafic or ultramafic melts (Eales and Cawthorn, 1996, Day *et al.*, 2008; Boudreau, 2016), and have high Zn contents, so are likely to be faithful representations of the Zn isotopes of mantle sources. In the case of the Bushveld and Stillwater chromitite horizons, these lie lower in the magmatic sequence than the PGE-enriched reefs (Peridotite zone and lower Critical Zone, respectively). A fertile plume-like mantle source has been suggested for the Muskox Intrusion and associated lower Coppermine CFB (*e.g.*, Griselin *et al.*, 1997; Day *et al.*, 2008, 2013). This is consistent with the δ^{66}Zn data for the CM19 glass flow (0.27 ‰) and the weighted average of Muskox chromitites (0.24 ‰), which are within the range of modern OIB or MORB compositions (Fig. 3). The CM19 glass flow has been interpreted as the extrusive manifestation of chromitite formation in the Muskox Intrusion (Day *et al.*, 2013) and the Zn isotope similarities remain consistent with such a hypothesis.

The Bushveld chromitite seams have a weighted average of 0.32 ‰, within the range of modern OIB or MORB compositions (Fig. 3). Notably, the chromitites in the Bushveld are

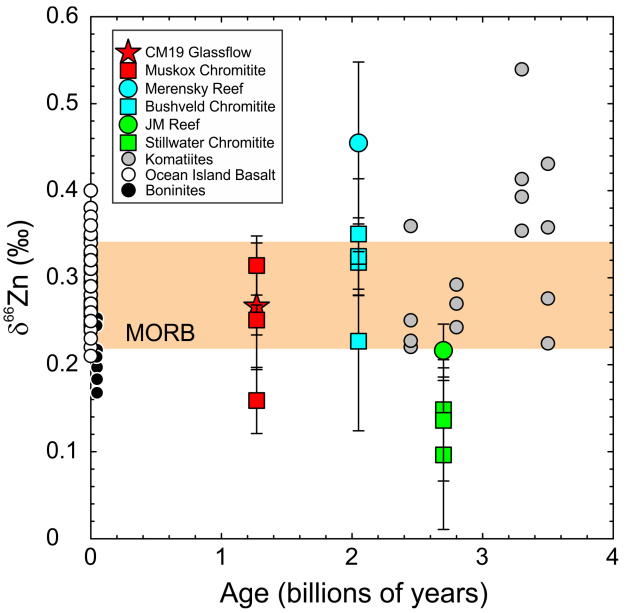

Figure 3 Zinc isotopic composition as a function of time for PGE-rich reefs and chromitite seams from the Stillwater, Bushveld and Muskox layered mafic intrusions, and the Coppermine CFB CM19 glass flow, bulk rock data for komatiites, ocean island basalts and boninites *versus* the MORB average (summarised in Day *et al.*, 2022).

isotopically lighter with respect to Zn than the Merensky Reef ($\Delta^{66}Zn_{chromitite\text{-}reef} = -0.1$ ‰). The chromitites are stratigraphically lower than the Merensky Reef, suggesting isotopic fractionation of Zn to heavier values towards the top of the Main Zone of the Rustenberg Layered Series, with no evidence of isotopic fractionation in the chromitites themselves. A similar scenario exists for the Stillwater chromitites, which are stratigraphically lower in the Ultramafic Series relative to the JM Reef, which is in the Lower Banded Series (*e.g.*, Boudreau, 2016), yet these show a similar sense of fractionation ($\Delta^{66}Zn_{chromitite\text{-}reef} = -0.1$ ‰). We suggest that these results point to similar formation processes whereby Rayleigh distillation type fractionation of Zn during cumulate crystallisation from parental type ultramafic melts, represented by the chromitites, followed by volatile fluid reprecipitation in the upper portions of the sequences, led to the formation of the PGE-rich reefs. In Figure 1, examples of Zn isotope fractionation during cumulate crystallisation of Cr-spinel are shown, accentuated by the isotopically light Zn composition of the spinel, yet the strong affinity for Zn into the spinel structure. This further emphasises the importance of spinel in hosting Zn in layered intrusive complexes.

A major distinction between the Stillwater chromitites, and those from the Muskox or Bushveld intrusive complexes is that the weighted average of δ^{66}Zn is anomalously low (0.13 ‰). No OIB, MORB or komatiite type melts have been measured with such low δ^{66}Zn (Fig. 3). The only modern mafic-ultramafic melts for which low δ^{66}Zn have been recorded to date are boninites (Day *et al.*, 2022). Boninite-type parental magmas have been widely invoked for the Stillwater parental magma (*e.g.*, McCallum, 1996; Boudreau, 2016) and the presented Zn isotope data for the chromitites may provide some of the strongest evidence yet for a depleted mantle source where fluid-assisted type melting was required to induce melting to produce the Stillwater igneous complex. Stillwater chromitites are characterised by lower PGE abundances and lower Pt/Pd than either Muskox or Bushveld chromitites (Naldrett *et al.*, 2009; Day and

O'Driscoll, 2019). Given that melt-depleted peridotites are typically strongly depleted in both Pd and Pt (*e.g.*, Lin *et al.*, 2024), the mantle source of the Stillwater Igneous Complex may have been metasomatically re-enriched in Pd and the rare earth elements to explain its composition.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2527.

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