

Evolution of ultrapotassic volcanism on the Kaapvaal craton: deepening the orangeite versus lamproite debate



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Abstract: Orangeites are a significant source of diamonds, yet ambiguity surrounds their status among groups of mantle-derived potassic rocks. This study reports mineralogical and geochemical data for a c. 140 Ma orangeite dyke swarm that intersects the Bushveld Complex on the Kaapvaal craton in South Africa. The dykes comprise distinctive petrographic varieties that are linked principally by olivine fractionation, with the most evolved members containing minor amounts of primary carbonate, sanidine and andradite garnet in the groundmass. Although abundant groundmass phlogopite and clinopyroxene have compositions that are similar to those of cratonic lamproites, these phases show notable Ti-depletion, which we consider a hallmark feature of type orangeites from the Kaapvaal craton. Ti-depletion is also characteristic of bulk rock compositions and is associated with strongly depleted Th–U–Nb–Ta contents at high Cs–Rb–Ba–K concentrations. The resultant high large ion lithophile element/high field strength element ratios of orangeites suggest that mantle source enrichment occurred by metasomatic processes in the proximity of ancient subduction zones. The Bushveld-intersecting orangeite dykes have strongly enriched Sr–Nd–Hf isotopic compositions (initial $^{87}\text{Sr}/^{86}\text{Sr} = 0.70701\text{--}0.70741$; $\epsilon\text{Nd} = -10.6$ to -5.8 ; $\epsilon\text{Hf} = -14.4$ to -2.5), similar to those of other orangeites from across South Africa. Combined with the strong Ti–Nb–Ta depletion, this ubiquitous isotopic feature points to the involvement of ancient metasomatized mantle lithosphere in the origin of Kaapvaal craton orangeites, where K-rich metasomes imparted a ‘fossil’ subduction geochemical signature. Previous geochronology studies identified ancient K-enrichment events within the Kaapvaal cratonic mantle lithosphere, possibly associated with collisional tectonics during the 1.2–1.1 Ga Namaqua–Natal orogeny of the Rodinia supercontinent cycle. It therefore seems permissible that the cratonic mantle root was preconditioned for ultrapotassic magma production by tectonomagmatic events that occurred along convergent plate margins during the Proterozoic. However, reactivation of the K-rich metasomes had to await establishment of an extensional tectonic regime, such as that during the Mesozoic breakup of Gondwana, which was accompanied by widespread (1000 × 750 km) small-volume orangeite volcanism between 200 and 110 Ma. Although similarities exist between orangeites and lamproites, these and other potassic rocks are sufficiently distinct in their compositions such that different magma formation processes must be considered. In addition to new investigations of the geodynamic triggers of K-rich ultramafic magmatism, future research should more stringently evaluate the relative roles of redox effects and volatile components such as H₂O–CO₂–F in the petrogeneses of these potentially diamondiferous alkaline rocks.

Supplementary material: Mineral and bulk rock compositions including Sr–Nd–Hf isotope ratios are available at <https://doi.org/10.6084/m9.figshare.c.5440652>

The Kaapvaal craton in southern Africa is host to numerous occurrences of kimberlite, which traditionally have been divided into two groups based on petrographic characteristics and compositions: the so-called (1) archetypal Group 1 kimberlites and (2) highly micaceous Group 2 kimberlites, also known as ‘orangeites’ (Wagner 1914; Smith *et al.* 1985; Skinner 1989; Mitchell 1995). Group 1

kimberlite magmatism of the Kaapvaal craton and surrounding mobile belts has occurred repeatedly since at least 1800 Ma, with prominent eruption episodes at around 1150, 520, 240 and 90 Ma (Jelsma *et al.* 2009; Griffin *et al.* 2014; Tappe *et al.* 2018). Archetypal Group 1 kimberlites are both petrographically and geochemically relatively homogeneous, and their moderately depleted to slightly enriched

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Sr–Nd–Hf isotopic compositions suggest an origin from the convecting upper mantle (Smith 1983; Nowell *et al.* 2004; Tappe *et al.* 2020a, b). A convecting upper mantle origin is also consistent with the lack of anomalous ^{182}W abundances in these CO_2 – H_2O -rich ultramafic magmas (Tappe *et al.* 2020b), which rules out significant melt contributions from lower mantle plumes to kimberlite magmatism, in contrast to many oceanic island basalt suites (Rizo *et al.* 2019; Mundl-Petermeier *et al.* 2020). The overwhelming majority of superlative diamond deposits are hosted by Group 1 kimberlites, not only across southern Africa but also on other cratons worldwide (Janse and Sheahan 1995; de Wit *et al.* 2016).

In contrast, Group 2 kimberlite *or* orangeite magmatism on the Kaapvaal craton and adjoining mobile belts appears to be temporally restricted to the Mesozoic between 200 and 110 Ma (Phillips *et al.* 1998; Griffin *et al.* 2014; Tappe *et al.* 2018), and its diamond potential is typically lower compared with that of Group 1 kimberlites (Field *et al.* 2008). Orangeites are a petrographically and geochemically heterogeneous group of potassic ultramafic rocks that can be reminiscent of cratonic lamproites or certain types of lamprophyres. Besides olivine-rich primitive varieties, there are several occurrences of more evolved olivine-poor orangeites that may contain groundmass sanidine or leucite (Mitchell 1995; Howarth *et al.* 2011). Extremely high large ion lithophile element (LILE) contents and the strongly enriched Sr–Nd–Hf isotopic compositions of orangeites are in stark contrast to archetypal kimberlites, and point to melt derivation from ancient K-metasomatized lithospheric mantle sources (Fraser *et al.* 1985; Nowell *et al.* 2004; Becker and le Roex 2006; Coe *et al.* 2008).

Compared with archetypal kimberlites, the mineralogy and geochemistry of type orangeite from the Kaapvaal craton are surprisingly poorly studied. For example, several decades of extensive platinum mining on the western lobe of the *c.* 2056 Ma Bushveld Complex exposed numerous highly micaceous ultramafic dykes that cut across the central Kaapvaal craton (Fig. 1), but there is a dearth of petrological information about these dykes in the public domain. They have been loosely referred to as ‘micaceous’ or ‘lamprophyric’ (Leeb-Du Toit 1986; Hughes *et al.* 2016; Daya 2019; Lerobane 2020), and most investigations have focussed on their geotechnical properties because these dykes are associated with life-threatening gas outbursts and tunnel failures in underground platinum mining operations (Daya *et al.* 2018; Moate 2019). Geochronology databases list the emplacement age of these diamond-poor to barren potassic ultramafic dykes on the western lobe of the Bushveld Complex at *c.* 140 Ma (Jelsma *et al.* 2009; Tappe *et al.* 2018). This suggests a

genetic link to the *c.* 145 Ma diamondiferous orangeite dyke swarm near Swartruggens (Mitchell 1995), which is located *c.* 60 km west of the Bushveld Complex and is host to Helam Diamond Mine (Gurney and Kirkley 1996; McKenna *et al.* 2004; Field *et al.* 2008; Fig. 1).

In this contribution, we present a detailed account on the mineralogy and geochemistry, including Sr–Nd–Hf isotope data, of the potassic ultramafic dykes from the western Bushveld Complex, sampled at the Impala and Sibanye–Stillwater platinum mine leases near Rustenburg in 2019 (Figs 1 & 2). Our data show that these dyke occurrences represent typical Group 2 kimberlites *or* orangeites, many of which are compositionally evolved, suggesting petrographic resemblance to ultramafic lamprophyres. Although olivine fractionation was the key driver in the evolution of the orangeite magmatic system on the central Kaapvaal craton, it remains unclear why this process was apparently much more efficient compared with orangeite magmatism on the western Kaapvaal craton (Mitchell 1995). The case of the mostly barren Mesozoic orangeite dykes transecting the western Bushveld Complex provides a tangible example of how the diamond potential of potassic ultramafic magmas can rapidly diminish upon ascent – opening further new research avenues into this commercially relevant topic.

Kaapvaal craton and Bushveld Complex: an overview

The Kaapvaal craton in southern Africa presents one of the best preserved remnants of an Archean continent (de Wit *et al.* 1992; Kröner *et al.* 2019). Its rock record dates back to *c.* 3.66 Ga and vestiges of Hadean proto-crust have recently been identified in detrital zircon populations from the Early Archean Barberton Supergroup (Byerly *et al.* 2018). On the basis of its Archean geological evolution, the Kaapvaal craton has traditionally been divided into four geographical domains (i.e. eastern, northern, western, central) comprising typical granite–greenstone rock assemblages (Poujol *et al.* 2003), which amalgamated into a larger proto-continent during the Late Archean (Shirey *et al.* 2002; Smart *et al.* 2016). The central and eastern domains form the Witwatersrand block, which grew by magmatic and tectonic accretion between 3.6 and 3 Ga, as opposed to the younger Kimberley block (3.0–2.7 Ga) that comprises large parts of the western Kaapvaal craton (Schmitz *et al.* 2004). Late Archean cratonization of the Kaapvaal lithosphere was complete by 2.7 Ga, accompanied by the development of large cratonic basins (e.g. the ‘Supergroups’ filling the Pongola, Witwatersrand and Ventersdorp basins), which were to some extent rift-related

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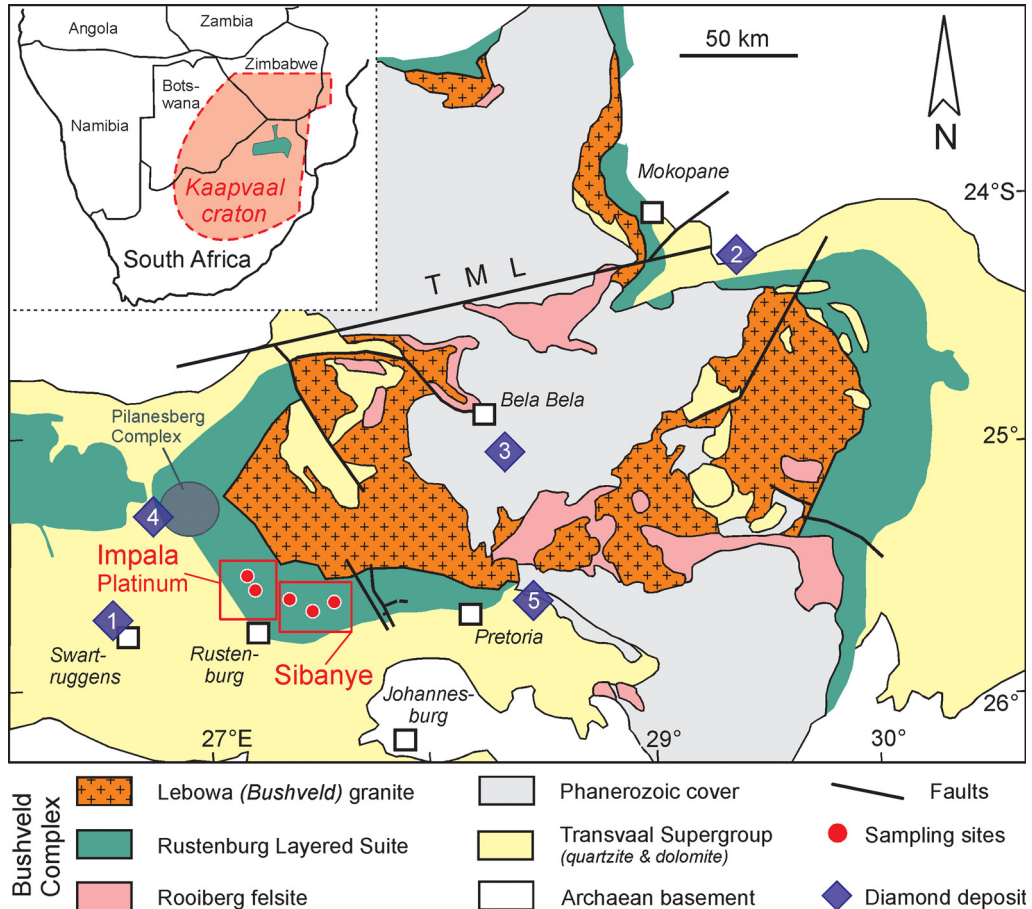


Fig. 1. Geological overview map of the c. 2056 Ma Bushveld Complex and surrounding areas on the central Kaapvaal craton in South Africa (modified from Zeh *et al.* 2019). The inset shows the location of the Kaapvaal craton in southern Africa. The c. 140 Ma orangeite dyke swarm on the western lobe of the Bushveld Complex is mainly exposed in the underground mines of the Impala Platinum Holdings Limited and Sibanye–Stillwater Limited leases near Rustenburg. A c. 145 Ma orangeite dyke system is mined for diamonds near the town of Swart-ruggens some 60 km west of the Bushveld Complex. TML, Thabazimbi–Murchison Lineament. Primary diamond deposits at: (1) ‘Swart-ruggens/Helam’ in a c. 145 Ma orangeite dyke system; (2) ‘Klipspringer–Marsfontein’ in a c. 155 Ma orangeite dyke–blow system; (3) ‘Palmietgat’ in c. 160 Ma kimberlite pipes; (4) ‘Palmietfontein’ in a c. 75 Ma kimberlite pipe; and (5) ‘Premier/Cullinan’ in a c. 1150 Ma kimberlite pipe (the ages are after Griffin *et al.* 2014; Tappe *et al.* 2018; Smart *et al.* 2021).

(Frimmel *et al.* 2009; Agangi *et al.* 2020; Gumsley *et al.* 2020). The Kaapvaal craton was mostly submerged below sea-level between 2.65 and 2 Ga when deposition of the >10 km thick Transvaal Supergroup occurred. At c. 2056 Ma, the Transvaal basal rock succession was intruded by the giant Bushveld Complex on the central Kaapvaal craton (Zeh *et al.* 2015, 2019).

The 400 × 300 × 9 km saucer-shaped Bushveld Complex presents the largest known continental layered mafic intrusion, but it also includes significant volumes of felsic rocks such as the vast Lebowa

granite suite and Rooiberg Group (Cawthorn *et al.* 2006; Fig. 1). The Rustenburg Layered Suite, up to 9 km thick, comprises various types of mafic–ultramafic cumulate rocks, with numerous relatively thin mineralized layers that form chromite and platinum-group-mineral-enriched zones commonly referred to as ‘reefs’, exploited on the three main lobes of the Bushveld Complex (i.e. western, eastern and northern lobes; Fig. 1). The pyroxenitic–anorthositic Merensky Reef and underlying UG2 chromite reef represent world’s largest proven ore reserves of Pt–Pd–Rh and Cr, currently extracted in numerous

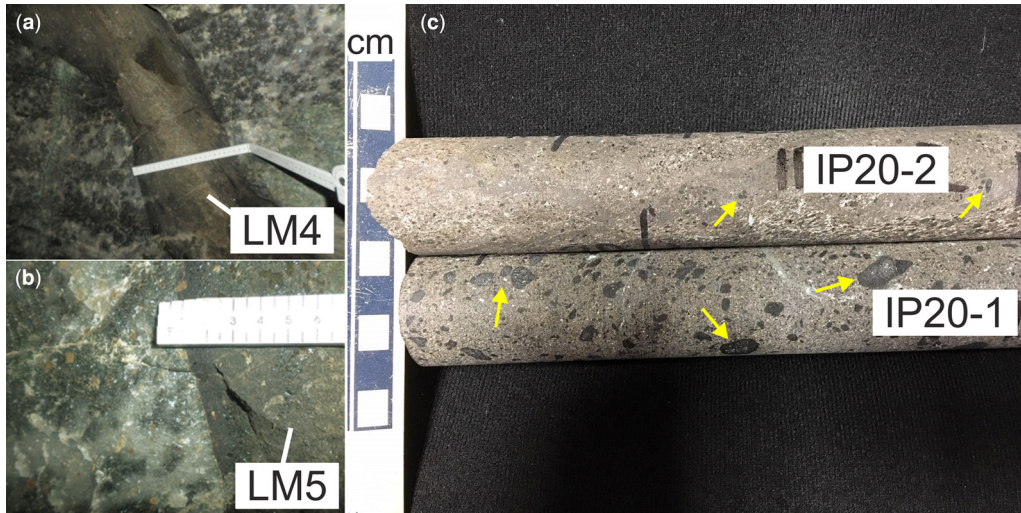


Fig. 2. Typical orangeite dyke exposures in underground mining tunnels and exploration drillcore intersections. (a, b) Approximately 0.2 m wide, near-vertical orangeite dykes exposed in the vicinity of the Merensky Reef at Karee-3 Shaft (samples LM4 and LM5). Note the sharp contacts against footwall anorthosites of the Critical Zone of the Rustenburg Layered Suite. (c) Characteristic micaceous drillcore intersections of so-called olivine-rich (IP20-1) and olivine-poor *or* evolved (IP20-2) orangeite dykes in the vicinity of Impala Shaft-20. The arrows point at relatively abundant olivine macrocrysts that are mostly serpentinized.

underground mining operations between 300 and 1000 m depth below the surface. On the western lobe of the Bushveld Complex, these mining activities exposed a diamond-poor to barren swarm of thin potassic ultramafic dykes of Mesozoic age that forms the subject of this study (Fig. 2).

Methodology

Acquisition and preparation of samples

Fifteen discrete 0.1–1 m wide potassic ultramafic dykes were sampled on the western lobe of the Bushveld Complex near Rustenburg during the course of 2019 (Figs 1 & 2). The samples are derived from the underground mining operations and exploration drilling activities of Impala Platinum (Shaft-20 and surrounding areas, $n = 11$) and Sibanye–Stillwater (formerly Lonmin Platinum Limited, Saffy and Karee-3 shafts, $n = 4$). Underground sampling of the dykes occurred in production tunnels at variable depth levels in footwall drives to the Merensky and UG2 ore reefs, *c.* 800–1000 m below the surface (Fig. 2).

The freshest samples were cut into thin slabs with a rock saw and small blocks were used for the preparation of polished petrographic thin sections at the University of Johannesburg, South Africa. The rock slabs were washed under running water and only materials visibly free from crust-derived rock

fragments were processed further. The rock slabs (*c.* 250 g per sample) were then wrapped in thick plastic sleeves and crushed with a hammer into <8 mm large chips. Approximately 100 g of rock chips per sample were then processed in an agate mill at the University of Johannesburg to obtain analytical grade powder.

Electron microprobe analysis

Quantitative major and minor element analysis of rock-forming minerals was conducted on representative thin sections employing a Cameca SX100 electron probe micro-analyser at the University of Johannesburg. The polished thin sections were covered with *c.* 25 nm thick carbon layers under vacuum in a Quorum Q300T ES coater. The electron microprobe instrument was calibrated using a variety of natural standards: jadeite (Na), olivine (Mg), almandine (Al), diopside (Si), orthoclase (K), wollastonite (Ca), rhodonite (Mn), hematite (Fe), barite (Ba), fluorite (F) and halite (Cl). Synthetic pure metal oxides were used as calibration standards for Ti, Cr and Ni. Chemical elements of interest were measured on their respective X-ray K_{α} lines, and matrix corrections were performed with the ‘X-PHI’ method, which is a $\varphi(\rho z)$ offline correction routine. The lower limit of detection for the quantified major and minor element abundances is *c.* 0.05 wt%. The diverse mineralogy of the potassic ultramafic dykes required different electron

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probe micro-analyser setups for 'dry' phases (e.g. olivine, diopside, spinel-group minerals) and 'volatile-bearing' phases (e.g. phlogopite). Therefore, the electron beam parameters and element signal counting times varied between setups. For example, the electron beam acceleration voltage was adjusted to 15 kV for phlogopite analyses, whereas 20 kV was applied during analysis of the 'dry' phases. The electron beam current was adjusted to 20 nA for both analytical routines. Peak intensity counting times varied between 10 and 60 s depending on the chemical element of interest. The electron beam diameter for spot analysis varied between setups: i.e. 1 μm for 'dry' and 2 μm for 'volatile-bearing' phases. The in-house almandine-rich garnet (Alma-PH1) and diopside-rich clinopyroxene (Diop-PH1) standards were repeatedly analysed during this study to monitor data accuracy and precision (Ngwenya and Tappe 2021).

X-ray fluorescence analysis and CO₂ determination

Bulk rock major and minor element concentrations were determined with a PANalytical MagiX PRO X-ray fluorescence spectrometer (XRF) at the University of Johannesburg. Sample powders were dried in an oven at 105°C prior to fusion into glass discs with a Li₂B₄O₇-LiBO₂ flux. The XRF method detection limit is *c.* 0.05 wt% for all chemical elements reported in Table 1. Instrument calibration was conducted with mixtures of pure metal oxides, and the accuracy of the XRF method was monitored by analysis of certified reference materials (BE-N, JSy-1, SARM-2, SARM-16), as well as analysis of carbonatite ST199 and kimberlite ST220II in-house standards (Dongre and Tappe 2019). For the in-house standards, all major and minor elements reported in Table 1 and Supplementary Material 1 were reproduced within 3% and typically better than 1% of the values listed in Tappe *et al.* (2006), except for MnO (better than 8%), Na₂O (12%) and K₂O (6%). Loss on ignition was determined after heating of the sample powders to 930°C in air and holding at this temperature for 30 min.

Bulk rock CO₂ contents were determined at the University of Münster (Germany) by liberation of CO₂ gas from the powders in a reaction with H₃PO₄. Liberated CO₂ gas was then cryogenically purified and collected into break-seal Pyrex tubes. The sample CO₂ concentrations were determined relative to a natural carbonate standard using a Thermo-Finnigan Delta Plus gas source mass spectrometer fitted with a dual inlet. The detection limit for the CO₂ concentration measurements was 0.01 wt% (Table 1).

Inductively coupled plasma mass spectrometry

Sample powder digestion for trace element inductively coupled plasma mass spectrometry (ICP-MS) analysis was undertaken at the University of the Witwatersrand in Johannesburg. Approximately 50 mg of rock powder per sample were loaded into microwave Teflon vessels. Concentrated HF-HNO₃ (6 ml) was added to each vessel, and powders were dissolved in a MARS microwave digester for 60 min. The sample solutions were then transferred to 15 ml Savillex beakers, which were placed on a hotplate at 60°C for 24 h and subsequently dried down at 70°C. Approximately 2 ml HNO₃ were then added to each beaker, and the previous heating step was repeated twice. Trace element analysis was performed with a Perkin Elmer ELAN Drc-e Q-ICP-MS instrument. Internal standards were Rh, Re, In and Bi, and ICP-MS calibration standards were made from certified solutions (Wilson 2012). During this study, replicate analyses of the USGS basaltic reference material BHVO-2 (Jochum *et al.* 2016) and the Mintek kimberlite reference material SARM-39 (Roy *et al.* 2007; Tappe *et al.* 2020a) yielded trace element concentrations within 10% of the recommended values, excluding Gd, Tm, Nb and Ta (within 20%).

Sr-Nd-Hf isotope ratio measurements

Purification of bulk rock Sr, Nd and Hf cuts for isotope ratio measurements was achieved through ion exchange chromatography at the University of Münster, and detailed descriptions of the clean-laboratory and mass spectrometry methods are provided in Tappe *et al.* (2020a, b). Approximately 100 mg of each sample powder were dissolved in HF-HNO₃ at 180°C (in Savillex beakers) and re-dissolved twice in 6 M HCl until clear solutions were obtained. Final sample digests were split into two solution aliquots for further processing for Sr-Nd and Hf isotope analysis, respectively. Strontium and the REE fractions were collected using conventional cation exchange chromatography (AG50W-X8, 200–400 mesh). Neodymium was subsequently isolated from the REE fraction using LN Spec resin (50–100 mesh). Hafnium was isolated in a separate procedure that also uses LN Spec resin. The total procedural blanks are negligible at <50 pg Sr, <30 pg Nd and <10 pg Hf (Bast *et al.* 2015; Tappe *et al.* 2020a).

Strontium isotope ratio measurements were performed by thermal ionization mass spectrometry on a Thermo Scientific Triton instrument, whereas the Nd and Hf isotope ratios were measured by plasma ionization with a Thermo Scientific Neptune Plus instrument. Measured ⁸⁷Sr/⁸⁶Sr, ¹⁴³Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf ratios were normalized using an

Table 1. Major (wt%) and trace (ppm) element compositions, as well as Sr–Nd–Hf isotopic compositions of Mesozoic Group 2 kimberlite dykes (orangeites) that intersect the western Bushveld Complex, central Kaapvaal craton, South Africa

Sample ID: Mining block:	Impala										Sibanye (Marikana)				
	Orangeite					Evolved orangeite					Evolved orangeite				
	IP20-1 Shaft-20	IP20-3 Shaft-20	IP20-4 Shaft-20	IP20-5 Shaft-20	IP20-7 Shaft-20	IP20-11 Shaft-20	IP20-2 Shaft-20	IP20-6 Shaft-20	IP20-8 Shaft-20	IP20-9 Shaft-20	IP20-10 Shaft-20	LM-4 Karee-3	LM-5 Karee-3	LM-6B1 Saffy	LM-6B2 Saffy
SiO ₂	39.0	36.7	41.2	40.2	40.2	38.4	38.6	36.4	32.2	38.2	40.1	40.1	38.6	40.1	38.7
TiO ₂	0.91	0.78	1.24	1.08	1.09	0.87	2.93	2.83	3.04	1.63	1.31	1.88	1.93	1.89	1.78
Al ₂ O ₃	4.74	6.22	5.55	4.77	4.64	6.64	4.41	4.55	5.66	5.86	5.46	6.37	6.59	6.48	6.28
Fe ₂ O ₃ ^T	7.64	8.05	9.46	8.56	9.39	7.70	12.3	11.3	11.7	10.2	9.06	10.2	10.1	10.3	10.1
MnO	0.13	0.18	0.16	0.14	0.14	0.16	0.22	0.19	0.18	0.19	0.15	0.18	0.17	0.16	0.17
MgO	26.1	22.6	22.8	25.7	22.9	23.2	16.6	16.5	15.1	18.8	20.2	15.5	14.9	15.8	17.1
CaO	7.47	8.69	7.51	6.24	8.68	7.22	10.9	11.7	13.4	8.33	9.76	11.3	10.9	11.8	10.4
Na ₂ O	0.05	0.12	0.68	0.49	0.58	0.08	1.65	1.48	1.06	1.27	1.53	1.61	1.23	1.14	1.00
K ₂ O	4.31	6.19	3.21	3.28	3.15	6.22	5.01	5.23	5.28	6.90	3.29	3.23	4.38	3.15	3.76
P ₂ O ₅	0.35	0.10	0.61	0.48	0.66	b.d.	1.72	1.70	1.18	0.61	0.54	0.91	0.97	0.90	0.89
Cr ₂ O ₃	0.38	0.31	0.22	0.22	0.22	0.29	0.15	0.13	0.15	0.23	0.20	0.16	0.16	0.16	0.17
NiO	0.14	0.10	0.13	0.15	0.14	0.11	0.07	0.07	0.05	0.08	0.11	0.06	0.05	0.06	0.06
LOI	6.9	8.3	5.3	6.7	5.8	7.0	3.0	4.8	7.7	5.3	5.8	6.3	7.5	6.0	8.3
Total	98.0	98.4	98.0	97.9	97.7	97.9	97.6	96.8	96.8	97.5	97.5	97.8	97.4	97.9	98.7
CO ₂	2.0	4.7	0.28	1.6	0.45	2.1	0.60	0.33	3.1	0.25	0.40	1.1	3.0	0.63	3.0
SO ₃	0.19	0.35	0.51	0.65	1.2	b.d.	0.38	0.44	0.76	0.10	1.0	0.29	0.12	0.32	0.21
Co	56.1	53.8	55.1	57.3	56.4	70.4	51.5	48.5	47.3	59.6	65.0	58.3	54.8	58.5	59.7
Cr	811	1503	454	419	465	1694	701	558	415	1260	485	496	529	460	563
Ni	871	655	807	930	850	776	441	432	302	579	798	430	341	427	454
Cu	38.8	5.9	37.5	29.2	37.1	8.5	96.5	94.6	39.6	52.9	49.1	63.5	63.2	63.3	59.7
Zn	37.1	34.4	51.6	40.6	51.9	47.6	66.4	65.3	59.6	76.4	74.6	82.8	85.8	88.5	82.0
Sc	8.6	7.4	11.5	10.2	11.3	19.6	24.8	25.8	21.3	15.6	18.6	23.1	23.2	22.9	21.1
V	112	17.0	123	96.4	115	27.9	51.3	59.9	110	115	292	215	287	144	286
Ga	6.5	6.7	6.9	6.5	6.6	11.6	10.0	10.2	10.4	15.0	11.9	15.9	16.3	17.0	15.9
Ba	1802	2946	2786	2115	3036	2734	4047	4938	4651	4647	6106	1864	2938	3327	3154
Li	1.62	1.98	5.18	4.39	4.14	4.00	8.08	7.55	5.88	9.69	5.55	10.5	11.0	7.45	10.4
Cs	1.95	2.24	1.35	1.18	1.41	2.72	2.88	2.77	2.49	3.66	2.00	1.91	2.06	1.54	1.84
Rb	153	145	106	91.8	93.3	266	179	173	191	340	153	194	230	172	202
Sr	435	808	720	448	668	797	1061	1189	1417	1047	957	858	942	746	858
Zr	84.2	66.3	110	106	130	68.3	389	397	174	189	149	310	306	307	272
Hf	2.53	2.45	3.88	3.61	4.16	2.57	12.2	13.0	5.84	4.81	4.21	7.80	7.70	7.42	6.67
Nb	50.9	42.2	61.3	48.4	61.4	46.7	120	109	102	60.6	82.9	108	103	107	94.0

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Ta	3.07	2.42	3.88	3.14	3.90	4.17	5.81	5.96	5.72	2.43	4.30	4.97	4.89	4.93	4.48
Y	4.51	11.4	9.59	6.19	10.4	12.1	33.9	34.9	19.6	20.4	14.1	21.0	21.2	20.7	19.2
Pb	8.84	4.01	14.0	7.11	13.9	2.74	17.7	19.6	25.2	14.8	41.9	28.3	24.6	27.7	23.4
Th	5.53	8.40	8.71	5.25	9.47	12.9	20.7	25.2	20.2	9.56	13.2	15.9	15.7	15.4	14.2
U	1.76	0.97	1.58	1.77	2.45	1.50	5.91	6.28	5.59	4.85	4.08	4.93	4.40	4.78	4.16
W	5.3	0.25	0.30	0.57	1.5	0.12	0.55	0.65	0.54	0.80	2.5	0.30	1.3	0.20	0.33
La	45.8	55.2	65.7	46.5	67.9	61.2	172.2	199.0	169.0	80.5	83.9	108.3	107.0	102.3	95.9
Ce	91.6	114.5	130.9	92.3	134.6	134.1	344.6	395.8	330.2	147.5	159.5	225.7	212.5	214.2	190.5
Pr	10.63	13.35	15.31	10.90	15.85	15.95	39.57	44.79	36.39	16.82	18.08	24.70	24.94	23.40	22.46
Nd	38.9	47.5	55.4	40.8	57.4	58.9	141.4	157.9	122.2	61.1	65.1	85.1	92.0	79.2	80.1
Sm	6.20	8.38	8.98	6.57	9.65	9.17	25.11	28.30	20.42	11.54	12.38	13.20	14.10	13.36	12.89
Eu	1.66	2.42	2.47	1.77	2.64	2.55	6.53	7.40	5.32	3.72	4.01	3.17	3.63	3.51	3.34
Gd	3.95	5.66	6.30	4.52	6.64	6.34	18.61	20.65	14.42	7.67	7.39	9.96	10.23	9.54	9.28
Tb	0.43	0.73	0.73	0.52	0.80	0.73	2.42	2.67	1.69	0.99	0.84	1.19	1.22	1.14	1.12
Dy	1.48	3.23	2.85	1.99	3.11	2.77	10.44	11.28	6.50	4.35	3.15	4.79	4.83	4.54	4.30
Ho	0.20	0.51	0.43	0.30	0.49	0.40	1.59	1.73	0.96	0.69	0.47	0.70	0.73	0.67	0.64
Er	0.51	1.14	1.01	0.71	1.18	1.10	3.44	3.67	2.26	1.58	1.20	1.76	1.79	1.73	1.57
Tm	0.06	0.17	0.14	0.09	0.16	0.14	0.46	0.49	0.28	0.20	0.15	0.22	0.21	0.22	0.20
Yb	0.35	1.05	0.83	0.58	0.95	0.91	2.54	2.77	1.69	1.10	0.94	1.30	1.31	1.25	1.17
Lu	0.05	0.15	0.12	0.09	0.13	0.13	0.34	0.36	0.24	0.15	0.14	0.19	0.19	0.18	0.17
⁸⁷ Sr/ ⁸⁶ Sr (m)	0.709390(6)	0.708025(7)					0.707962(6)					0.708486(5)	0.708555(7)	0.708592(7)	0.708620(7)
* ⁸⁷ Sr/ ⁸⁶ Sr (i)	0.707410	0.707013					0.707012					0.707218	0.707183	0.707299	0.707293
¹⁴³ Nd/ ¹⁴⁴ Nd (m)	0.512243(6)	0.512054(8)					0.512030(6)					0.511993(6)	0.511997(5)	0.512003(7)	0.512010(5)
* ¹⁴³ Nd/ ¹⁴⁴ Nd (i)	0.512154	0.511956					0.511932					0.511907	0.511912	0.511909	0.511921
ϵ Nd (i)	-5.8	-9.6					-10.1					-10.6	-10.5	-10.6	-10.3
2 SD	0.44	0.42					0.41					0.44	0.44	0.43	0.43
¹⁷⁶ Hf/ ¹⁷⁷ Hf (m)	0.282633(7)	0.282476(7)					0.282301(6)					0.282358(7)	0.282350(6)	0.282352(9)	0.282350(7)
* ¹⁷⁶ Hf/ ¹⁷⁷ Hf (i)	0.282626	0.282454					0.282291					0.282349	0.282342	0.282343	0.282340
ϵ Hf (i)	-2.5	-8.6					-14.4					-12.3	-12.6	-12.5	-12.6
2 SD	0.73	0.65					0.70					0.72	0.71	0.75	0.72
$\Delta\epsilon$ Hf (i)	5.4	5.5					0.4					3.3	2.8	3.0	2.5

Major and minor element concentrations are X-ray fluorescence spectrometry data determined at the University of Johannesburg; b.d., below detection.

Trace element concentrations are solution ICP–MS data determined at the University of the Witwatersrand. The CO₂ concentrations were determined at the University of Münster.

Fe₂O₃^T = total Fe as ferric iron; LOI, loss on ignition, is defined as the difference in sample weight after ignition at 930°C for 30 min.

Sr isotope ratios were determined by thermal ionization mass spectrometry, and Nd–Hf isotope ratios were determined by MC–ICP–MS at the University of Münster.

*Initial isotope ratios calculated for a magma emplacement age of 140 Ma (Tappe *et al.* 2018) using the following decay constants: ⁸⁷Rb 1.42 × 10⁻¹¹, ¹⁴⁷Sm 6.54 × 10⁻¹² and ¹⁷⁶Lu 1.865 × 10⁻¹¹ per year.

[†]Initial ϵ Nd values calculated using ¹⁴⁷Sm decay constant of 6.54 × 10⁻¹² per year (Lugmair and Marti 1978); (¹⁴³Nd/¹⁴⁴Nd)_{CHUR} = 0.512630 and (¹⁴⁷Sm/¹⁴⁴Nd)_{CHUR} = 0.1960 (Bouvier *et al.* 2008).

[‡]Initial ϵ Hf values calculated using ¹⁷⁶Lu decay constant of 1.865 × 10⁻¹¹ per year (Scherer *et al.* 2001); (¹⁷⁶Hf/¹⁷⁷Hf)_{CHUR} = 0.282785 and (¹⁷⁶Lu/¹⁷⁷Hf)_{CHUR} = 0.0336 (Bouvier *et al.* 2008). CHUR, Chondritic uniform reservoir.

The 2 SD uncertainties of the ϵ Nd and ϵ Hf values entail a full error propagation, where the uncertainties of the emplacement age and parent–daughter element ratios were set at 20 and 5%, respectively.

$\Delta\epsilon$ Hf (i) is defined as $\epsilon_{\text{Hf}}^i - (1.59 \times \epsilon_{\text{Nd}}^i + 1.28)$, such that analyses with positive values fall by definition above the Nd–Hf isotope regression line of Chauvel *et al.* (2008).

Numbers in parentheses are 2-sigma-of-the-mean uncertainties for individual isotope ratio measurements.

exponential mass fractionation law and $^{88}\text{Sr}/^{86}\text{Sr}$ of 8.37521, $^{146}\text{Nd}/^{144}\text{Nd}$ of 0.7219 and $^{179}\text{Hf}/^{177}\text{Hf}$ of 0.7325, respectively. The measured Sr–Nd–Hf isotope ratios for all ‘unknowns’ including the secondary standards are reported relative to $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.710245 for NBS987, $^{143}\text{Nd}/^{144}\text{Nd}$ of 0.512115 for JNdi-1 and $^{176}\text{Hf}/^{177}\text{Hf}$ of 0.282160 for AMES/ JMC-475 standard solutions, respectively. During the analyses in 2020, repeated measurements of $^{87}\text{Sr}/^{86}\text{Sr}$ for the NBS987 primary standard averaged 0.710282 ± 21 (2 SD, $n = 4$), $^{143}\text{Nd}/^{144}\text{Nd}$ for the JNdi-1 primary standard averaged 0.512069 ± 8 (2 SD, $n = 12$) and $^{176}\text{Hf}/^{177}\text{Hf}$ for the AMES primary standard averaged 0.282137 ± 17 (2 SD, $n = 14$).

We also processed the USGS basaltic reference materials BCR-2 and BHVO-2 alongside our samples, and the results are in excellent agreement with the recommended isotope ratios. For BCR-2 the measured values are: $^{87}\text{Sr}/^{86}\text{Sr} = 0.704996 \pm 6$; $^{143}\text{Nd}/^{144}\text{Nd} = 0.512634 \pm 25$; and $^{176}\text{Hf}/^{177}\text{Hf} = 0.282869 \pm 17$. For BHVO-2 the measured values are: $^{87}\text{Sr}/^{86}\text{Sr} = 0.703471 \pm 6$; $^{143}\text{Nd}/^{144}\text{Nd} = 0.512990 \pm 9$; and $^{176}\text{Hf}/^{177}\text{Hf} = 0.283100 \pm 37$. We also processed and analysed the Mintek kimberlite reference material SARM-39 during this analytical session in 2020, and the measured values are: $^{87}\text{Sr}/^{86}\text{Sr} = 0.708849 \pm 7$; $^{143}\text{Nd}/^{144}\text{Nd} = 0.512610 \pm 7$; and $^{176}\text{Hf}/^{177}\text{Hf} = 0.282595 \pm 63$. The results for this matrix-matched reference material are in excellent agreement with the recommended Sr–Nd–Hf isotope ratios for SARM-39 (Tappe *et al.* 2020a).

For our samples, the parent–daughter element ratios were calculated using the Rb, Sr, Sm, Nd, Lu and Hf concentrations determined by solution-mode ICP-MS analysis. The initial isotope ratios and notations are calculated at 140 Ma (Tappe *et al.* 2018) and the 2 SD uncertainties of the calculated initial epsilon Nd and Hf values reported in Table 1 entail a full propagation of ‘errors’. The error propagation comprises highly conservative uncertainty estimates, i.e. 20% for the c. 140 Ma magma emplacement age (140 ± 30 Ma) and 5% for the parent–daughter element ratios. The age corrections are based on the following decay constants: ^{87}Rb 1.42×10^{-11} , ^{147}Sm 6.54×10^{-12} and ^{176}Lu 1.865×10^{-11} per year.

Results

For convenience, results are described separately for the samples from the Impala and Sibanye mining concessions, despite the fact that they are derived from a single large potassic ultramafic dyke swarm. All data generated during this study (i.e. mineral and bulk rock compositions including Sr–Nd–Hf isotope ratios) are provided online in Supplementary

Material 1. In addition, the complete bulk rock geochemical dataset is reported as Table 1 in the main article.

Petrography and classification

Our mineralogy and geochemistry data (see below) reveal that the highly micaceous ultramafic dykes intersecting the western Bushveld Complex represent ‘typical’ Kaapvaal orangeites (formerly Group 2 kimberlites), in contrast to their previous categorization as lamprophyres (cf. Hughes *et al.* 2016). A lamprophyric character can also be excluded on the basis of petrography (Fig. 3), with the absence or paucity of hydrous mafic phenocrysts/macrocysts and phlogopite being largely confined to the groundmass (Rock 1991; Tappe *et al.* 2005). On a macroscopic scale, the orangeite dykes studied comprise olivine-rich and olivine-poor varieties; the latter are, hereafter, designated as ‘evolved’ orangeites (Fig. 2c).

The Impala orangeite samples have inequigranular textures with fairly abundant olivine macrocrysts (up to 15 vol%), ranging in size from 1 to 10 mm, as well as highly abundant microphenocrysts (100–500 μm) of phlogopite laths and clinopyroxene prisms (Fig. 3). These coarse to fine crystal populations are set in a very fine-grained groundmass (<100 μm) dominated by phlogopite and clinopyroxene with minor amounts of spinel-group minerals, apatite, carbonate and serpentine (Fig. 3). The olivine macrocrysts are largely serpentinized but rare relict olivine cores have been observed and analysed (see below). The phlogopite laths are typically zoned displaying pale yellow cores and deep orange rims under plane polarized light (Fig. 3a). Rare serpentinized peridotitic microxenoliths up to 20 mm across also occur.

The more evolved orangeite dykes from the Impala and Sibanye mining concessions mostly lack inequigranular textures because olivine macrocrysts are very rare or even absent (<2 vol%; Fig. 2c). However, the common occurrence of aggregates of phlogopite laths >500 μm in length results in a micro-glomeroporphyritic texture. Locally, phlogopite laths show preferred orientations as part of prominent flow structures (Fig. 3c). Rare olivine microcrysts displaying core–rim zoning are observed as inclusions within larger complexly zoned phlogopite plates (Fig. 3f). The phlogopite–diopside dominated groundmass of the evolved orangeites is petrographically similar to its counterpart in the more primitive orangeite dykes, but in addition it may contain subordinate amounts of K-feldspar (?sanidine) and andradite-rich magmatic garnet. Groundmass phlogopite in the evolved orangeite dykes studied may show a decussate texture with randomly oriented and interlocking laths (Fig. 3d–f).

Nature and origin of Kaapvaal orangeites

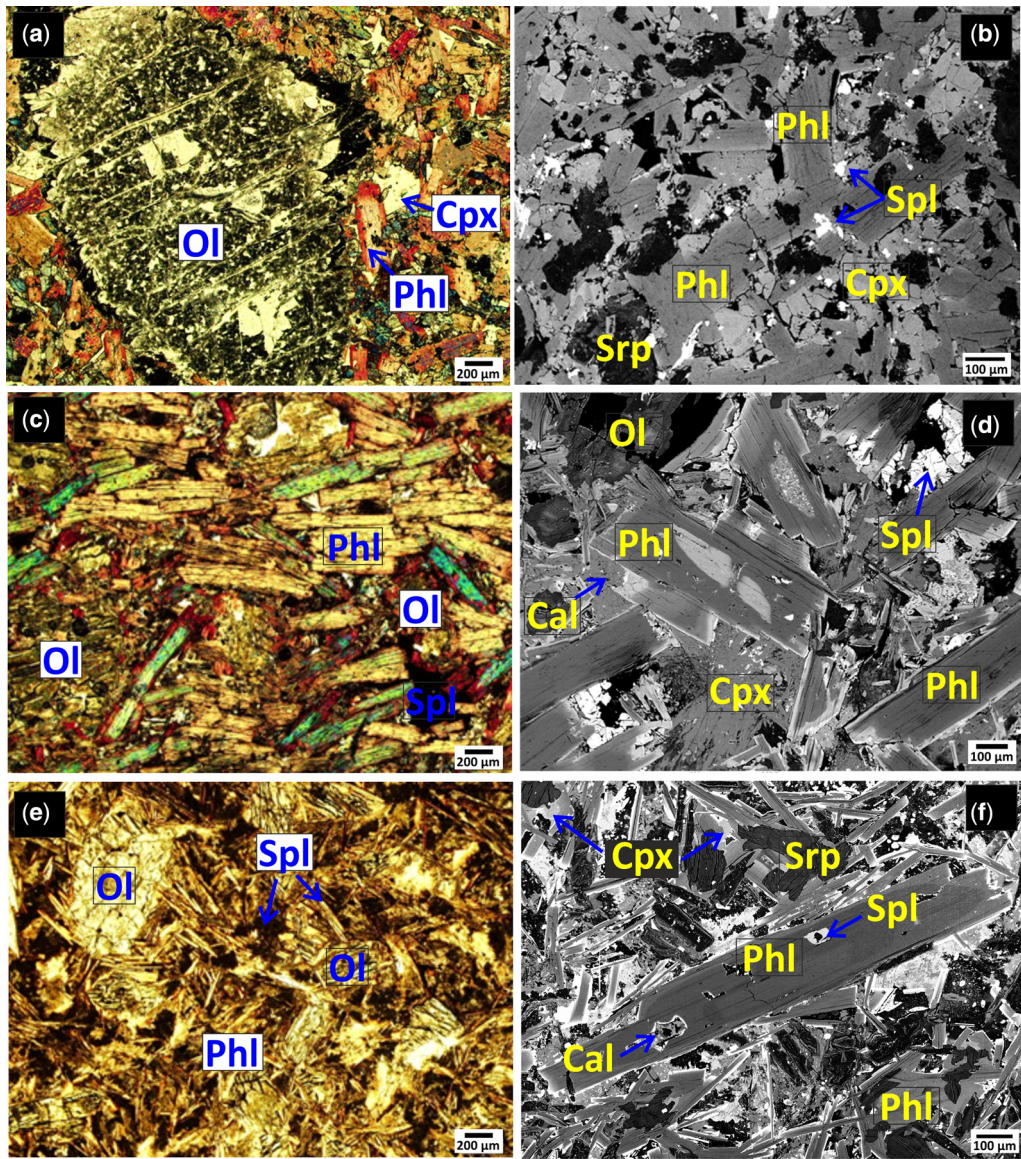


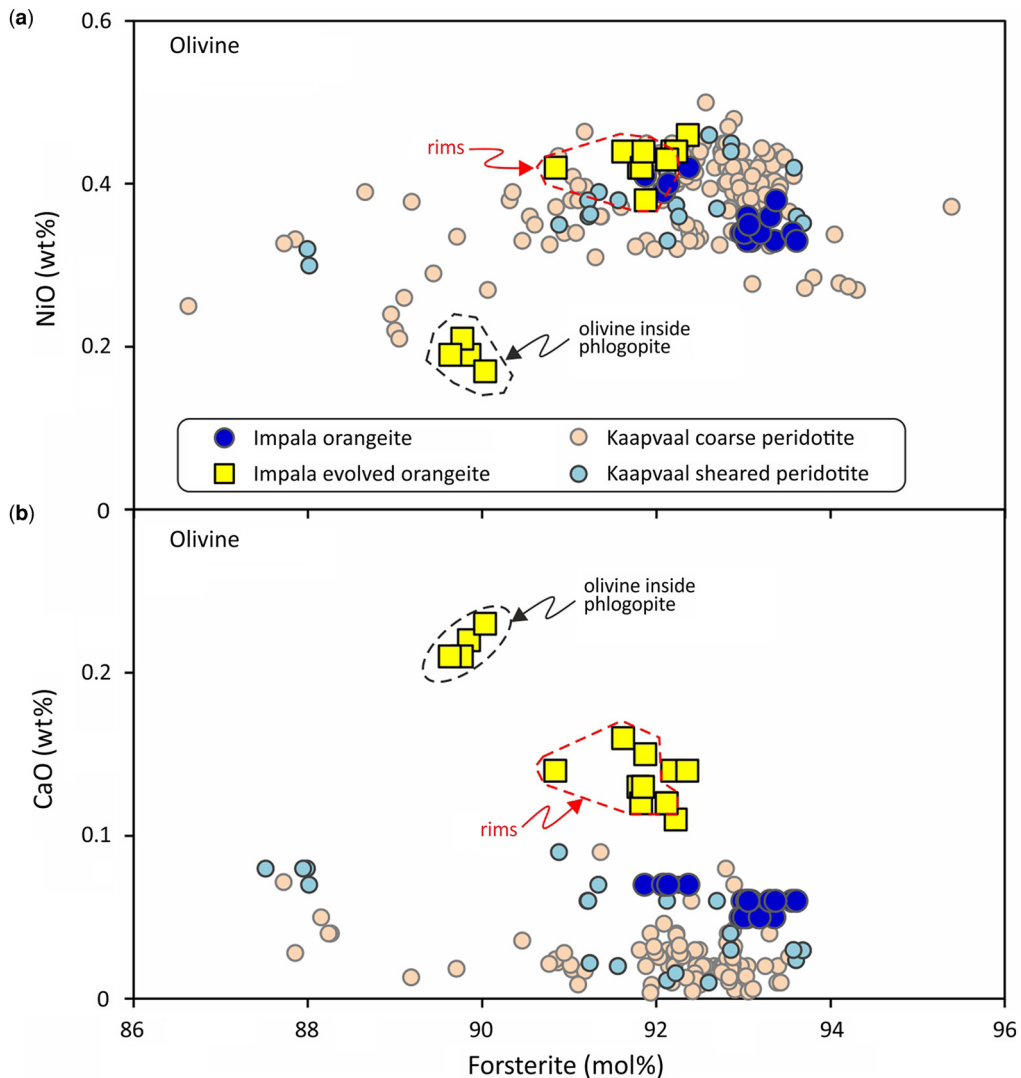
Fig. 3. Photomicrographs and BSE images of representative samples from Bushveld-intersecting orangeite dykes. (a) Impala orangeite dyke IP20-1 exhibiting a serpentinized olivine macrocryst set in a phlogopite–clinopyroxene dominated groundmass (plane polarized light). (b) Impala orangeite dyke IP20-1 exhibiting abundant zoned phlogopite laths in the groundmass plus interstitial clinopyroxene, Cr-rich spinel and serpentine (?pseudomorph after olivine microphenocrysts). (c) Impala evolved orangeite dyke IP20-9 exhibiting flow-aligned phlogopite laths that drape around serpentinized olivine microphenocrysts. Note the discrete groundmass Cr-rich spinel grains (cross polarized light). (d) Impala evolved orangeite dyke IP20-2 exhibiting complexly zoned phlogopite laths and plates in the groundmass plus interstitial acicular clinopyroxene prisms, equant Cr-rich spinel grains and calcite. Note also the serpentinized subhedral olivine microphenocryst. (e) Sibanye evolved orangeite dyke LM4 characterized by rare serpentinized subhedral olivine microphenocrysts set in a fine groundmass that is dominated by acicular phlogopite and clinopyroxene plus rare equant Cr-rich spinel grains (plane polarized light). (f) Sibanye evolved orangeite dyke LM5 showing a zoned and inclusion-bearing phlogopite microphenocryst set in a fine groundmass that is dominated by acicular phlogopite and clinopyroxene laths forming a decussate micro-texture. Cal, Calcite; Cpx, clinopyroxene; Ol, olivine; Phl, phlogopite; Spl, spinel; Srp, serpentine.

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523 *Mineral compositions*

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 525 *Olivine.* Owing to widespread alteration of olivine to serpentine (Fig. 3a), olivine major and minor
 526 element analysis was only possible for orangeite dyke samples from the Impala mining concession
 527 (Supplementary Material 1). Olivine in the Impala orangeite dykes shows a rather restricted
 528 compositional range with high forsterite (91.9–93.6 mol%) and NiO (0.33–0.42 wt%) contents at
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<0.07 wt% CaO (Fig. 4). These compositions resemble those of peridotite-derived mantle xenocrysts reported from kimberlites and related rocks on cratons worldwide (Kamenetsky *et al.* 2008; Tappe *et al.* 2009; Bussweiler *et al.* 2015; Howarth and Taylor 2016; Giuliani 2018; Shaikh *et al.* 2019; Rooney *et al.* 2020; Ngwenya and Tappe 2021). Olivine in the evolved orangeite dykes has slightly lower forsterite contents (90.8–92.4 mol%) at elevated NiO (0.38–0.46 wt%), CaO (0.11–0.



577
 578 **Fig. 4.** Olivine major and minor element compositions in Bushveld-intersecting orangeite and evolved orangeite
 579 dykes from Impala. (a) NiO v. forsterite contents. (b) CaO v. forsterite contents. Published olivine compositions for
 580 coarse and sheared peridotite xenoliths from the Kaapvaal craton are shown for comparison and the data are listed in
 Supplementary Material 1 together with the literature sources.

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16 wt%) and MnO (0.11–0.16 wt%) concentrations (Fig. 4), which is more typical of magmatic olivine in ultrapotassic systems such as lamproites (Prelevic and Foley 2007; Jaques and Foley 2018; Howarth and Giuliani 2020). Olivine inclusions in phlogopite have the lowest forsterite (89.6–90.0 mol%) and NiO (0.17–0.21 wt%) contents at elevated CaO (0.21–0.23 wt%) and MnO (0.20–0.22 wt%) (Fig. 4), suggestive of a magmatic origin during the early stages of groundmass crystallization.

Phlogopite. Phlogopite is the dominant mica type in the Impala and Sibanye orangeite dykes, where it is largely restricted to the groundmass (Fig. 3). However, phlogopite microphenocrysts also occur in the form of laths and plates ranging in size between 0.3 and 1 mm. They show complex zoning and typically contain minute inclusions of spinel, olivine and carbonate (Fig. 3f). The phlogopite laths and plates are commonly intersected by prismatic groundmass diopside establishing their somewhat earlier crystallization during fast cooling of the narrow dykes (Fig. 3d).

Phlogopite laths and plates in both the olivine-rich and olivine-poor (or evolved) orangeite dykes from Impala have similar core compositions that are MgO-rich and relatively poor in Al₂O₃ and FeO, similar to the phlogopite core compositions in the evolved orangeites from Sibanye (Fig. 5a–c). Although phlogopite from the three sample subsets has evolved by Al-depletion and Fe-enrichment (i.e. a tetraferriphlogopite trend), we note subtle differences in the individual core–rim trends relating to the magnitude of the Al–Fe fractionation (Fig. 5a, b). Moreover, phlogopite from the olivine-rich Impala orangeites shows strong Ti-depletion towards the rims, whereas micas in the evolved orangeites from Impala and Sibanye have constant or increasing TiO₂ concentration levels, respectively, towards the rims (up to 4.8 wt% TiO₂; Fig. 5a). In general, the Al–Ti–Fe systematics of phlogopite in the potassic ultramafic dykes from the western Bushveld Complex are typical for Kaapvaal craton orangeites (Mitchell 1995), and only the most evolved dyke compositions from the Sibanye mining concession show a subtle lamproitic mica affinity (e.g. Ti-enrichment). An affinity of the Impala and Sibanye dykes to ultramafic lamprophyres can be largely ruled out because of their generally low Al₂O₃ contents of mica (\ll 13.5 wt%), which is in contrast to the relatively Al-rich phlogopite compositions ($>$ 14 wt% Al₂O₃) of aillikites from localities worldwide (Tappe *et al.* 2004; Tappe *et al.* 2006; Nielsen *et al.* 2009; Nasir *et al.* 2011; Dalton *et al.* 2019; Fig. 5a).

The Cr₂O₃ contents of phlogopite in the orangeite dykes from the western Bushveld Complex are mostly $<$ 1 wt%. However, rare resorbed mica

cores in the olivine-rich Impala dykes and more evolved Sibanye dykes reach higher Cr₂O₃ concentration levels of up to 1.8 wt% (Fig. 5c). The phlogopite BaO and F concentration levels are typically below 1 wt%, which presents a marked compositional difference to archetypal kimberlites (Giuliani *et al.* 2016) and cratonic lamproites (Ngwenya and Tappe 2021). These Ba–F systematics are similar to those of phlogopite in ultramafic lamprophyres such as type aillikite (Tappe *et al.* 2006). Very rare biotite laths occur only in the evolved orangeite dykes from the Impala and Sibanye mining concessions, and they show some of the highest Al₂O₃ (up to 14.3 wt%) and F (up to 1.1 wt%) contents among the analysed micas at very low Mg# ($<$ 45) and Cr₂O₃ ($<$ 0.1 wt%) (Fig. 5c).

Clinopyroxene. Clinopyroxene is diopside-rich and occurs as small prismatic microphenocrysts (coeval with the phlogopite microphenocrysts) and needle-shaped crystals in the groundmass of the orangeite dykes that intrude the western Bushveld Complex. It also occupies interstitial space between slightly larger interlocking phlogopite laths (Fig. 3b, d). Although subtle compositional zoning has been observed for the clinopyroxene prisms, it is not as prominent as for the coexisting phlogopite laths and plates.

Clinopyroxene in the Impala orangeite dykes has an almost pure diopside composition (En_{44.3–49.6}Wo_{50.3–51.6}Fs_{0–5.3}) with very low Al₂O₃ ($<$ 0.5 wt%) and TiO₂ ($<$ 1 wt%) contents (Fig. 5d). Diopside-rich clinopyroxene in the groundmass of the evolved orangeite dykes from both the Impala and Sibanye mining concessions (En_{40.0–49.1}Wo_{49.7–51.6}Fs_{0–10}) is also Al₂O₃ poor ($<$ 0.5 wt%), but it has notably higher TiO₂ concentrations compared with diopside in the more primitive olivine-rich dykes from Impala, ranging between 1.6 and 2.6 wt% (Fig. 5d). The Cr₂O₃ and Na₂O contents of clinopyroxene are similarly low for the three sample subsets, reaching up to 0.2 and 1.36 wt%, respectively. These groundmass clinopyroxene compositions are typical for Kaapvaal craton orangeites (Mitchell 1995), whereas they differ greatly from the much more Al- and Ti-enriched groundmass diopside compositions of ultramafic lamprophyres from localities worldwide (e.g. Tappe *et al.* 2004, 2006; Dalton *et al.* 2019; Fig. 5d).

Spinel-group minerals. Spinel-group minerals are common accessory constituents of the petrographically diverse samples from the orangeite dyke swarm examined here. They typically occur as $<$ 50 μ m euhedral to subhedral inclusions within microphenocrystic olivine and phlogopite, but are only rarely observed as $<$ 100 μ m groundmass phase occupying textural interstices (Fig. 3). Atoll-

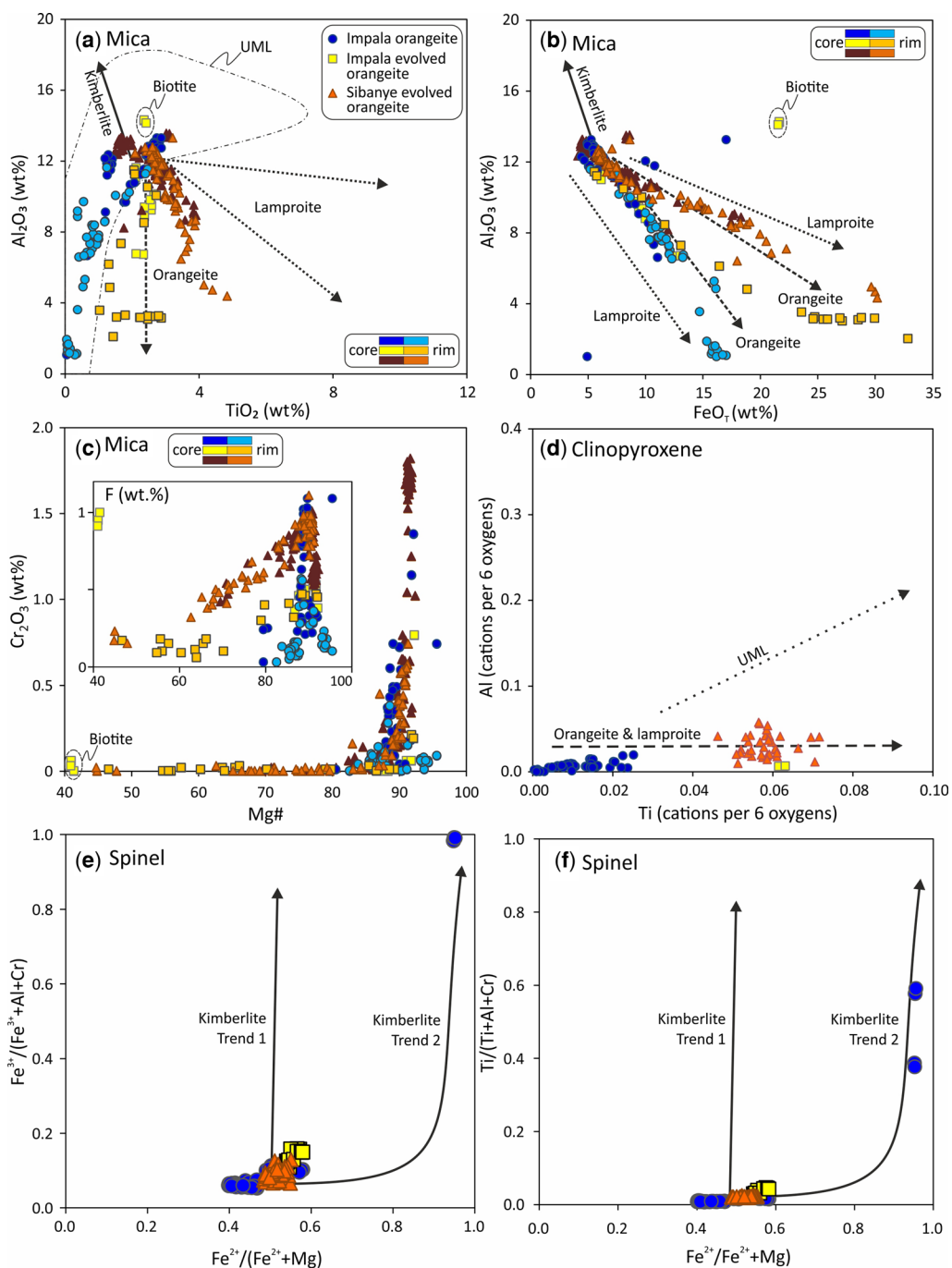


Fig. 5. Major and minor element compositions of petrogenetically important mineral constituents of Bushveld-intersecting orangeite dykes. (a) Al_2O_3 v. TiO_2 ; (b) Al_2O_3 v. total FeO ; and (c) Cr_2O_3 and F v. Mg-number for abundant phlogopite and rare biotite in the groundmass. (d) Atomic Al v. Ti contents of diopside-rich groundmass clinopyroxene. (e) $\text{Fe}^{3+}/(\text{Fe}^{3+} + \text{Al} + \text{Cr})$ and (f) $\text{Ti}/(\text{Ti} + \text{Al} + \text{Cr})$ v. $\text{Fe}^{2+}/(\text{Fe}^{2+} + \text{Mg})$ for groundmass spinel. Mineral compositional trends that are typical for kimberlites, lamproites and orangeites are taken from Mitchell (1995), whereas those for ultramafic lamprophyres or UML are adopted from Tappe *et al.* (2004, 2005, 2006).

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697 textures, as commonly observed for groundmass spinel
 698 in archetypal kimberlite and aillikite (Mitchell
 699 1986; Tappe *et al.* 2006), are notably absent in our
 700 orangeite samples. Spinel in the Impala orangeite
 701 dykes evolved from chromite to Ti-rich magnetite
 702 along a trajectory reminiscent of ‘Kimberlite Trend
 703 2’, although intermediate compositions are conspicu-
 704 ously absent (Fig. 5e, f). Such an evolutionary trend
 705 at increasing Fe^{2+}/Mg is typical for Kaapvaal craton
 706 orangeites, but also for cratonic lamproites from
 707 localities worldwide (Mitchell and Bergman 1991;
 708 Mitchell 1995). In contrast, groundmass spinel com-
 709 positional evolution in bona fide kimberlite mag-
 710 matic systems occurs along relatively constant and
 711 low Fe^{2+}/Mg , the so-called ‘Kimberlite Trend 1’
 712 (Mitchell 1986; Roeder and Schulze 2008). How-
 713 ever, recent work has shown that evolutionary
 714 paths in between the two major ‘kimberlitic’ spinel
 715 trends may be followed by both archetypal kimber-
 716 lites and aillikites (Tappe *et al.* 2009; Tappe *et al.*
 717 2014; Dongre and Tappe 2019). This observation
 718 renders spinel compositions less diagnostic for the
 719 identification of volatile-rich ultramafic magma
 720 types than previously thought. Regardless, the
 721 evolved orangeites from the Impala and Sibanye
 722 mining concessions apparently contain only Cr-rich
 723 spinel grains (87.2–91.6 Cr#) that overlap with the
 724 compositions of chromite from the more primitive
 725 olivine-rich orangeite dykes at Impala (82.1–90.3
 726 Cr#) (Fig. 5e, f).

728 *Andradite garnet.* Andradite-rich garnet occurs as
 729 <100 μm large groundmass grains within the
 730 evolved orangeite dykes of the Impala mining con-
 731 cession. The magmatic garnet grains exhibit very
 732 high CaO (32.0–32.5 wt%), TiO_2 (15.8–18.0 wt%)
 733 and FeO (17.5–18.2 wt%) contents at <0.01 wt%
 734 Cr_2O_3 (Fig. 6). They represent a solid solution series
 735 between the andradite, Ti-andradite and schorlomite
 736 garnet endmembers, with only a very small pyrope
 737 component. The Ti-rich andradite grains analysed
 738 here have similar compositions to the primary mag-
 739 matic Ti-andradite reported from the groundmass of
 740 ultramafic lamprophyres on the North Atlantic cra-
 741 ton in eastern Canada and West Greenland (Tappe
 742 *et al.* 2004; Tappe *et al.* 2006; Tappe *et al.* 2009),
 743 as well as from orangeites on the Dharwar craton
 744 in southern India (Dongre *et al.* 2016; Choudhary
 745 *et al.* 2020; Fig. 6). They are more Ti-enriched
 746 than the relatively pure andradite compositions
 747 reported from carbonatite dykes of the Premier kim-
 748 berlite pipe on the central Kaapvaal craton (Fig. 6),
 749 which were interpreted to have a secondary origin
 750 (Dongre and Tappe 2019). We note that Hammond
 751 and Mitchell (2002) reported an ‘unidentified
 752 Ca–Ti–Fe–silicate’ phase from the Swartruggens
 753 orangeite dyke swarm some 60 km west of the Bush-
 754 veld Complex on the central Kaapvaal craton

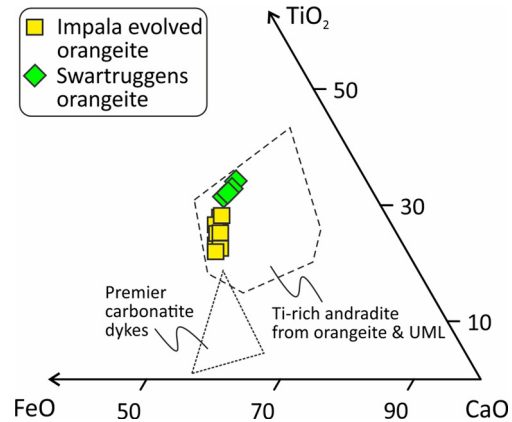


Fig. 6. Andradite garnet CaO–TiO₂–FeO compositions in Bushveld-intersecting evolved orangeite dykes from Impala. Data from a Swartruggens orangeite dyke represent the ‘unidentified Ca–Ti–Fe–silicate’ phase reported in Hammond and Mitchell (2002), which is interpreted here as magmatic Ti-rich andradite. The field for andradite garnet from carbonatite dykes of the Premier kimberlite pipe on the central Kaapvaal craton is adopted from Dongre and Tappe (2019). The andradite garnet compositional field for ultramafic lamprophyres and orangeites is based on the data reported in Tappe *et al.* (2004, 2006, 2009), Dongre *et al.* (2016) and Choudhary *et al.* (2020).

(Fig. 1), and this phase possibly represents Ti-rich andradite of a primary magmatic origin (Fig. 6). Regardless, it appears as though magmatic Ti-andradite is a relatively common accessory mineral in evolved orangeites and, thus, less diagnostic for ultramafic lamprophyres than previously thought (cf. Tappe *et al.* 2005).

Bulk rock major and trace element compositions

The Impala orangeite dykes have higher MgO (22.6–26.1 wt%), Cr (419–1694 ppm) and Ni (655–930 ppm) contents than the evolved orangeite dykes from this locality (15.1–20.2 wt% MgO, 415–1260 ppm Cr, 302–798 ppm Ni) and from the Sibanye mining concession (14.9–17.1 wt% MgO, 460–563 ppm Cr, 341–454 ppm Ni). The TiO_2 (0.78–1.24 wt%), CaO (6.24–8.69 wt%), Na_2O (0.05–0.68 wt%) and P_2O_5 (0.10–0.66 wt%) contents are notably lower for the Impala orangeite dykes compared with the evolved varieties from Impala (1.31–3.04 wt% TiO_2 , 8.33–13.4 wt% CaO, 1.06–1.65 wt% Na_2O , 0.54–1.72 wt% P_2O_5) and Sibanye (1.78–1.93 wt% TiO_2 , 10.4–11.8 wt% CaO, 1.00–1.61 wt% Na_2O , 0.89–0.97 wt% P_2O_5 ; Fig. 7). The other major and minor elements show

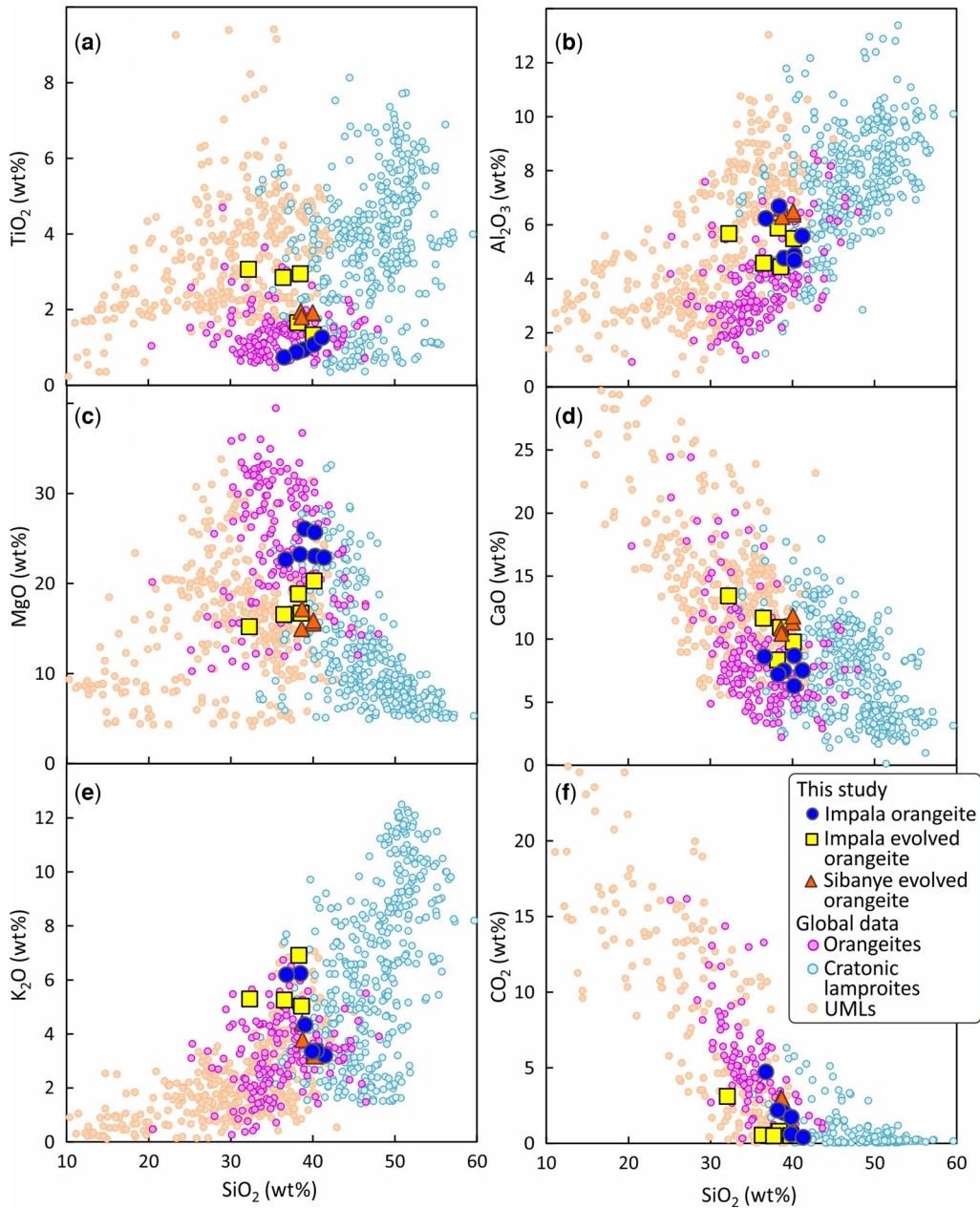


Fig. 7. Bulk rock major element variation diagrams for Bushveld-intersecting orangeite dykes. Data for comparisons with cratonic lamproites and ultramafic lamprophyres from localities worldwide are taken from the compilation of [Ngwenya and Tappe \(2021\)](#), which also contains the summary of data for type orangeites from the Kaapvaal craton as shown here.

broad compositional overlap between the three sample subsets. For example, the SiO_2 (32.2–41.2 wt%), Al_2O_3 (4.41–6.64 wt%) and CO_2 (0.25–4.71 wt%) contents are variably low, whereas the K_2O (3.15–6.90 wt%) concentration levels are collectively high

(Fig. 7b, e, f). The entire suite of samples from the Bushveld-intersecting orangeite dyke swarm has a truly ‘ultrapotassic’ character according to the criteria of [Foley *et al.* \(1987\)](#), with >3 wt% MgO and K_2O , respectively, at $\text{K}_2\text{O}/\text{Na}_2\text{O} \geq 2$ (2.0–86.2)

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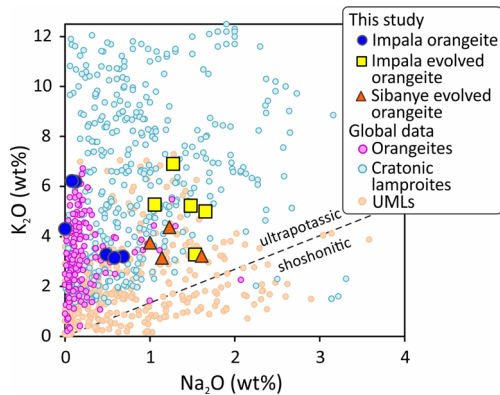


Fig. 8. Bulk rock K_2O – Na_2O variation in Bushveld-intersecting orangeite dykes. Data for comparisons with cratonic lamproites and ultramafic lamprophyres from localities worldwide are taken from the compilation of [Ngwenya and Tappe \(2021\)](#), which also contains the summary of data for type orangeites from the Kaapvaal craton. Note the exclusively ultrapotassic character of Kaapvaal orangeites including the dykes studied here. The ultrapotassic–shoshonitic rock divide is after [Peccerillo and Taylor \(1976\)](#).

([Figs 7 & 8](#)). Compared with other potassic ultramafic rock types, the Bushveld-intersecting ultrapotassic dykes show a good compositional match to

typical Kaapvaal craton orangeites ([Fig. 7](#)), which is in agreement with their mineral assemblages and compositions (see above). However, there is also some overlap in terms of major element compositions with cratonic lamproites and ultramafic lamprophyres from continental shields worldwide ([Figs 7 & 8](#)).

The orangeite dykes analysed here are strongly enriched in incompatible trace elements and they have similar primitive mantle normalized distributions compared with other Kaapvaal craton orangeites ([Becker and le Roex 2006](#); [Coe *et al.* 2008](#)) ([Fig. 9](#)). Whereas the olivine-rich Impala dykes fall at the lower end of the incompatible element concentration range of Kaapvaal orangeites, the olivine-poor *or* evolved orangeites from the Impala and Sibanye mining concessions cover the middle and upper ranges ([Fig. 9](#)), but the trace element patterns for the three sample subsets are very similar ([Figs 9 & 10](#)). A characteristic feature of Kaapvaal craton orangeites is their strong LILE (Cs–Rb–Ba–K) enrichment coupled to a marked relative depletion of certain high field strength elements (HFSE; Th–U–Nb–Ta; [Coe *et al.* 2008](#)), and the Bushveld-intersecting orangeite dykes studied here are no exception ([Fig. 9](#)). This geochemical feature also displays prominently in the Ba/Nb v. La/Nb diagram ([Fig. 11a](#)), in which the Impala dykes even extend the level of LILE/HFSE fractionation that is

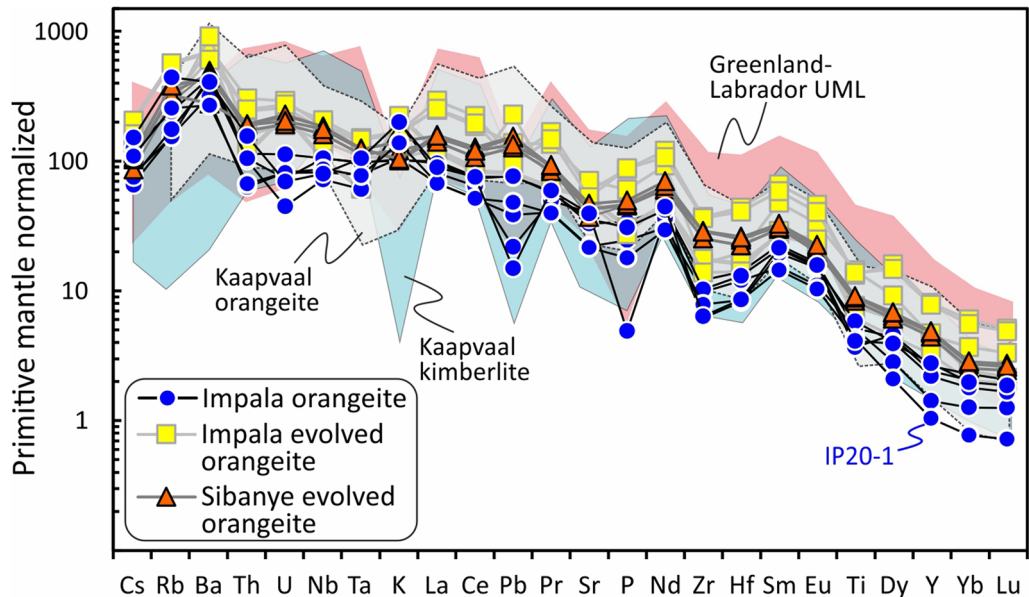


Fig. 9. Normalized incompatible element distributions of Bushveld-intersecting orangeite dykes. Fields for comparisons are based on data from the following literature sources: Kaapvaal kimberlites – [le Roex *et al.* \(2003\)](#), [Becker and le Roex \(2006\)](#), [Tappe *et al.* \(2020a, b\)](#); Kaapvaal orangeites – [Becker and le Roex \(2006\)](#), [Coe *et al.* \(2008\)](#); Greenland–Labrador UML – [Tappe *et al.* \(2006, 2008, 2011, 2017\)](#), [Nielsen *et al.* \(2009\)](#). The primitive mantle compositional estimate is after [Palme and O’Neill \(2003\)](#). UML, ultramafic lamprophyres.

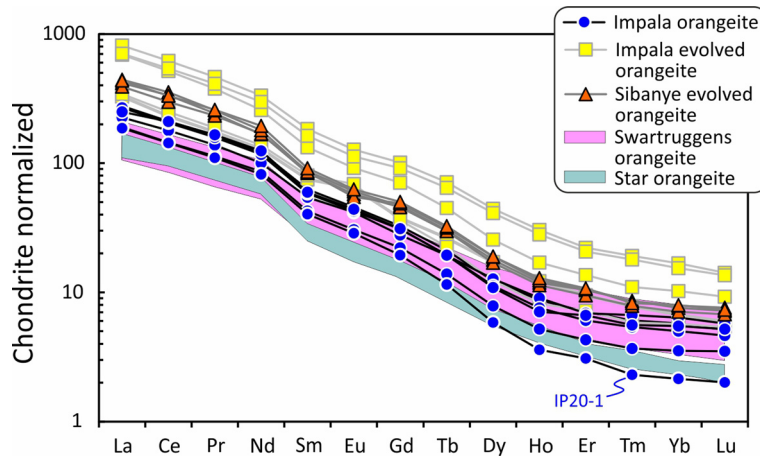


Fig. 10. Normalized rare-earth element (REE) distributions of Bushveld-intersecting orangeite dykes. Fields for the Swartruggens and Star orangeites are based on the data reported in Coe *et al.* (2008). The values for chondrite are after Palme and Jones (2003).

commonly observed for South African orangeites (Becker and le Roex 2006).

Bulk rock Sr–Nd–Hf isotopic compositions

The initial Sr–Nd–Hf isotopic compositions of the Bushveld-intersecting orangeite dykes from the central Kaapvaal craton are calculated for a magma emplacement age of 140 Ma (Table 1; Supplementary Material 1). The initial $^{87}\text{Sr}/^{86}\text{Sr}$ of the three sample subsets are all elevated and fall within a relatively narrow range between 0.707012 and 0.707410. The range displayed by the corresponding initial $^{143}\text{Nd}/^{144}\text{Nd}$ is relatively wide, with subchondritic initial ϵNd values from -10.6 to -5.8 , although the majority of samples fall between -10.6 and -9.6 (Fig. 12). The olivine-rich orangeite dykes from Impala tend to have more radiogenic Nd isotopic compositions compared with the olivine-poor *or* evolved orangeite dykes from the Impala and Sibanye mining concessions (Figs 12 & 13). The corresponding initial $^{176}\text{Hf}/^{177}\text{Hf}$ ratios show even larger variations, with subchondritic initial ϵHf values from -14.4 to -2.5 (Fig. 13). Similar to the Nd isotope systematics, the olivine-rich orangeite dykes from Impala have more radiogenic Hf isotopic compositions than the evolved varieties. The Bushveld-intersecting orangeite dykes fall firmly within the terrestrial Nd–Hf isotope array (Chauvel *et al.* 2008; Fig. 13), with initial $\Delta\epsilon\text{Hf}$ values between $+0.4$ and $+5.5$ (Table 1). The positively correlated Nd–Hf isotope systematics are in contrast to the commonly observed isotopic decoupling that characterizes many archetypal kimberlite sample suites from cratons worldwide (for a review see

Tappe *et al.* 2020a). In general, the combined Sr–Nd–Hf isotope systematics of the Bushveld-intersecting orangeite dykes reveal moderately to strongly enriched source compositions relative to primitive mantle, very similar to those of other orangeite occurrences from across the Kaapvaal craton (Figs 12 & 13).

Discussion

Mesozoic orangeite magmatism on the central Kaapvaal craton

The Swartruggens dyke swarm as a benchmark. The *c.* 145 Ma potassic ultramafic dyke swarm near Swartruggens is one of the best-studied orangeite occurrences on the central Kaapvaal craton and thus provides a benchmark against which to compare the petrogenesis of the Bushveld-intersecting orangeite dykes near Rustenburg (Fig. 1). The Swartruggens dykes, exploited at Helam Diamond Mine, have been variably referred to as kimberlites, lamprophyres, minettes, lamproites and, more recently, as orangeites (Fourie 1958; Skinner and Scott 1979; Mitchell and Bergman 1991; Mitchell 1995; Gurney and Kirkley 1996; Klump and Gurney 1998; Hammond and Mitchell 2002; McKenna *et al.* 2004; Coe *et al.* 2008). Although some studies used this terminology interchangeably, several workers pointed out that the Swartruggens dyke system is structurally and compositionally highly complex, comprising multiple intrusive phases and showing evidence of internal differentiation processes that may have created the highly variable petrographic characteristics and diverse mineralogy

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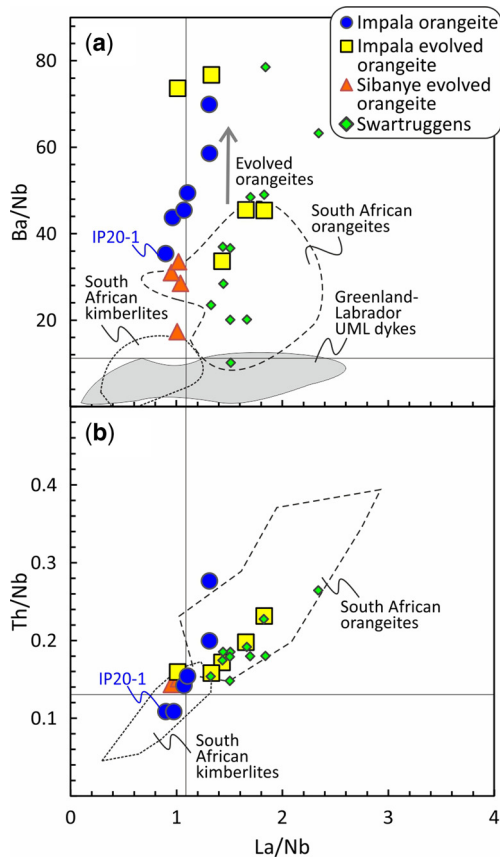


Fig. 11. Incompatible trace element ratios of Bushveld-intersecting orangeite dykes. (a) Ba/Nb and (b) Th/Nb v. La/Nb. Fields for comparisons are based on data from the following literature sources: South African kimberlites – *le Roex et al. (2003), Becker and le Roex (2006), Tappe et al. (2020a, b)*; South African orangeites including the Swartruggens dykes – *Becker and le Roex (2006), Coe et al. (2008)*; Greenland-Labrador UML dykes – *Tappe et al. (2006, 2008, 2011, 2017), Nielsen et al. (2009)*.

(e.g. *Skinner and Scott 1979; Mitchell 1995; Gurney and Kirkley 1996*). A key observation was that the dykes are unusually poor in or even devoid of olivine macrocrysts. This feature is mirrored by the typically low abundances of mantle-derived indicator minerals such as pyrope garnet and Cr-diopside, compared with much higher indicator mineral counts for diamondiferous orangeites on the western Kaapvaal craton (*Field et al. 2008*). The paucity of mantle-derived indicator minerals stands in contrast to the very high diamond grades in some of the Swartruggens dykes such as the Main Fissure with >200 carats per hundred tonnes of kimberlitic ore. However, diamond-free ‘fissures’ such as the Muil Dyke occur within a few metres of lateral distance

from the highly diamondiferous dykes (*Klump and Gurney 1998; McKenna et al. 2004*). On the basis of a detailed geochemical investigation, *Coe et al. (2008)* concluded that the compositionally highly variable Swartruggens orangeite dykes are related to a common parental magma, which suggests that differentiation processes have a strong control on primitive potassic magma type diversification upon ascent, including redistribution of any possibly existing diamond cargo.

The *c.* 140 Ma orangeite dykes on the western Bushveld Complex studied here are structurally and compositionally very similar to the Swartruggens dyke swarm some 60 km further west (*Fig. 1*). Our sample suite from these Bushveld-intersecting orangeite dykes allows for an evaluation of the role of crystal fractionation in the evolution of these potassic ultramafic magmas, which is an often-neglected process that operates in the plumbing system of deep-sourced volatile-rich melts on and around cratons worldwide.

Ultrapotassic magma evolution at Impala and Sibanye. The Bushveld-intersecting orangeite dykes comprise relatively olivine-rich and very olivine-poor varieties, illustrated by the example of drillcore intersections in *Figure 2c*. Importantly, both varieties contain compositionally overlapping Cr-spinel inclusions and discrete groundmass grains (*Fig. 5e, f*), which suggests a common parental magma to the entire orangeite dyke swarm on the central Kaapvaal craton, similar to the interpretation by *Coe et al. (2008)* for the Swartruggens dyke system.

Although a significant portion of the macrocrystic olivine in the olivine-rich orangeite dykes from Impala is derived from disaggregated cratonic mantle peridotites (i.e. xenocrystic), magmatic olivine is present in these rocks and reveals the diagnostically high Ni and elevated Ca contents commonly observed in olivine that crystallized from ultrapotassic magmas such as lamproites (*Prelevic et al. 2013; Jaques and Foley 2018*). These Ni- and Ca-enriched magmatic olivine compositions partially overlap with those of rare olivine microphenocrysts in the more evolved orangeite dykes. The evolved orangeite dykes contain olivine inclusions in groundmass phlogopite laths and they record magmatic evolution towards lower forsterite and Ni contents at elevated CaO (*Fig. 4*). Hence, olivine compositions in both the olivine-rich and olivine-poor or evolved orangeites provide evidence for magmatic olivine fractionation within this ultrapotassic system, superimposed on the effect of relatively minor xenocrystic olivine addition.

The geochemical variation diagrams in *Figure 14*, using MgO as a ‘differentiation’ index, show that the olivine-rich orangeite dykes from Impala have slightly higher MgO contents between 22.6 and

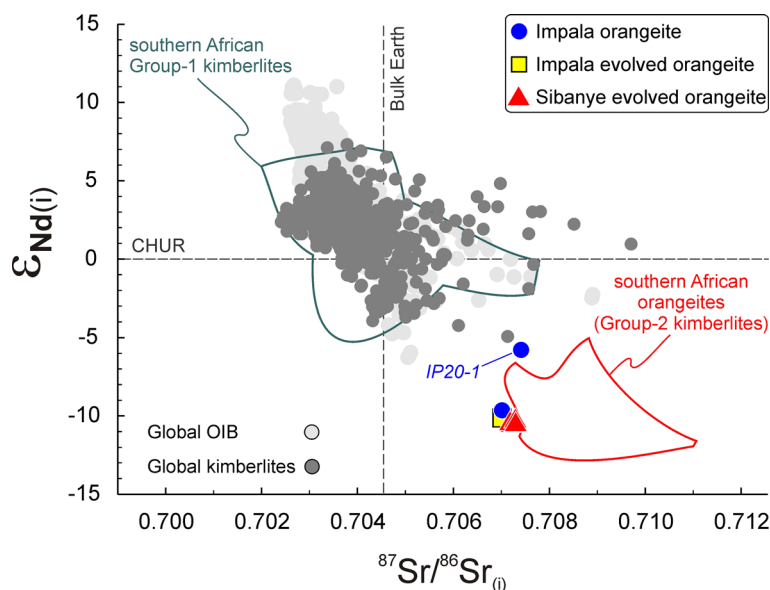


Fig. 12. Initial Sr–Nd isotopic compositions of Bushveld-intersecting orangeite dykes. Analytical uncertainties including a full propagation of errors are smaller than or similar to symbol size (Table 1). Fields for comparisons are based on data from the following literature sources: southern African Group 1 kimberlites – Smith (1983), le Roex *et al.* (2003), Nowell *et al.* (2004), Becker and le Roex (2006), Griffin *et al.* (2014), Sarkar *et al.* (2014), Tappe *et al.* (2020a, b); southern African Group 2 kimberlites or orangeites – Fraser *et al.* (1985), Nowell *et al.* (2004), Becker and le Roex (2006), Coe *et al.* (2008); global kimberlites including groundmass perovskite – Tappe *et al.* (2017); global OIB – Stracke (2012). CHUR, Chondritic uniform reservoir; OIB, oceanic island basalts.

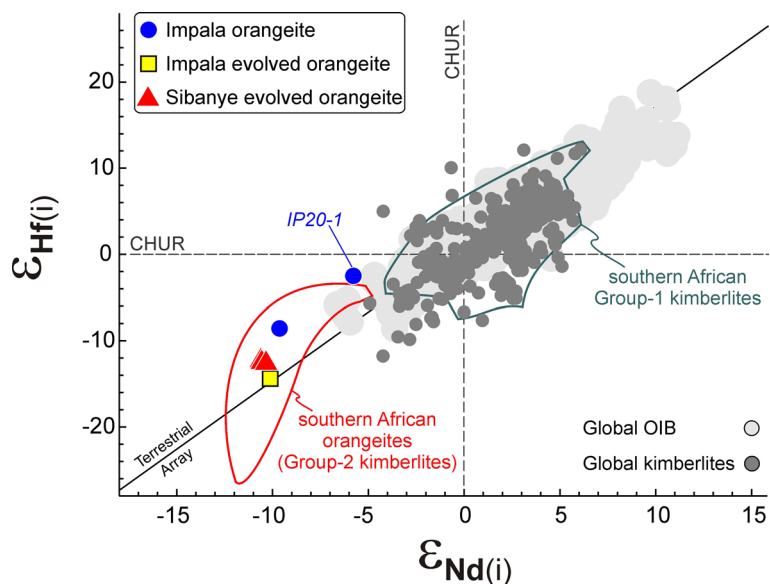


Fig. 13. Initial Nd–Hf isotopic compositions of Bushveld-intersecting orangeite dykes. Analytical uncertainties including a full propagation of errors are smaller than or similar to symbol size (Table 1). Fields for comparisons are based on data from the following literature sources: southern African Group 1 kimberlites – Nowell *et al.* (2004), Tappe *et al.* (2020a, b); southern African Group 2 kimberlites or orangeites – Nowell *et al.* (2004), Coe *et al.* (2008); global kimberlites – Tappe *et al.* (2017); global OIB – Stracke (2012). The terrestrial array regression line is taken from Chauvel *et al.* (2008).

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26.1 wt% compared with the estimated close-to-primary magma composition of the Swartruggens orangeite dyke swarm at c. 21 wt% MgO (Coe *et al.* 2008). Given the similar age and Sr–Nd–Hf isotopic compositions, plus the close geographic proximity on the central Kaapvaal craton (Fig. 1), it is reasonable to infer that a compositionally similar parental magma supplied both dyke systems. If correct, up to 10% of xenocrystic olivine addition to the parental Impala orangeite magma can explain

their higher MgO contents relative to the close-to-primary ultrapotassic magma composition of the Swartruggens dyke swarm (Fig. 14). Regarding the more evolved orangeite varieties from the Bushveld-intersecting dyke swarm, their MgO contents range from 20.2 down to 14.9 wt%, which suggests between 2 and 12% of olivine fractionation, arguably controlled by the removal of magmatic olivine. Alternatively, flowage-segregation and filter-pressing may have contributed to the observed

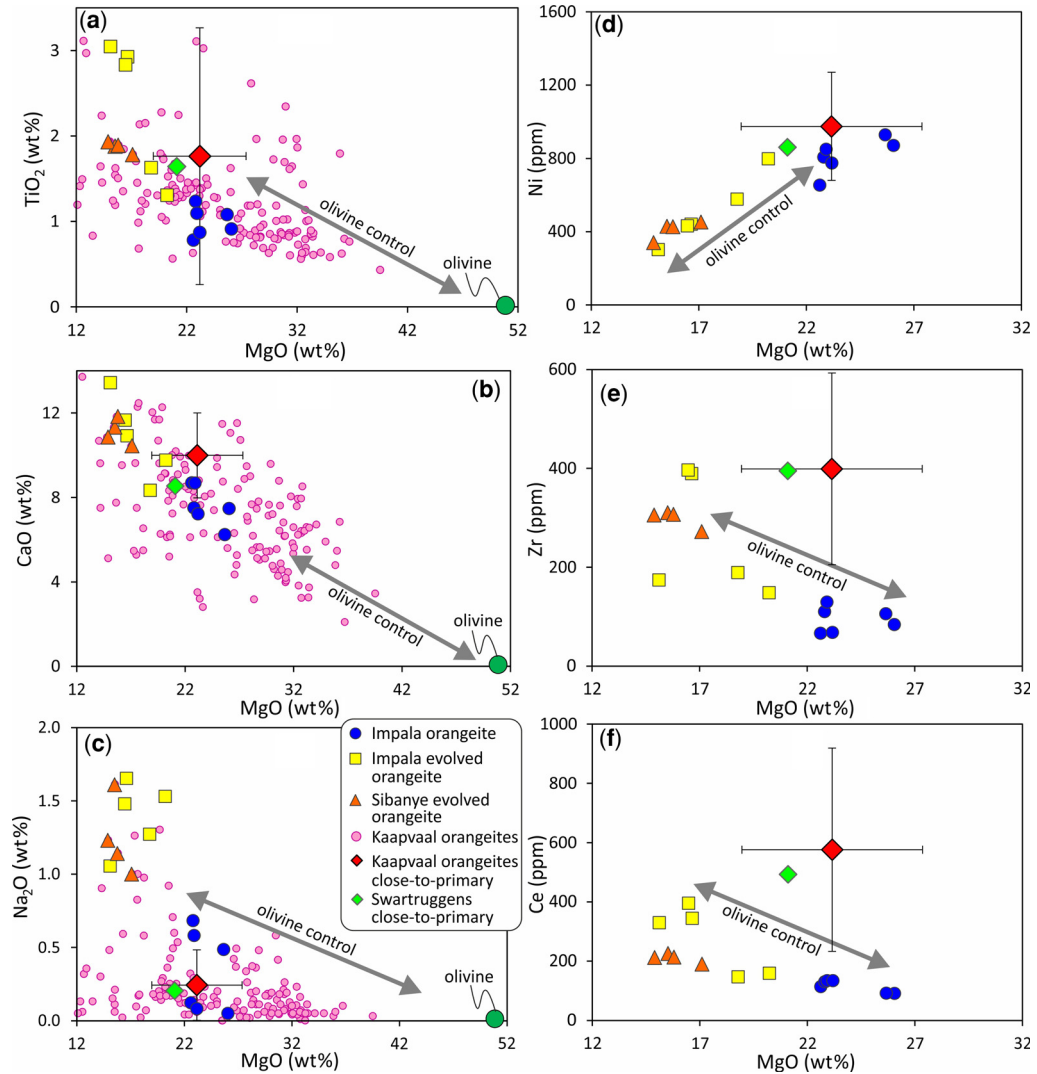


Fig. 14. Bulk rock major (a–c) and trace (d–f) element variation diagrams for Bushveld-intersecting orangeite dykes using MgO as a differentiation index. The data compilation for type orangeites from the Kaapvaal craton is taken from Ngwenya and Tappe (2021). The estimated close-to-primary magma compositions of Kaapvaal orangeites (error bars at 1-sigma) and Swartruggens orangeites are adopted from Becker and le Roex (2006) and Coe *et al.* (2008), respectively. The inferred olivine control line is guided by olivine compositions from the Impala orangeite dykes (Supplementary Material 1).

1103 geochemical ‘olivine control’, but such intra-dyke
 1104 differentiation mechanisms are difficult to quantify
 1105 in kimberlitic magmatic systems (Kjarsgaard *et al.*
 1106 2009; Zurevinski and Mitchell 2011; Tappe *et al.*
 1107 2014; Abersteiner *et al.* 2019). Regardless of the
 1108 circumstances of olivine control, it is generally
 1109 accepted that olivine and diamond crystals are trans-
 1110 ported in a similar manner within flowing and erupt-
 1111 ing kimberlitic magmas, mainly because of their
 1112 similar densities (Grunsky and Kjarsgaard 2008;
 1113 Field *et al.* 2009; Moss *et al.* 2010). Thus, identifica-
 1114 tion of olivine fractionation helps to explain the lack
 1115 of diamonds in the Bushveld-intersecting orangeite
 1116 dykes on the central Kaapvaal craton, a geological
 1117 domain that has a highly diamondiferous litho-
 1118 spheric mantle root (Viljoen *et al.* 2004; Field
 1119 *et al.* 2008; Korolev *et al.* 2018; Tappe *et al.* 2018;
 1120 Smart *et al.* 2021; Fig. 1).

1121 A firm olivine control on the petrogenetic link
 1122 between the Impala orangeites and the more evolved
 1123 varieties from Impala and Sibanye is also supported
 1124 by both compatible (e.g. Ni) and incompatible (e.g.
 1125 Zr, Ce) trace element variations with varying MgO
 1126 contents (Fig. 14d–f). Furthermore, the similar
 1127 primitive mantle normalized incompatible element
 1128 patterns of the olivine-rich and olivine-poor orange-
 1129 ite varieties, with a systematic offset to higher
 1130 trace element abundances in the samples with the
 1131 lowest modal olivine content, are best explained
 1132 by olivine fractionation (Fig. 9). This effect is also
 1133 evident in the chondrite-normalized REE diagram
 1134 (Fig. 10), where complete compositional overlap
 1135 exists between the most primitive Swartruggens
 1136 and Impala orangeite dykes, whereas the more
 1137 evolved varieties from Impala and Sibanye exhibit
 1138 up to an order of magnitude higher REE concentra-
 1139 tions, at broadly parallel element distributions for
 1140 all rock suites discussed here (Fig. 10).

1141 *K-metasomatized cratonic mantle sources.* Similar
 1142 to global anorogenic lamproite volcanism, type
 1143 orangeites from the Kaapvaal craton and adjoining
 1144 mobile belts are characterized by strongly enriched
 1145 Sr–Nd–Hf isotopic compositions (Nowell *et al.*
 1146 2004). The long-term enriched geochemical signa-
 1147 ture of the mantle source to orangeite magmatism
 1148 stands in contrast to the more primitive and moder-
 1149 ately depleted isotopic compositions of archetypal
 1150 kimberlites in southern Africa (Figs 12 & 13). The
 1151 striking isotopic difference between the two major
 1152 diamond ‘delivery’ systems on the Kaapvaal craton
 1153 forms the basis of petrogenetic models that invoke
 1154 ancient metasomatized lithospheric mantle sources
 1155 for orangeites as opposed to asthenospheric sources
 1156 for kimberlites (Smith 1983; Fraser *et al.* 1985;
 1157 Nowell *et al.* 2004; Tappe *et al.* 2020a, b).

1159 The Sr–Nd–Hf isotopic compositions of the
 1160 Bushveld-intersecting orangeite dykes from the

central Kaapvaal craton are similar to those of
 other orangeite localities in South Africa, including
 the Swartruggens dyke swarm (Coe *et al.* 2008;
 Figs 12 & 13). For the Impala and Sibanye dykes,
 the olivine-rich and more evolved orangeite varieties
 have broadly overlapping isotopic compositions, but
 we note that dyke IP20-1 shows significantly higher
 Nd and Hf isotope ratios at similarly radiogenic
 $^{87}\text{Sr}/^{86}\text{Sr}$, when compared with the other orangeite
 dykes (Figs 12 & 13). Dyke IP20-1 from Impala
 also shows more extreme HREE depletion and, alto-
 gether, these geochemical features suggest a more
 notable interaction of the ultrapotassic melt with
 much less enriched lithologies within the Kaapvaal
 cratonic mantle root (Figs 9–13). This material was
 probably clinopyroxene bearing, because among
 common cratonic mantle minerals it is the only
 phase that can significantly influence the Nd and
 Hf budgets of passing melts alike, retaining isotopic
 coupling (Griffin *et al.* 2000). Alternatively, the
 detected variability in the Nd–Hf isotopic composi-
 tions of the Bushveld-intersecting orangeite dykes
 may reflect lithospheric mantle source heterogeneity.
 For example, MARID-type metasomes have fre-
 quently been invoked as source components to
 potassic ultramafic magmas (Foley 1992; Ulmer
 and Sweeney 2002; Tappe *et al.* 2008; Smart *et al.*
 2019), and isotopic investigations of such heavily
 K-metasomatized lithospheric mantle materials
 revealed extreme heterogeneity down to the centi-
 metre scale (Kramers *et al.* 1983; Grégoire *et al.*
 2002; Choukroun *et al.* 2005; Fitzpayne *et al.* 2019).

*A ‘stored’ subduction signature in Kaapvaal cra-
 tonic mantle metasomes.* Type orangeites from
 South Africa collectively exhibit a strong Cs–Rb–
 Ba–K enrichment that is coupled to a relative deple-
 tion in Th–U–Nb–Ta (Becker and le Roex 2006; Coe
et al. 2008; this study). This distinctive trace element
 signature is associated with remarkably low TiO₂
 contents of <2 wt% for the most primitive orangeite
 varieties (Figs 7a & 9), much lower than what is typi-
 cally observed for other potassic ultramafic magma
 types on cratons, such as anorogenic lamproites and
 aillikites, which reach up to 8 wt% TiO₂ (Figs 7a &
 9). The high LILE/HFSE ratios of Kaapvaal craton
 orangeites are reminiscent of a classic subduction
 zone signature (Fig. 11), where residual titanate min-
 erals (e.g. rutile) in the downgoing oceanic slab
 retain in particular Ti–Nb–Ta while the LILE (and
 LREE) become enriched in extracted melts/fluids
 and enter the lithospheric mantle of the overriding
 tectonic plate (Foley and Wheller 1990; Pearce and
 Peate 1995).

As Coe *et al.* (2008) pointed out, the Kaapvaal
 craton was last subjected to subduction and collisional
 events in its immediate vicinity during the Mesoproterozoic
 Namaqua–Natal orogeny at

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1161 1220–1090 Ma (Spencer *et al.* 2015). Although the
1162 geometry of these ancient subduction zones is far
1163 from understood (Jacobs *et al.* 2008; Van Schijndel
1164 *et al.* 2020), it is reasonable to assume that oceanic
1165 slab remnants temporarily stalled beneath the thick
1166 Kaapvaal lithosphere and released small volumes
1167 of metasomatic melts/fluids by which the wide-
1168 spread ‘subduction’ signature was introduced into
1169 the lowermost cratonic mantle. Structural responses
1170 of the Kaapvaal lithosphere to these Mesoprotero-
1171 zoic collisions are evident from its burial and unroof-
1172 ing history (Baughman and Flowers 2020), and also
1173 from the prolonged emplacement history of the Pre-
1174 mier kimberlite cluster in the central part of the cra-
1175 ton between *c.* 1155 and 1135 Ma (Tappe *et al.*
1176 2020a).

1177 A direct origin of potassic ultramafic magmas
1178 from slab remnants, including subducted sedimentary
1179 components (Murphy *et al.* 2002; Rapp *et al.*
1180 2008), within the deeper convecting mantle is
1181 excluded here. The main reason for this is that the
1182 phlogopite/richterite-carbonate bearing peridotitic
1183 source assemblages required to explain orangeites
1184 and related rocks are largely restricted to the pres-
1185 sure–temperature– fO_2 conditions of the lower cra-
1186 tonic mantle lithosphere (Tappe *et al.* 2007; Foley
1187 2011). Ulmer and Sweeney (2002) pointed out that
1188 even under such cratonic mantle conditions, mixed
1189 phlogopite/richterite–carbonate assemblages are
1190 only metastable and probably do not persist for pro-
1191 longed periods of geological time. This led Tappe
1192 *et al.* (2008) to propose a model in which ancient
1193 phlogopite–richterite-bearing cratonic mantle meta-
1194 somes are fused by small volumes of infiltrating
1195 CO_2 -rich melts from the underlying asthenosphere,
1196 giving rise to K-rich carbonated ultramafic silicate
1197 magmas such as aillikites and orangeites.

1198 An inherited subduction signature manifested
1199 in cratonic mantle metasomes is supported by
1200 $^{40}Ar/^{39}Ar$ age results for phlogopite from strongly
1201 metasomatized peridotites along the southern Kaap-
1202 vaal craton margin, yielding ages of 1.25–1.0 Ga
1203 (Hopp *et al.* 2008). In contrast, U–Pb geochronology
1204 of zircon in MARID-type xenoliths from the western
1205 Kaapvaal craton revealed exclusively Late Mesozoic
1206 ages (Konzett *et al.* 1998; Giuliani *et al.* 2015; Hoare
1207 *et al.* 2021), but it is uncertain whether these dates
1208 represent the timing of K-rich mantle metasomatism
1209 or simply the time of isotopic closure as a conse-
1210 quence of lithospheric mantle cooling after a short-
1211 lived, yet impactful regional heating event during
1212 the Early Mesozoic (Bell *et al.* 2003). Regardless
1213 of the timing of lithospheric mantle K-enrichment,
1214 it is remarkable how uniform this enriched domain
1215 apparently was in terms of its mineralogical and geo-
1216 chemical compositions, as evident from the similar
1217 incompatible element systematics (Figs 9–11) and
1218 Sr–Nd–Hf isotopic compositions (Figs 12 & 13) of

type orangeites. These deep-sourced ultrapotassic
magmas erupted in pulses on and off the Kaapvaal
craton, across an area that was 1000×750 km in
size, during progressive breakup of the Gondwana
superterrane between 200 and 110 Ma (Tappe *et al.*
2018).

Orangeite v. lamproite terminology: what’s in a name?

The potassic ultramafic dykes on the central Kaap-
vaal craton in South Africa highlight the problems
surrounding naming conventions for kimberlites
and related rocks. They have been variably referred
to as kimberlites, lamprophyres, minettes, lamproites
and orangeites (Skinner and Scott 1979; Leeb-Du
Toit 1986; Mitchell and Bergman 1991; Mitchell
1995; Gurney and Kirkley 1996; Hammond and
Mitchell 2002; McKenna *et al.* 2004; Coe *et al.*
2008), even by the same authors over short periods
of time (compare Mitchell and Bergman (1991,
p. 72) with Mitchell (1995, p. 30)). There appears
to be wide consensus within the alkaline rock petro-
logy community that the term Group 2 kimberlite
should be abandoned in favour of these potassic
rocks being called orangeites (Mitchell 1995), but
in recent years there has been some development to
fully assimilate the term orangeite into the lamproite
clan. What began with notions about orangeites
being the ‘lamproite variety Kaapvaal’ (Mitchell
2006) has now culminated in orangeites being con-
sidered as ‘ CO_2 -rich lamproites’ or ‘carbonate-rich
lamproites’ (Pearson *et al.* 2019). This development
is regrettable for three main reasons: first, lamproites
from classic potassic igneous provinces around the
globe are mostly free from primary magmatic car-
bonate but tend to have a silicate glass matrix
instead, such that the term CO_2 -rich lamproite pre-
sents an oxymoron in a resemblance classification
scheme that relies on similarities to standard mem-
bers (Foley *et al.* 1987). Second, type orangeite
from the Kaapvaal craton commonly comprises
primitive and evolved varieties, the latter of which
may approach CO_2 concentration levels of zero
(Fig. 7f) and thus cannot be referred to as CO_2 -rich
lamproites. Third, the introduction of a carbonate-
rich lamproite subgroup obstructs the classification
of ultramafic lamprophyres (Tappe *et al.* 2005),
because the carbonate-rich lamprophyre variety aillikite
would, by the same logic that was applied to type
orangeite, also belong to the newly proposed lamproite
class. Recently, Sarkar *et al.* (2018) referred
to carbonate-rich potassic ultramafic rocks, akin to
aillikite or orangeite, as lamproites, which highlights
the need for a more coherent classification scheme
that can satisfactorily accommodate the petrographical
variability of mantle-derived volatile-rich igneous
rocks (Mitchell and Tappe 2010).

Here we argue to maintain the status quo of orangeite as a discrete type of mantle-derived potassic magma with occurrences worldwide (Chalapathi Rao *et al.* 2011; Donatti-Filho *et al.* 2013; Kargin *et al.* 2014; Dalton *et al.* 2019; Chalapathi Rao *et al.* 2020; Choi *et al.* 2021; Krmíček *et al.* this volume, in press). The recognition that orangeites or Group 2 kimberlites may show closer compositional affinities to cratonic lamproites and aillikites than to archetypal Group 1 kimberlites was a milestone in alkaline rock petrology (Dawson 1987; Mitchell 1995), clearly serving a petrogenetic purpose (Tappe *et al.* 2005). However, modifications of and changes to the terminology of igneous rocks without any obvious advance in our understanding of magma petrogenesis are not endorsed here. Although the names of rocks do not affect what they really are and how they formed – alkaline rock terminology is highly complex and does not appear to benefit from incoherently applied changes – this only complicates matters further.

Conclusions

- The mineralogical and geochemical compositions of 15 individual potassic ultramafic dykes, intersecting the western Bushveld Complex, show that these Mesozoic minor intrusions represent ‘typical’ Kaapvaal craton orangeites, formerly known as Group 2 kimberlites. A strong olivine control is recognized in the magmatic evolution of the orangeite dykes, with olivine-poor more evolved varieties petrographically resembling lamprophyres. Olivine fractionation and physical sorting are held responsible for the absence of significant quantities of diamond, even though the orangeite magmas traversed one of the richest diamond repositories in the world, as represented by the central Kaapvaal cratonic mantle lithosphere.
- The enriched Sr–Nd–Hf isotopic compositions of the Bushveld-intersecting orangeite dykes suggest a magma origin from ancient K-metasomatized mantle lithosphere, similar to the source of other Kaapvaal craton orangeites. Despite the highly enriched incompatible trace element contents, the orangeite samples studied exhibit pronounced depletions in Th–U–Nb–Ta as well as Ti, resulting in high LILE/HFSE ratios. Such a subduction zone geochemical signature is common among the Mesozoic orangeites from across the Kaapvaal craton. The ‘stored’ subduction signature is best ascribed to K-rich metasomatism of the cratonic mantle lithosphere in the vicinity of Mesoproterozoic collision zones associated with the Namaqua–Natal orogeny during Rodinia supercontinent formation.

- Our study reinforces that orangeites represent a discrete type of mantle-derived potassic magma. Although compositional similarities with cratonic lamproites and ultramafic lamprophyres do exist, type orangeite from South Africa is sufficiently distinct such that different magma formation processes are implied.

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Data availability All data generated or analysed during this study are included in this published article (and its supplementary information files).

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