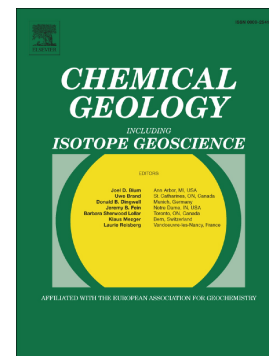


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Belingwe komatiites (2.7 Ga) originate from a plume with moderate water content, as inferred from inclusions in olivine.

Asafov E.V.<sup>1</sup>, Sobolev A.V.<sup>1,2</sup>, Gurenko A.A.<sup>3</sup>, Arndt N.T.<sup>2</sup>, Batanova V.G.<sup>1,2</sup>, Portnyagin M.V.<sup>1,4</sup>, Garbe-Schonberg D.<sup>5</sup>, Krasheninnikov S.P.<sup>1</sup>

1. Vernadsky Institute of Geochemistry and Analytical Chemistry, Russian Academy of Sciences, 19 ul. Kosygina, Moscow 119991, Russia.
2. ISTERre, Univ. Grenoble Alpes, CNRS, IRD, IFSTTAR, 38000 Grenoble, France.
3. Centre de Recherches Pétrographiques et Géochimiques (CRPG), UMR 7358, Université de Lorraine, 54501 Vandoeuvre-lès-Nancy, France.
4. GEOMAR Helmholtz Centre for Ocean Research Kiel, Wischhofstrasse 1-3, 24148 Kiel, Germany.
5. CAU Kiel University, Institute of Geosciences, Ludewig-Meyn-Strasse 10, 24118 Kiel, Germany.

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Corresponding author: Mr. Evgeny Vladimirovich Asafov, M.D.

Corresponding Author's Institution: Vernadsky Institute of Geochemistry and Analytical Chemistry, Russian Academy of Sciences, 19 ul. Kosygina, Moscow 119991, Russia.

## Abstract

Major and trace elements, and volatile components have been measured in melt inclusions in olivine from fresh 2.7 Ga old komatiites from the Reliance Formation of the Belingwe Greenstone Belt, Zimbabwe. Reconstructed compositions of melt inclusions contain 20-23.5 wt.% MgO and up to 0.3 wt.% H<sub>2</sub>O; these compositions probably represent those of the erupted lava. In inclusions in relatively evolved (low Fo) olivines, an excess of Na<sub>2</sub>O, CaO, Li, La, Cu, Rb, Y, Sc as well as volatile components (H<sub>2</sub>O, F, Cl and S) relative to other highly incompatible elements is attributed to assimilation of seawater altered mafic material. No assimilation signature is observed for the most primitive melt inclusions hosted in the magnesium rich olivines. The primary melt composition, estimated using melt inclusions in the most magnesian olivine (Fo<sub>93.5</sub>), contains up to 27.5 wt.% MgO and ca. 0.2 wt.% H<sub>2</sub>O. The presence of H<sub>2</sub>O slightly depressed the liquidus temperature to ca. 1513°C. Our results suggest formation of the Belingwe komatiite magma at ca. 7 GPa pressure and ca. 1790 °C temperature in a mantle plume. The plume picked up water and probably chlorine through interaction with a hydrous transition mantle zone in the way similar to that previously proposed by Sobolev et al. (2016) for komatiites in Canada.

## 1. Introduction

Komatiites are the most MgO-rich and hottest volcanic rocks on Earth. Their anhydrous liquidus temperatures are estimated to be about 1600°C, but their eruption temperatures and the mode of origin are uncertain because the amount of volatiles initially dissolved in komatiite magmas is poorly constrained (Arndt et al., 1998; Parman et al., 2004; Sobolev et al., 2016). The water contents in komatiite magmas have been debated since Viljoen and Viljoen (1969) first described komatiites. Most researchers now agree that komatiites result from melting in essentially dry and exceptionally hot mantle plumes (Arndt et al., 1998; Campbell et al., 1989; Connolly et al., 2011), and the alternative point of view – that komatiites are hydrous magmas that formed in subduction settings (Brooks and Hart, 1974; Allègre, 1982; Grove et al., 1997; Parman et al., 2004) – is now not widely accepted (e.g. Arndt et al. 2008; Berry et al., 2008).

Several attempts have been made to determine the concentration of water in komatiites. Parman et al. (2004) employed the compositions of pyroxenes to infer an extremely wet komatiite magma that originated in a supra-subduction setting, but most recent studies found far lower water contents. For example, Danyushevsky et al. (2002) estimated 0.2-0.3 wt.% of H<sub>2</sub>O in the melt inclusions in olivine from Belingwe Greenstone Belt komatiites. Berry et al. (2008) measured low (0.10±0.02) Fe<sup>3+</sup>/ΣFe ratios in the melt inclusions using X-ray absorption near-edge structure, and used this to rule out oxidation and related diffusional H-loss from melt inclusions. Connolly et al. (2011) then used H<sub>2</sub>O/Ce ratios to propose anhydrous and exceptionally high-temperature initial melts. On this basis, these authors proposed that the measured water contents are those of the original melt. However, the study by Shimizu et al. (2001) of melt inclusions in chrome spinel crystals from the Belingwe komatiites suggested the presence of up to 1.7 wt.% water in trapped melts and 0.8-0.9 wt.% H<sub>2</sub>O in the primary komatiite magma. Finally, direct SIMS measurements of H<sub>2</sub>O in melt inclusions in exceptionally magnesian olivine (up to Fo<sub>94.3</sub> where Fo = Mg/(Mg+Fe) at%), independently supported by Sc-Y olivine-melt geothermometry, revealed up to 0.6 wt% H<sub>2</sub>O in the primary melt of 2.7 Ga Abitibi komatiites (Sobolev et al., 2016).

Experimental approaches (Inoue and Sawamoto, 1992; Kawamoto et al., 1996) and combinations of experimental and petrological methods to study igneous minerals in komatiites (Allègre, 1982; Arndt et al., 1998; Arndt et al., 2008; Parman et al., 1997; Stiegler et al., 2012; Stone et al., 1997) have also been applied. Based on these studies, water contents in parental komatiite melts are now estimated to range from essentially zero up to 3wt.%.

Formation of komatiites in a hydrous mantle plume was originally proposed by Inoue (1994) and Hirose and Kawamoto (1995). Shimizu et al. (2001) supported the same model using

data from melt inclusions in chrome spinel and more recently, Sobolev et al. (2016) developed a similar idea using melt inclusions in olivine in Abitibi komatiites. The latter authors proposed that water was entrained into a mantle plume as it traversed the mantle transition zone deep in the Archean mantle. High Cl-concentrations (up to 0.2 wt%) measured in a series of melt inclusions implied a contribution from seawater. These Cl-enriched melt inclusions were found even in the most magnesium rich olivine (Fo<sub>94.1</sub>), suggesting that the contamination occurred at an early stage of magma fractionation, thereby precluding correct estimation of the initial water contents using the composition of late-stage igneous minerals such as pyroxene or amphibole, or H<sub>2</sub>O contents of the evolved melt inclusions in spinel. Shimizu et al.'s, (2001) estimates of water content of Belingwe komatiites were extrapolated from the compositions of melt trapped in chrome spinel that crystallized from evolved magma and thus these water contents do not represent those of the primary melt.

Here we report new data for two representative samples of 2.7 Ga Ngezi Group komatiites from the Belingwe Greenstone Belt, Zimbabwe. Using an electron microprobe (EPMA), laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) and an ion microprobe (SIMS), we obtained data on major and trace elements and volatile components (H<sub>2</sub>O, F, Cl, S, CO<sub>2</sub>) in the melt inclusions and in their host olivines. We also analyzed trace elements in the whole rocks, as well as the compositions of co-existing spinel and olivine in thin sections by ICP-MS and electron microprobe. The main goal of our study was to determine the pristine composition (i.e. unmodified by alteration, contamination and fractional crystallization) of the parental melts of these komatiites. We also estimate the volatile contents in the parental melts, discuss their chemical evolution (including the temperature and pressure of origin), and finally place new constraints on the deep mantle processes responsible for the origin of Archean komatiites.

## 2. Geological background

The Belingwe Greenstone Belt (Fig. 1) is located in the southern part of the Zimbabwe Craton, to the north of the Limpopo orogenic belt and directly east of the Great Dyke. This belt is a tight syncline, unconformably underlain by 3.5-3.6 Ga tonalitic crust, the Shabani Gneiss Complex (Bickle et al., 1975) and by greenstones of similar age (Bickle et al., 1977; Nisbet et al., 1977). The Belingwe Greenstone Belt comprises the Lower Greenstones (2.9 Ga Mtshingwe Group), which consists of three sedimentary formations whose deposition environment ranges from probable beach to deeper water conditions, and the unconformably overlying Upper Greenstones (2.7 Ga Ngezi Group). Dupré & Arndt (1990) reported a Pb-Pb age of 2692±9 Ma

for basalt within this unit. The Upper Greenstones comprise (from base to top) 300m of shallow-water sedimentary rocks of the Manjeri Formation, the predominantly basaltic and komatiitic 0.5-1-km-thick Reliance Formation, the 6-km-thick basaltic and andesitic Zeederbergs Formation, and the uppermost Cheshire Formation of shallow-water sedimentary rocks. There is strong evidence that the volcanic successions of the Reliance and Zeederbergs Formation erupted in shallow submarine conditions onto submerged continental crust (Bickle et al., 1975, Nisbet et al., 1977).

In this project we studied two olivine cumulates (samples Z4 and Z6) from the Reliance Formation. The samples were collected by N. Arndt in outcrops of Tony's flow, one of the best-exposed komatiite flows (Bickle et al., 1993). In addition we used new LA-ICP-MS data on the olivine phenocrysts from olivine cumulates MZ-4 from the same lava series studied previously by (Sobolev et al., 2007). By Archean standards, the samples are only moderately altered, as is reflected by their relatively low water content (ca. 3%), by totally serpentinized groundmass but partially preserved olivine phenocrysts, by the presence of serpentine and carbonate veins, and by oxygen, hydrogen and Rb-Sr isotopic systematics (Bickle et al., 1993). According to their Nd and Pb isotope compositions, the selected samples do not show evidence of continental crust contamination (Chauvel et al., 1993). Fresh chrome spinel and fresh relicts of olivine occur in the samples. Some of these contain crystallized melt inclusions, which we investigated in the course of this study.

### 3. Sample preparation

All sample preparation was performed at the Vernadsky Institute of Geochemistry and Analytical Chemistry Russian Academy of Sciences, Moscow, Russia. The hand specimens of Z6 and Z4 komatiites from the cumulate layers were crushed and several small pieces were used to prepare polished thin sections. Thin sections were then used for electron microprobe analysis of olivine, and of melt inclusions in olivine and spinel. Fragments with minimal signs of alteration were manually crushed in a steel mortar and ground to powder using an agate mortar. A portion of the powder was used for solution ICP-MS analysis and the remaining parts of the sample were separated into size fractions of <0.1 mm, 0.1-0.25 mm, 0.25-0.5 mm, from which fragments of fresh olivine containing melt inclusions were hand-picked under a binocular microscope.

Most melt inclusions were found to be concentrated near the grain cores. Melt inclusions typically have spherical or ellipsoidal shapes. They are up to 200  $\mu\text{m}$  in diameter and comprise glass, a shrinkage bubble and tiny crystals of spinel, olivine and pyroxene (Fig. 2). In order to

homogenize the partially crystallized melt inclusions, the separated fresh and clean olivine grains were heated at 1250-1350°C for 5 minutes in a 1 atmosphere, high-temperature vertical furnace in a C-O-H atmosphere with buffered oxygen fugacity corresponding to the quartz-fayalite-magnetite buffer (QFM), then rapidly quenched to glass in water. The olivine grains were mounted in epoxy resin, then ground and polished to expose the glass inclusions. Quenched melt inclusions contained glass, spinel crystals and a shrinkage bubble. Inclusions larger than 20  $\mu\text{m}$  were selected for chemical analysis. Smaller inclusions were ignored because their compositions could be affected by a boundary layer effect, which is expressed in elevated Al concentrations and inconsistent Si/Mg ratios.

Initially altered and/or cracked melt inclusions are easily recognized after heating because they have lost most of their volatiles due to direct connection with 1 atmosphere external pressure at high temperatures.

## 4. Methods

### 4.1. EPMA

Major element compositions of the melt inclusions and host olivines in the epoxy mounts, and the compositions of coexisting olivine-spinel pairs in thin sections, were measured by electron microprobe (EPMA). All samples were analyzed using the JEOL JXA 8230 at ISTERre, Grenoble, France. Operating conditions were 15 to 25 kV accelerating voltage and 900 nA probe current for olivine in thin sections and 300 nA probe current for separated olivine grains in epoxy mounts using protocol described in Batanova et al (2015). The accuracy of the data was controlled by measuring the reference San Carlos olivine during the analytical session (Batanova et al., 2015; Sobolev et al., 2007). For each olivine grain, two or more analyses were performed and the uncertainties were calculated as standard deviations. The melt inclusions were analyzed for the major elements, trace elements (titanium, potassium, manganese, chromium and phosphorus) and volatiles (chlorine and sulfur) at 15 kV voltage and 12 nA current using protocol described in Sobolev et al (2016). The analytical precision was assessed by including reference glasses GOR-132G (Jochum et al., 2006) and VG-2 (Jarosevich et al., 1980) measured as unknown in the analytical session. GOR-132G was used to monitor contents of major and trace elements and VG-2 for concentrations of Cl and S. The number of analyses performed for each inclusion ranged from 1 to 4, depending on the inclusion size. Uncertainties were estimated as standard deviations or, in the case of single analyses, the standard deviations for the reference material. Spinel was analyzed in thin sections at 20 kV voltage and 50nA current with a chromite standard as reference material (Batanova et al., 2011). The spinel crystals have skeletal habits

and only one measurement per grain was possible. Uncertainty was estimated as the standard deviation for reference chromite. ZAF corrections were used to eliminate matrix effects. Compositions of olivine are listed in Table 1, unprocessed EPMA data on the melt inclusions is listed in the Table A.2 (online supplementary material), data on the melt inclusions corrected for post entrapment crystallization (see Section 4.5 for details) and host olivines are given in the Table 2 and the compositions of spinel and coexisting spinel-olivine pairs are listed in Table A.1 and Table A.3 respectively (Online supplementary material).

#### **4.2. SIMS**

The CAMECA IMS 1280 HR2 and IMS 1270 E7 secondary ion mass spectrometers (SIMS) in CRPG, Nancy, France, were used to analyze CO<sub>2</sub>, H<sub>2</sub>O, Cl, S and F in the melt inclusions. A set of standards (ALV981-R23, ALV519-4-1, 30-2, 40-2, KL2-G, CY8406-02 and San Carlos olivine) was used to define calibration lines. The analytical details are provided in Sobolev et al. (2016).

#### **4.3. LA-ICP-MS**

We analyzed trace-element concentrations in melt inclusions and host olivine by LA-ICP-MS using an Agilent 7500s quadrupole mass-spectrometer coupled with a 193 nm Excimer laser ablation system GeoLas Pro (Coherent) at the Institute of Geosciences of the Christian-Albrecht University in Kiel, Germany. Laser spot size was 24- $\mu$ m and 32- $\mu$ m for inclusions and olivine, respectively. Ablation was performed with laser pulse frequency of 10 Hz and laser fluence of 10 J cm<sup>-2</sup>. Olivine analyses were performed on three spots surrounding each inclusion, on areas pre-ablated for 3 s to reduce surface contamination. All analyses were performed in a large volume ‘Zürich’ ablation cell. Carrier gas was He (~1 L min<sup>-1</sup>) with addition of H<sub>2</sub> (0.014 L min<sup>-1</sup>), which were mixed with Ar (0.85 L min<sup>-1</sup>) before introduction into spectrometer. Analyses were performed in time-resolved mode and included 20 s background measurement followed by 20 s sample ablation and signal measurement. Dwell time was 10 ms for all elements. Detailed description of the method is given in Sobolev et al. (2016). All trace element contents are listed in the Table 2.

#### **4.4. Solution ICP-MS**

Bulk rocks were analyzed by solution ICP-MS on a Thermo X series II at ISTerre, Grenoble, France following the (Chauvel et al., 2011) protocol. For analysis, 100 mg of rock powder were dissolved in Savillex beakers and Paar bombs in mixture of high purity HF and HNO<sub>3</sub>. Final solution in 2% HNO<sub>3</sub> was diluted by factor of 5000. We used BR-24 as an external

standard and a set of internal standards (BHVO-2, BCR-2, Bir-1a, BEN and UBN). A five-element spike (Be, Ge, In, Tm, Bi) was added to the solution to correct for machine drift. Trace-element concentrations were calculated using methods published by Barrat et al. (1996) and Ionov et al. (2006). Operating conditions were: RF power 1400W, coolant gas 13L/min, Aux gas 0.8L/min, nebulizer gas 0.9L/min, pump speed 1mL/min, T°C cyclonic chamber = 2°C, Time uptake = 90s. Three runs were performed per analysis with the dwell time 10ms per element. The ICP-MS was washed out with an acid solution (HNO<sub>3</sub> 5% + HNO<sub>3</sub> 2% + HNO<sub>3</sub> 2%) three times after every analysis

#### 4.5. Data processing

Compositions of melt inclusions are listed in the Table A.2 (supplementary online material). Cracked inclusions that had lost volatiles during or before heating were identified by low Na, S and Cl concentrations and filtered out. As the compositions of the melt inclusions were affected by post entrapment crystallization of olivine on the walls of the melt inclusions and Fe-Mg diffusion between host olivine and included melt, we reconstructed the composition of originally trapped melts using the reverse Fe-Mg diffusion method (Sobolev and Danyushevsky, 1994). This method is based on the olivine-melt equilibrium model of (Ford et al., 1983) and uses the original FeO content of the melt. The latter was determined using the Fo content of the host olivine by modeling the crystallization of sample Z11 (Bickle et al., 1993). This sample was taken from chilled margin of komatiite flow and is thought to represent the quenched komatiite melt composition. The calculations were performed with the Petrolog3 software (Danyushevsky and Plechov, 2011).

Trace element and volatile element concentrations of the trapped melts were reconstructed using measured trace element contents in the melt inclusions normalized to the Al<sub>2</sub>O<sub>3</sub> of the reconstructed melts in order to take into account the amount of the olivine, which crystallized on the walls of inclusions cavities.

Due to low solubility at low pressure, 40 to 90% of CO<sub>2</sub> can be lost to the bubbles in the inclusions (Esposito et al., 2011; Hartley et al., 2014; Mironov et al., 2015; Moore et al., 2015; Steele-Macinnis et al., 2011; Wallace et al., 2015). All studied melt inclusions do indeed contain a fluid bubble and therefore CO<sub>2</sub> concentrations measured in glasses represent only a small fraction of the CO<sub>2</sub> initially dissolved in the trapped melt. The measured water contents are also affected by this process but to lesser extent. To reconstruct the bulk CO<sub>2</sub> and H<sub>2</sub>O contents in melts, we applied the method of Wallace et al. (2015) and used the VolatileCalc (Newman and Lowenstern, 2002) and Flincor (Brown, 1989) programs. Our calculations demonstrate that on average ~87% of the original CO<sub>2</sub> has been lost to the bubble. All measured CO<sub>2</sub> and water



contents were corrected using the described method. All reconstructed major, trace elements and volatile contents of the melt inclusions are listed in the Table 2.

#### 4.6. Geothermometers and geobarometers

We used three geothermometers to estimate crystallization temperatures of the komatiite melt: a) partitioning of Sc and Y between olivine and melt (Mallmann and O'Neill, 2013), b) partitioning of Fe and Mg between olivine and melt (Ford et al., 1983), and c) partitioning of Al between olivine and spinel (Coogan et al., 2014). The temperatures calculated from Fe and Mg partitioning were then corrected for water dissolved in the melt using the equation of (Falloon and Danyushevsky, 2000).

To calculate Sc/Y temperatures, we used measured composition of olivines and the evaluated concentrations of Sc and Y in the komatiite melt as functions of MgO constrained using composition of bulk rocks following approach described in Sobolev et al. (2016):

$$\text{Sc melt} = 46.176 - 0.898 * \text{MgO}$$

$$\text{Y melt} = 17.318 - 0.505 * \text{MgO}$$

We used the published whole-rock data for Sc and Y (Bickle et al., 1993; Jochum et al., 1991; Nisbet et al., 1987) because Sc contents in melt inclusions appeared to have been strongly affected by diffusion like that of Fe (see Section 5.3 and 6.2). The Y contents in the melts calculated using the equation above are similar within error but overall are lower than those measured directly in inclusions by LA ICP-MS and then corrected for post entrapment crystallization.

The olivine-spinel thermometry was applied to olivine containing less than 91 mol% Fo, because only these co-exist with spinel (see Section 5.2 below). For the uncertainties on the temperature estimates, we used reported errors for these geothermometers:  $\pm 20$  °C (2 standard errors) for Al-olivine-spinel geothermometer (Coogan et al., 2014) and  $\pm 24$  °C (1 standard error) for Sc/Y olivine-melt geothermometer (Mallmann and O'Neil, 2013).

To estimate oxygen fugacity, we first used oxygen geobarometers based on the partitioning of V between olivine and melt (Mallman and O'Neil 2013). Measured V-concentrations in the melt inclusions could be affected by the redistribution between host olivine, daughter spinel and the melt (see Section 5.3 below) and therefore we estimated content of V as a function of MgO using the whole rock data of (Puchtel et al., 2009) following method of Sobolev et al. (2016)

$$\text{V melt} = 297.18 - 5.8206 * \text{MgO}.$$

This method gives a precision of  $0.08 \log_{10} fO_2$  units (one standard deviation, Mallmann and O'Neill, 2013). We also used compositions of the coexisting olivine-spinel pairs measured in the thin sections and the method of Ballhaus et al. (1991). The reported 1 standard error precision of this protocol is  $\pm 0.4$  log units.

All uncertainties on our measurements are reported as 2 standard errors.

## 5. Results

### 5.1. Olivine

Representative compositions of olivine from Belingwe komatiites are listed in Table 1. These compositions are consistent with those reported by Renner et al. (1994) and Sobolev et al. (2007). The analyzed olivine cores are un-zoned and have Fo contents from 93.5 to 87.5 mol% (Fig. 3) while the rims show normal zoning to Fo85 (see Fig. 4 and Renner et al., 1994). Mapping of the trace-element distribution demonstrates that P, Cr and Al possess oscillatory zoning, typical for magmatic olivine (see Fig. 4 and Milman-Barris et al., 2008). On this basis, we conclude that the fresh olivine relicts were not affected by post-magmatic alteration or metamorphism and likely preserved their initial magmatic patterns.

### 5.2. Chrome Spinel

Chrome spinel occurs as late stage microcrystals in close contact with, but not included in, olivine. This phase started to crystallize when the olivine had the composition Fo90.9, as indicated by the kink in the trend in the Cr vs. Fo diagram (Fig 3). Compositions of spinel grains are given in Table A.1 (online supplementary material). Spinel crystals are un-zoned except for thin Fe-rich reaction rims composed of microcrystalline magnetite aggregates. However, spinel grains have a variety of compositions with Mg#  $[=Mg/(Mg+Fe^{2+})]$  ranging from 0.34 to 0.63 and Cr#  $[=Cr/(Cr+Al+Ti+Fe^{3+})]$  from 0.64 to 0.74. FeO/Fe<sub>2</sub>O<sub>3</sub> ratios in spinel were calculated from stoichiometry. All studied spinel grains show no evidence of low-temperature re-equilibration with residual melt nor signatures of metamorphic alteration, excepting the magnetite rims (Barnes, 1998).

### 5.3. Melt Inclusions: major and trace elements

The studied melt inclusions are hosted in olivine Fo92.5-90.9 and their compositions are not significantly affected by spinel crystallization because olivine with this composition crystallizes alone. However, all heated melt inclusions contain small spinel crystals that crystallized after entrapment within the inclusions and were not melted completely, probably

because of insufficient experimental temperatures (1350°C or lower). Complete melting of the daughter spinel crystals would have increased the concentrations of Cr and V in the melts. Concentrations of these elements in melt inclusions should therefore be considered as minima.

The concentrations of major and trace elements in melt inclusions are listed in Table A.2 (uncorrected compositions, online supplementary material) and Table 2 (corrected for post-entrapment crystallization and volatile loss in the bubble) and plotted against MgO in Fig. 5 and Fig. 6. Reconstructed trapped melts have a relatively small range of compositions, from 19.8 to 23.6 wt.% MgO, which is much less than in the whole rock samples which cover a wide range of MgO contents from 15 wt.% in spinifex textured komatiites to ~45-50 wt.% MgO in olivine cumulates (Bickle et al., 1993, Nisbet et al., 1987, Puchtel et al., 2009).

Most of the measured immobile major and trace element contents in the melt inclusions are consistent with the data from whole rock samples and vary regularly as a function of MgO content. Differences in the Na, K, Ba, Rb and Pb contents of whole rocks and melt inclusions are related to the alteration that affected the Belingwe komatiites. The contents of these elements in bulk rocks are thus not representative for the original melt, whereas the melt inclusions preserved initial magmatic concentrations of these elements. Finally, a group of immobile trace elements (Sc, Y, V) correlate with MgO both in the melt inclusions and in the whole rock samples (Fig. 6) but there are small but systematic shifts between the melt inclusions and whole rock data.

The abundances of TiO<sub>2</sub>, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, K<sub>2</sub>O in the melt inclusions in more magnesian olivines plot on the tight trends that coincide with olivine control lines (Fig. 5). Na<sub>2</sub>O and CaO also plot on tight trends, but these have steeper slopes than predicted by olivine fractionation. These elements display slight excess in the melt inclusions hosted by less magnesian olivine. The Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> of the Belingwe komatiites yields an average of 21.3, a value close to the chondritic, which classifies them as Al-undepleted komatiites (Jahn et al., 1982; Nesbitt and Sun, 1976).

Trace element abundances in melt inclusions show larger scatter than the whole rock data (because of the larger analytical uncertainty of analyses of small inclusions) but they mostly plot close to the trends defined by the whole rock compositions (Fig. 6). Trace element concentrations of the melt inclusions, as well as previously reported whole rock data from the same flow, (Tony's Flow, Bickle et al., 1993) were normalized to 0.39 wt.% TiO<sub>2</sub> (average concentration in the studied melt inclusions) and to primitive mantle (Hofmann, 1988) to eliminate the effect of olivine crystallization or accumulation during or following eruption (Fig. 7). Averaged mantle-normalized trace-element patterns of melt inclusions are smooth but feature pronounced positive Sr anomalies, negative Pb anomalies and significant positive anomalies of volatiles (H<sub>2</sub>O, F, Cl). Whole rock samples show similar REE patterns, positive Sr anomalies and significant positive Pb anomalies. In the whole rock samples, the mobile elements Pb, Rb, Ba and K do not preserve

initial features and have very wide ranges due to alteration, whereas in the melt inclusions they display tight areas without significant deviations. On this basis we conclude that the melt inclusions are minimally affected by post-entrapment alteration and preserve the original melt compositions.

The trace element patterns from melt inclusions display depletion in light rare earth elements (LREE;  $(\text{La}/\text{Sm})_n=0.65\pm 0.03$ ) and flat heavy rare earth element (HREE) patterns ( $([\text{Gd}/\text{Yb}]_n=1.01\pm 0.06)$ ). These values are characteristic of Al-undepleted (Group 1) komatiites described by (Jahn et al., 1982) and are consistent with previous studies of the Belingwe komatiites (Bickle et al., 1993; Puchtel et al., 2009; Shimizu et al., 2005).

#### 5.4. Volatile contents in the melt inclusions

Representative analyses of volatiles in the melt inclusions, corrected for  $\text{CO}_2$  loss and for post-entrapment crystallization (see Section 4.5), are listed in Table 2 and the uncorrected data in Table A.2 (online supplementary material). Measured  $\text{CO}_2$  in glasses ranges from 3 to 43 ppm.

Water contents measured directly by SIMS in melt inclusions are subject to uncertainty due to a relatively high permeability of hydrogen in olivine, as demonstrated by numerous studies (Mei and Kohlstedt, 2000; Mironov et al., 2015; Portnyagin et al., 2008; Sobolev and Danyushevsky, 1994). Hence, prior to interpreting their original concentrations, it is important to determine whether the measured water contents were affected by diffusive loss or gain of H after entrapment. Measured water contents differ by 30-40% (averaged measured  $\text{H}_2\text{O} = 0.37$  wt.% versus average corrected  $\text{H}_2\text{O} = 0.24$  wt.%), from values corrected for post-entrapment crystallization and loss to the vapor bubble. The water contents show a significant negative correlation with the Fo content of host olivine ( $R^2=0.46$  for 20 inclusions, confidence level 0.95) and hence with the MgO contents of the trapped melt. Plotted on Fig. 8 are correlation lines between the volatile contents in the melt inclusions hosted in  $\text{Fo} > 91.5$  and the Fo contents of the host olivines. Despite the low  $R^2$ , these melt inclusions are consistent within error with the olivine control line and hence are representative of the parental melt compositions. Of the major and trace elements that are incompatible with olivine (e.g.  $\text{TiO}_2$ ,  $\text{Na}_2\text{O}$ , Rb, Li, Sc, V, Cu, Ce), most display positive correlations with the corrected water contents of the melt inclusions. There are also significant (on 95% confidence level) positive correlations between  $\text{H}_2\text{O}$  and S and F (Fig. 9). In contrast, no correlations are observed between the size of the melt inclusions and  $\text{H}_2\text{O}$  concentrations. All these observations suggest that water-concentrations in melt inclusions represent those of the originally trapped melts and do not show any significant loss or gain due to

diffusion through the host olivine, as also was demonstrated for the olivine hosted melt inclusions by Berry et al. (2008).

Questionable data on water contents was acquired from four data points in small (7-21 $\mu$ m) melt inclusions from the cumulate sample Z4. The average water content is 0.07 wt.% H<sub>2</sub>O, markedly lower than 0.2-0.3 wt.% H<sub>2</sub>O in sample Z6. The small size of the inclusions and the low water contents means that H<sub>2</sub>O loss by diffusion cannot be excluded.

Corrected CO<sub>2</sub> and H<sub>2</sub>O contents were used to estimate the equilibrium pressure using the VolatileCalc program. These calculations yielded an average pressure of 357 bars (Fig. 10) or 1.2 km depth. Our estimates likely represent the minimum pressure of magma fractionation due to partial degassing during magma ascent of CO<sub>2</sub> that was originally dissolved in the melt (Dixon et al., 1995). This is supported by relatively low CO<sub>2</sub>/Ba (5-41.4) and CO<sub>2</sub>/Nb (54.7-432.1) ratios compared to estimated ratios for the modern depleted mantle (CO<sub>2</sub>/Ba=133 $\pm$ 44, CO<sub>2</sub>/Nb=505 $\pm$ 168, Rosenthal et al., 2015). Thus, the data probably apply to the last vapor-melt equilibrium in the magmatic system. The calculated equilibrium fluid should have low H<sub>2</sub>O/CO<sub>2</sub> mol ratios (less than 0.025). Hence, if the degassing of the melt occurred, the effect on H<sub>2</sub>O contents was negligible because it should not exceed few % of total CO<sub>2</sub> loss.

Water, fluorine, chlorine and sulfur concentrations in the corrected trapped melts also show negative correlation with the forsterite content of the host olivine (Fig. 8). The melts trapped by the most MgO rich olivines closely follow olivine fractionation line, while those included in more evolved olivines tend to have more elevated volatile concentrations than predicted by olivine control alone.

Chlorine concentrations in the melts range from 269 to 595 ppm with an average of 407  $\pm$  40 ppm. Two outliers are not included in the average. One of these is an extremely Cl-rich inclusion containing up to 1280 ppm and the other is a small melt inclusion (20  $\mu$ m) with an unusually low Cl (84 ppm). The latter was excluded from further consideration. The major and trace element composition of the melt inclusion with extremely high Cl is similar to those in the other melt inclusions for both mobile and immobile elements (Fig. 5, Fig. 6).

Water, chlorine and fluorine contents of the trapped melts show significant excess when normalized to primitive mantle (Fig. 7).

## 5.5. Geothermometers

We applied two independent thermometers – Al in olivine-spinel and Sc/Y olivine-melt – to estimate melt temperatures and water contents (see Section 4.6). Estimated temperatures are listed in the Table 2.

The Sc/Y method was applied to olivines with Fo contents from 91 to 93.3 and to cases in which we know the compositions of the melts. (Fig. 11). The estimates demonstrate a wide scatter plotting from the dry liquidus down to the 2 wt.% H<sub>2</sub>O liquidus line. The average estimated temperature for the most magnesian olivines (Fo 93.3-91.7, average Fo 92.2, N=20), based on Sc-Y thermometer, is 1448±21°C. This temperature is 31°C less than the dry liquidus temperature for the melt and thus provides evidence of a small but significant amount of water in the melt.

Al-in-olivine-spinel temperatures obtained for the more evolved olivines (88.7-90.9 mol.% Fo, listed in the Table A.3, online supplementary material) scatter between the dry and 0.5 wt.% H<sub>2</sub>O liquidus lines (Fig. 11) and tend towards the dry liquidus line with decreasing Fo. The most primitive olivine-spinel pairs (Fo 90.2-90.9, average Fo 90.5, N=7) yield an average temperature of 1391±26°C, which is ~36°C lower than the average dry liquidus temperature for this range.

### 5.6. Oxygen fugacity in the komatiite magma

Oxygen fugacity in magmatic systems can be used to distinguish between magmas originating in different tectonic settings (Evans et al., 2012). Magmas in modern subduction settings tend to be more oxidized than magmas generated in modern plumes and mid-ocean ridges. The tectonic setting of komatiites is uncertain and the redox state of the komatiite magma may provide additional information on their origin and on the oxidation state of the Archean mantle. Our estimates of  $fO_2$  are based on the Mallmann and O'Neil (2013) or Ballhaus et al. (1991) protocols (see Section 4.6), and the results are plotted in Fig. 11.

The oxygen fugacity for the most MgO rich olivines (Fo 91.7-93.3), calculated using the method of Mallmann and O'Neil (2013), implies relatively reduced conditions corresponding to QFM -0.3 to QFM -0.9 equilibria.  $\Delta QFM$  values exhibit a negative correlation with olivine Fo and follow closed-system olivine crystallization conditions from  $\Delta QFM$ -1.1 at Fo93.5. The method of Ballhaus et al. (1991) for olivine-spinel pairs gives results that follow closely the same line, within the reported errors.

## 6. Discussion

Even though the komatiites in the Belingwe belt are among the best-preserved Archean komatiites, they have undergone alteration and regional metamorphism that partially replaced igneous minerals with secondary hydrous minerals (Bickle et al., 1993). Although some of the

immobile major and trace elements (e.g., HFSE, REE, V, Sc, Y) preserve the initial melt characteristics, concentrations of mobile elements (Na, K, Ba, Rb, Pb) and volatiles were strongly affected by post-magmatic processes. Despite the alteration, Belingwe komatiites contain abundant relicts of fresh olivine. Melt inclusions hosted in these relicts have immobile major and trace element concentrations consistent with the whole rock data. They also preserved the concentrations of mobile trace elements and volatiles in the trapped melts. Moreover, melt inclusions in the most magnesian olivines follow olivine fractionation trends and thus can be used to reconstruct the initial melt composition and the original water contents.

### **6.1. Signs of contamination**

Contents of  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$ ,  $\text{K}_2\text{O}$  and most of the trace elements in the trapped melts are consistent with olivine control lines for the entire compositional range of the host olivines (Fig. 4, 5). However, melt inclusions in the more evolved olivines display scatter about, but mainly above, the olivine fractionation line for the concentrations of  $\text{Na}_2\text{O}$ ,  $\text{CaO}$ ,  $\text{Li}$ ,  $\text{La}$ ,  $\text{Cu}$ ,  $\text{Rb}$ ,  $\text{Y}$ ,  $\text{Sc}$ , as well as  $\text{H}_2\text{O}$ ,  $\text{Cl}$ ,  $\text{F}$  and  $\text{S}$ . This enrichment cannot result from the fractional crystallization of olivine but may be related to assimilation and fractional crystallization (AFC, except for  $\text{Sc}$  and  $\text{Y}$  – as discussed in a later section), which probably affected komatiites at a late stage of crystallization.

One possible source for the  $\text{H}_2\text{O}$  and  $\text{Cl}$  enrichment is seawater or high-salinity brines (Kent et al., 2002; Kent et al., 1999; Lassiter et al., 2002). As shown in Figure 12, two-component mixing of komatiite melt with seawater is inconsistent with the observed  $\text{H}_2\text{O}/\text{Cl}$  and  $\text{K}/\text{Cl}$  ratios in the melt, whereas mixing with high-salinity brine yields a range of compositions that coincides with the melt inclusion data as well as with data for altered MORB and OIB glasses (Kendrick et al, 2017). The contamination of komatiite melt could have taken place on its way to the surface via the assimilation of brine saturated sea-water altered oceanic crust (e.g. Lassiter et al, 2002). The relative excess of fluid-mobile elements in komatiite melt inclusions is not very large and is observed only for the most evolved melts, and it is therefore not possible to accurately model the contamination. Nonetheless, the compositions of the most primitive melt inclusions are consistent with the model of olivine fractionation and hence are representative of the parental melt.

### **6.2. Reconstruction of parental melt**

To exclude the contamination effect from the calculations of the parental melt composition, we used a set of 7 inclusions that lacked pronounced contamination signatures in the most  $\text{MgO}$  rich olivines (Fo 92.1-92.5, Table 2). In our calculations, we assumed that the

initial melt was in equilibrium with Fo 93.5 – the most MgO-rich olivine analyzed in this and earlier studies (Bickle et al., 1993, Sobolev et al., 2007). To calculate the melt composition in equilibrium with this olivine, we used the reverse crystallization of olivine procedure in the Petrolog 3.0 software of Danyushevsky and Plechov (2011). To calculate the compositions of the melt in equilibrium with olivine we applied the models of Ford et al. (1983) and Herzberg and O'Hara (2002) and averaged the compositions of melts in equilibria with Fo 93.5 acquired using the two protocols. This makes sense, because as was shown recently by Krashennnikov et al. (2017) testing of these models displays a certain temperature underestimation for the Ford et al. (1983) model and temperature overestimation for the Herzberg and O'Hara (2002) model for the temperatures exceeding 1450°C. The oxygen fugacity was based on V partitioning between the host olivine and melt (Mallmann and O'Neill, 2013), which corresponds to closed system crystallization starting at  $\Delta\text{QFM} -1.1$  (Fig. 11).

Our estimated primary komatiite melt contained  $47\pm 0.7$  wt.%  $\text{SiO}_2$ ,  $27.5\pm 2.9$  wt.% MgO and  $0.18\pm 0.02$  wt.%  $\text{H}_2\text{O}$  (Table 2). The calculated major element contents for the primary komatiite melt of the Belingwe komatiites are consistent with the 50-60% partial melting of peridotite KR4003 (Fig. 13), unlike the Abitibi komatiites which have too little  $\text{SiO}_2$  (Herzberg, 2016; Herzberg and O'Hara, 2002; Walter, 1998). The pressure of melting is estimated to be ca. 7 GPa (Lee et al., 2009), and 60% partial melting of mantle peridotite at such depth would produce a residue composed of olivine with small amount of orthopyroxene, which is consistent with the proposed origin of Al-undepleted komatiite (Herzberg, 2004).

The systematic differences between the Sc and Y contents of melt inclusions and the whole rock data must be taken into account. Both elements are sensitive to temperature and may redistribute between the solid and liquid during cooling, and V, which has  $\text{V}^{3+}$  and  $\text{V}^{5+}$  forms, partitions differently between solid and liquid depending on the temperature and oxidation state of the magma. All studied melt inclusions come from cumulates, which are the lower parts of the flows and do not quench rapidly. We therefore suggest that the differences in the concentrations of Sc and Y between melt inclusions and whole rock samples results from redistribution of these elements between the trapped melts and host olivines. The V shift may be due to the presence of spinel crystals in the melt inclusions that were not completely resorbed during the experimental heating. As V is relatively compatible in spinel, these could reduce the V concentrations in the trapped melts. Thus, the whole-rock sample data may best represent the initial komatiite melt for these elements.



### 6.3. Independent approach for water contents

To check water contents measured directly in the melt inclusions by SIMS, we applied two independent geothermometers to estimate melt temperatures (see Section 4.6 for the uncertainties of the geothermometers) and evaluate the equilibrium water contents (see 5.5 Section). The estimated temperatures are listed in the Table 2.

Data obtained using the S/Y geothermometer may be influenced by Sc and Y redistribution between olivine and the residual melt during cooling (Fig. 11) and may therefore represent only the temperatures of the last olivine-melt equilibrium. But the temperatures obtained for the most MgO rich olivines (Fo 91.7 – 93.3) are likely to be unaffected by such re-equilibration and hence are more reliable. This is consistent with the fact that these olivines preserved their high Fo contents, suggesting that there was not enough time, or enough interstitial melt, to significantly change their compositions during later cooling. The estimated average temperature for the most magnesian olivines (average Fo 92.2, N=20), is  $1450\pm 22^\circ\text{C}$ , which is  $30^\circ\text{C}$  lower than the dry liquidus temperature of melt in equilibrium with the olivine composition. The model of Falloon and Danyushevsky (2000) can be used to estimate the amount of  $\text{H}_2\text{O}$  that is needed to depress the temperature by  $30^\circ\text{C}$ . We calculate  $\text{H}_2\text{O}$  contents between 0.0-0.37 wt.% with a mean of  $0.1\pm 0.1$  wt.%  $\text{H}_2\text{O}$ , which is consistent with the direct estimates using SIMS data ( $0.20\pm 0.02$  wt.%).

Even though the uncertainty of Sc/Y temperatures is large, this independent approach yields similar water contents, as obtained from direct SIMS measurements. This argues against the significant water gain or loss during the post-entrapment history.

Al-in-olivine-spinel temperatures scatter between the dry and 0.5 wt.%  $\text{H}_2\text{O}$  liquidus lines (Fig. 11) and trend towards the dry liquidus with decreasing Fo number. This variation is attributed to degassing of the melt in shallow crustal conditions. The most primitive olivine-spinel pairs (Fo 90.2-90.9, average Fo 90.5, N=7) yield an average temperature of  $1391\pm 26^\circ\text{C}$ , which is  $\sim 36^\circ\text{C}$  lower than the average dry liquidus temperature for this range. This difference corresponds to 0-0.5 wt.%  $\text{H}_2\text{O}$  (mean 0.13 wt.) in the melt and again is consistent within error with the direct SIMS measurements. However, these concentrations are significantly lower than those obtained by direct measurements of water in melt inclusions in spinel by Shimizu et al. (2001). We suspect that these melt inclusions were contaminated and do not accurately record the initial volatile contents of the magma. In contrast, our estimates on water obtained by direct measurements are consistent with the data reported by (Danyushevsky et al., 2002) and represent the maximum possible value.

#### 6.4. Excess of H<sub>2</sub>O and Cl in the parental komatiite melt

Our estimates of volatile contents in the parental melt are based on measurements of inclusions trapped in the most Mg-rich olivines. The concentrations of volatiles (H<sub>2</sub>O, F, Cl and S) in these inclusions correlate with both the MgO of the melt and the Fo content of the host olivines and closely follow closed-system olivine fractionation paths (Fig. 8). We estimate 0.2 wt.% H<sub>2</sub>O, 290 ppm Cl, 192 ppm S and 32 ppm F in the melt in equilibrium with Fo<sub>93.5</sub>.

Multiple correlations between water contents and concentrations of other volatiles, incompatible trace elements in melt inclusions and the forsterite content of the host olivine argue against H loss through diffusion. In addition, the estimated very low H<sub>2</sub>O/CO<sub>2</sub> ratio of the equilibrium fluid ([H<sub>2</sub>O/CO<sub>2</sub>] = 0.025; see Section 5.4) precludes significant degassing of H<sub>2</sub>O before entrapment of melt inclusions, because the effect of degassing on H<sub>2</sub>O contents was negligible as it should not exceed few % of the total CO<sub>2</sub> loss. All these suggest that the estimated H<sub>2</sub>O contents, as well as concentrations of F, Cl and S in the parental melt, are reliable. However, the estimated concentrations of H<sub>2</sub>O and Cl in Belingwe komatiites are much higher (H<sub>2</sub>O/Ce > 1100, Cl/K ~ 1.3) than those estimated for modern mantle melts (H<sub>2</sub>O/Ce = 166, Cl/K = 0.1) (Hofmann, 1988; Kendrick et al., 2017) (Fig. 14). On the other hand, recent experimental studies of high-pressure olivine polymorphs (wadsleyite and ringwoodite) have demonstrated that at the conditions corresponding to the mantle transition zone (14-22 GPa, 1100-1400 °C), these phases are capable of holding significant amounts of Cl, F and H<sub>2</sub>O (Roberge et al., 2015; Roberge et al., 2017).

#### 6.5. Implications for the source of excess of water and chlorine

One possible source of excessive volatiles is H<sub>2</sub>O-rich rock such as serpentinite, which could have been assimilated by the komatiite melt. In the results section, we noted that inclusions trapped in evolved olivine grains contain amounts of H<sub>2</sub>O, F, S, Ca, Na in excess over those expected from closed-system olivine fractionation. This implies minor contamination of the komatiite magma during crystallization. However, a strong correlation between water contents of the trapped melts with the composition of the host high-Mg olivines (>92 Fo), consistent with closed-system fractionation argues against contamination at an early stage of crystallization. Thus, the excessive contents of H<sub>2</sub>O, as well as Cl and F, estimated for the less-evolved melt likely represent those of the primary magma and hence its mantle source.

In this case two scenarios are possible: a supra-subduction origin of the komatiite melts (Grove et al., 1997; Parman et al., 1997; Parman et al., 2004) or derivation from a hydrous mantle plume (Shimizu et al., 2001; Sobolev et al., 2016). As shown on Fig. 11; the calculated parental melt for Zimbabwe komatiites has a smooth and flat trace-element pattern with

moderate depletion in the LREE, a strong negative Pb anomaly, and large positive H, Cl and F anomalies. The absence of typical supra-subduction features such as negative Nb-Ta anomalies and positive anomalies of large ion lithophile elements (Rb, Ba, K, Sr) and Pb suggests a non-subduction origin of both komatiites, as argued previously for Belingwe by McDonough and Ireland (1993) and Shimizu et al. (2001). While the subduction related geochemical fingerprints (e.g. negative Nb-Ta anomalies) do in fact exist in the Archean ultramafic and mafic volcanic rocks (Polat et al., 2010). The low oxygen fugacity of primary melts (lower than QFM- 1) also argues against a supra-subduction setting. Instead, these observations support a hydrous mantle plume origin of Belingwe komatiite as in the model recently proposed by Sobolev et al. (2016). In this model, the mantle transition zone between 410-660 km contained significant amounts of H<sub>2</sub>O, F and Cl (Roberge et al., 2015; Roberge et al., 2017) which contaminated the plume en route to the lithosphere.

Sobolev et al. (2016) inferred 0.6 wt.% H<sub>2</sub>O in the initial melts of 2.7 Ga Abitibi komatiites, a slightly higher value than inferred for the Belingwe melt. On the other hand, the H<sub>2</sub>O/Ce ratio of the estimated primary melt is more than 6 times higher and Cl/K ratio is 13 times higher than those estimated ratios for modern depleted mantle. This difference probably reflects heterogeneous distribution of H<sub>2</sub>O and Cl in the transition zone and/or differing extents of entrainment of hydrous material into the mantle plume

#### 6.6. Magma generation conditions

Our estimated initial melt composition (Table 2) has MgO=27.5±2.9wt% and a liquidus temperature of 1513±54°C (2 standard errors). Magma generation pressures and temperatures were calculated using the method of Lee et al. (2009), which takes into account the presence of water and gives 1787°±54°C at 6.7±0.2 GPa or 223±6.7 (2 standard errors) km depth. These estimates correspond to 60% of partial melting, however, the initial melting depth was probably much deeper and could have started in the transition zone of mantle (410-660 km depth or >13 GPa), which allows mantle plume to entrain the transition zone material (Sobolev et al., 2016). The temperatures of the contemporary 2.7 Ga ambient mantle are estimated to be ~1600°C according to Korenaga (2008) and Herzberg et al. (2010) or 1500°C according to Ganne and Feng (2017). Thus, the calculated magma generation temperatures are at least 150 to 300°C hotter than ambient Archean mantle providing further evidence of a plume origin of Belingwe komatiites (McDonough and Ireland, 1993; Puchtel et al., 2009; Shimizu et al., 2001).

## 7. Concluding remarks

- Excess concentrations of Na<sub>2</sub>O, CaO, Li, La, Cu, Rb, Y, Sc as well as of H<sub>2</sub>O, Cl, F and S over the olivine fractionation line in the evolved trapped melts suggest contamination of the komatiite lava at shallow levels of during emplacement.
- No signs of contamination are observed for the melts hosted in the most magnesium rich olivines. Hence, these melts are representative of the parental komatiite magma.
- Belingwe komatiite lavas contained up to 23.5 wt.% MgO and 0.3 wt.% H<sub>2</sub>O when they erupted. The parental melt was more magnesian, with up to 27.5 wt.% MgO. It contained ~0.2 wt. of water and had a liquidus temperature of 1513°C.
- Estimates of the parental melt water contents obtained directly by the SIMS analyses of melt inclusions are consistent with those obtained indirectly using the Sc/Y olivine-melt and Al-in-olivine-spinel geothermometers.
- The high H<sub>2</sub>O and Cl contents estimated in the parental komatiite melt are thought to result from interaction of a mantle plume with a hydrous mantle transition zone.

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## Figure Captions

Fig. 1. Geological map of Belingwe Greenstone Belt, south-central Zimbabwe (modified after Shimizu et al., 2005). Analyzed samples z6 and z4 come from Tony's Flow, Belingwe

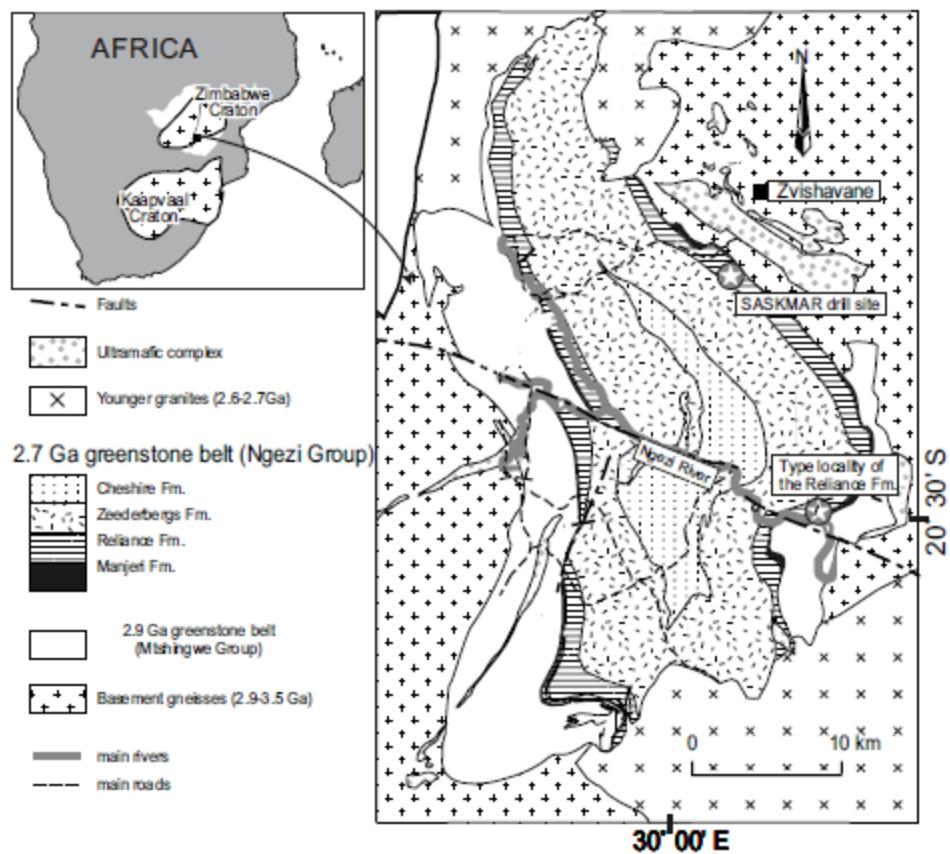


Fig.1

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Fig. 2. Melt inclusions in olivine from Belingwe belt komatiite z6. A,b,c – heated and quenched inclusions in the transmitted light; d – unheated inclusion hosted in the olivine in the thin section, back scattered electron image. Inclusions contain glass and a gas bubble, b inclusions also contains spinel crystal.

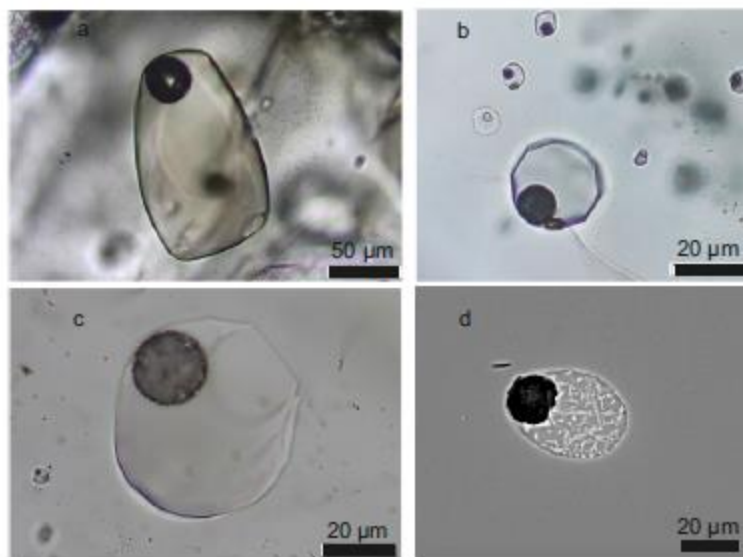


Fig. 2.

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Fig. 3. Major element compositions of olivine from the Z4 and Z6 olivine cumulates. Black arrows display the crystallizing phases.  $\text{Cr}_2\text{O}_3$  demonstrates the start of the spinel crystallization when olivine has the composition Fo91.

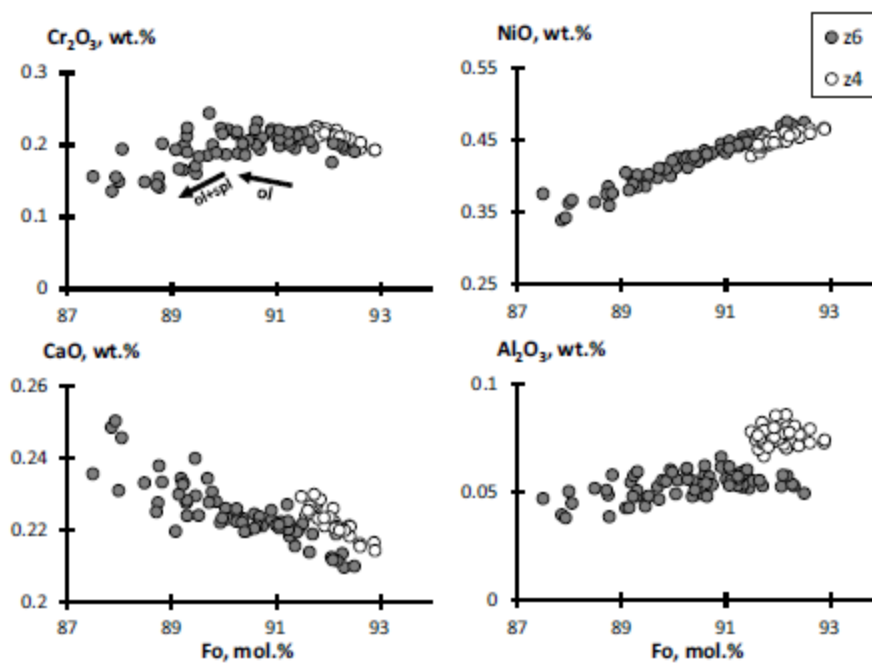


Fig.3.

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Fig. 4. Back scattered image and element qualitative distribution maps for the olivine grain in the thin section of Z6 komatiite. Normal magmatic shape and original magmatic zoning in Cr, Al and P provide evidence that the olivine grains escaped post-magmatic alteration and retain the originally trapped melts. The approximate concentrations of elements are reproduced in the color scales.

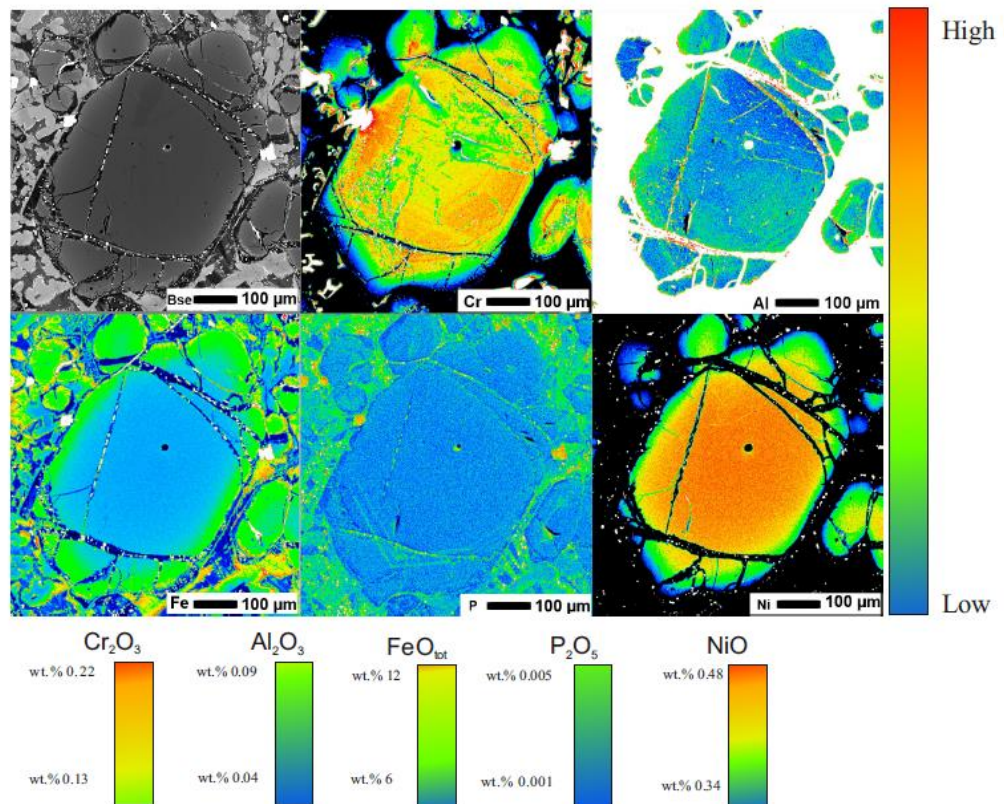


Fig. 4.

Fig. 5. Compositions of melt inclusions. Literature data from Nisbet et al., 1987, Bickle et al., 1993, Shimizu et al., 2005, Puchtel et al., 2009). Z6-Cl - chlorine rich melt inclusion; wr (Z, ZV, TN) – whole rock data. Some samples are out of scale

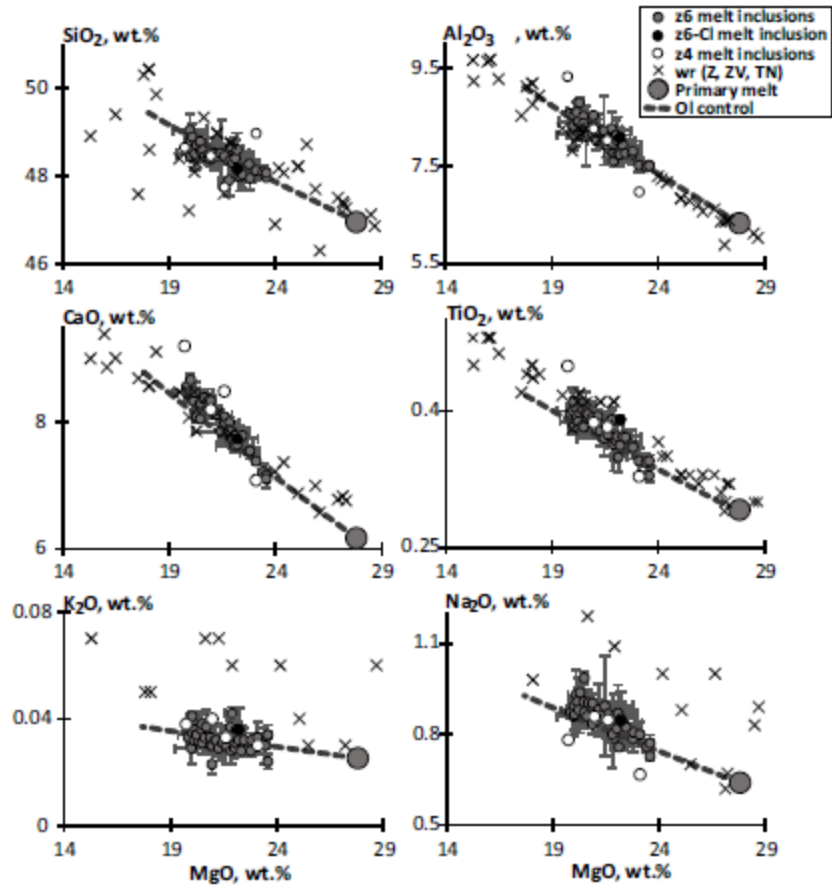


Fig. 5.

Fig. 6. Trace element contents of melt inclusions. z6-Cl – melt inclusion with extremely high Cl; wr (Z, ZV, TN) – whole rock data from literature (Nisbet et al., 1987, Bickle et al., 1993, Shimizu et al., 2005, Puchtel et al., 2009), some samples are out of scale. Sc demonstrates a systematic shift compared to the whole rock data due to the redistribution into the host olivine. Sc and Rb plot away from the olivine control line and were affected by the AFC process. Y, Ba and Ce follow the olivine control line within the error. Z6-Cl - chlorine rich melt inclusion is consistent with the melt inclusions data set. For Sc, V and Y – whole rock samples are more representative for the primary melt (see Section 4.6 for methods and Section 6.2 for discussion).

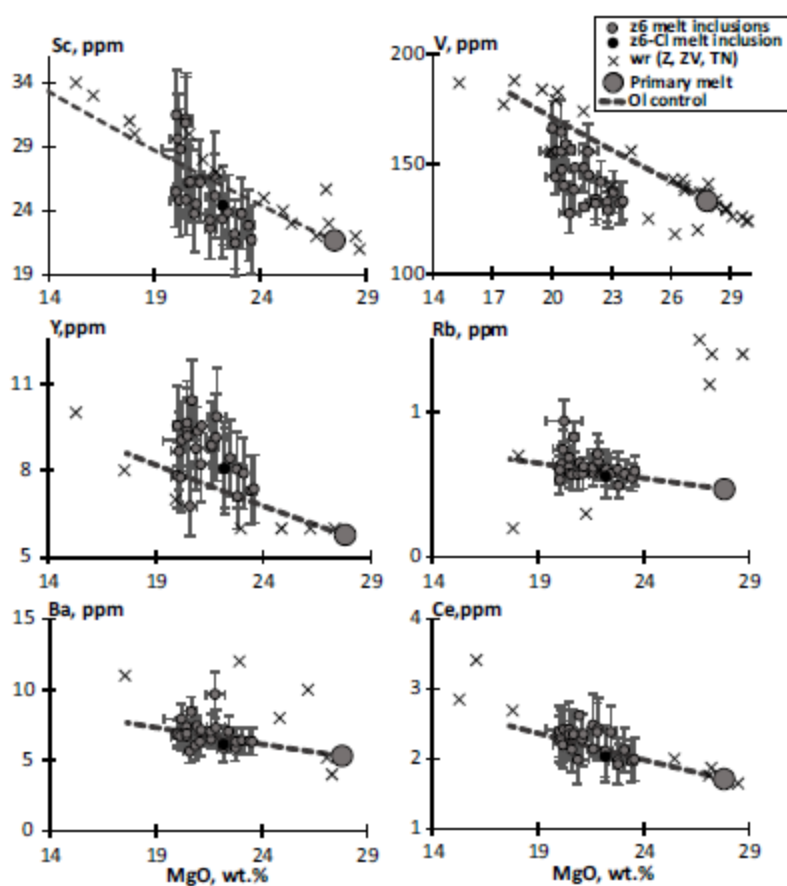


Fig.6.

Fig. 7. Spider diagram of melt inclusions and Tony flow whole rock samples normalized to 0.39 wt.% TiO<sub>2</sub> (average for the trapped melt inclusions). All data normalized for the incompatible lithophile elements in the primitive mantle after Hofmann (1988). Water, fluorine and chlorine were estimated from the ratios of H<sub>2</sub>O/Ce=166, Cl/K=0.10, F/Nd=22.7. Average ratios for uncontaminated OIB and MORB glasses after Kendrick et al. (2017). Whole rock data from Bickle et al. (1993). Grey shaded field – range of the trapped melt compositions.

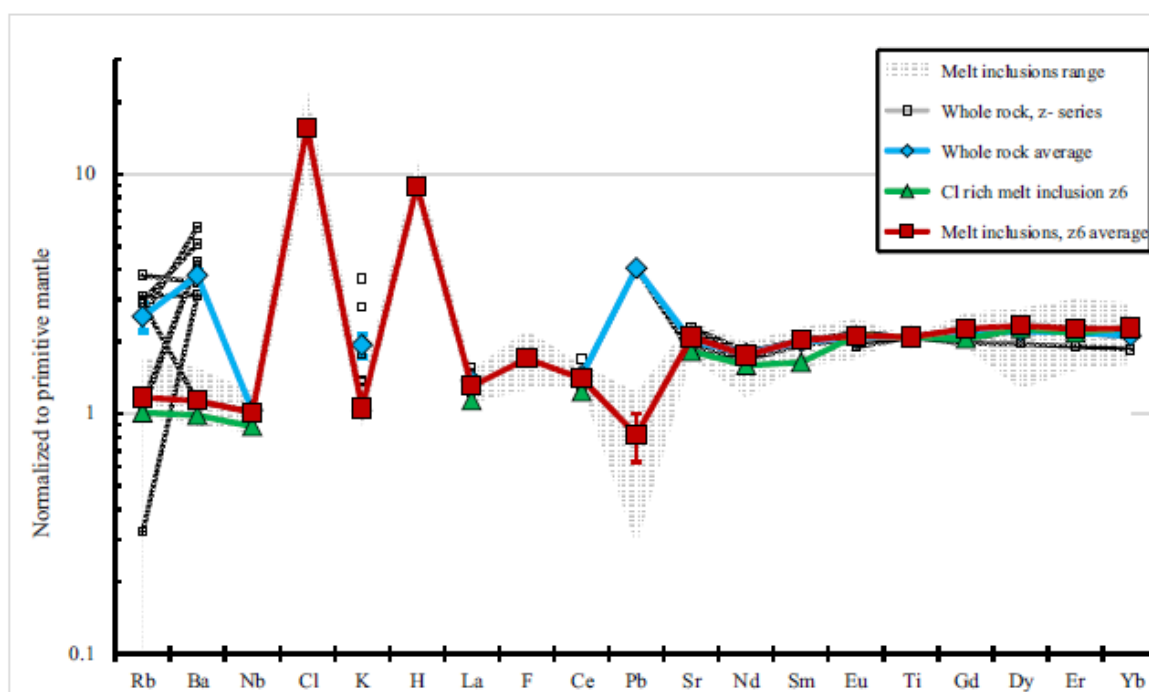


Fig. 7.



Fig. 8. Volatile contents in the melt inclusions. Data show correlation with the forsterite content of the olivine but the slopes are steeper than the olivine control line.

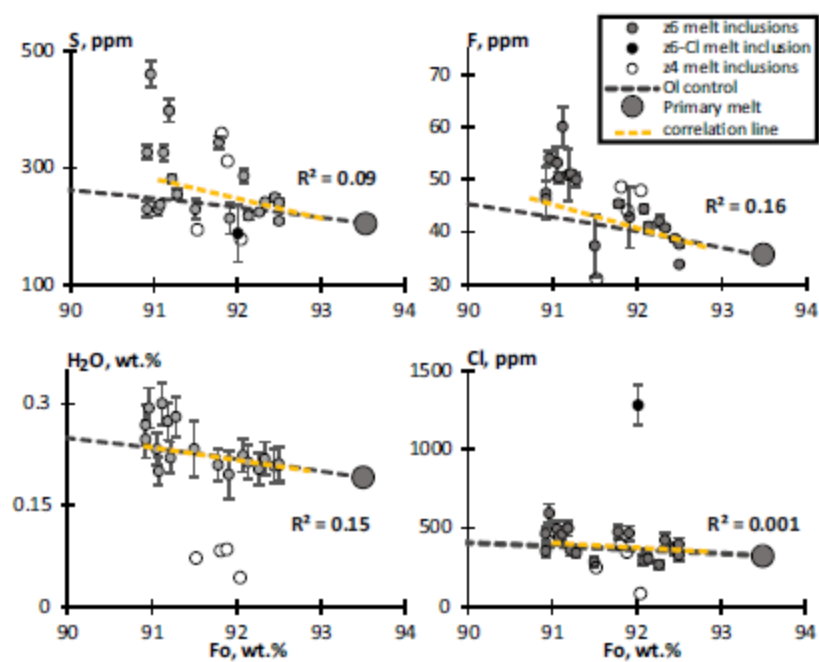


Fig. 8.

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Fig. 9. Water contents versus Cl, S, F and size of the melt inclusions. Mi – melt inclusions; Corr – correlation lines. Correlations between H<sub>2</sub>O and other volatiles suggest that melt inclusions did not lose or gain water after the entrapment and represent the initial magma contents. The absence of a correlation with the size of olivine grains provides strong evidence against diffusion of H<sub>2</sub>O.

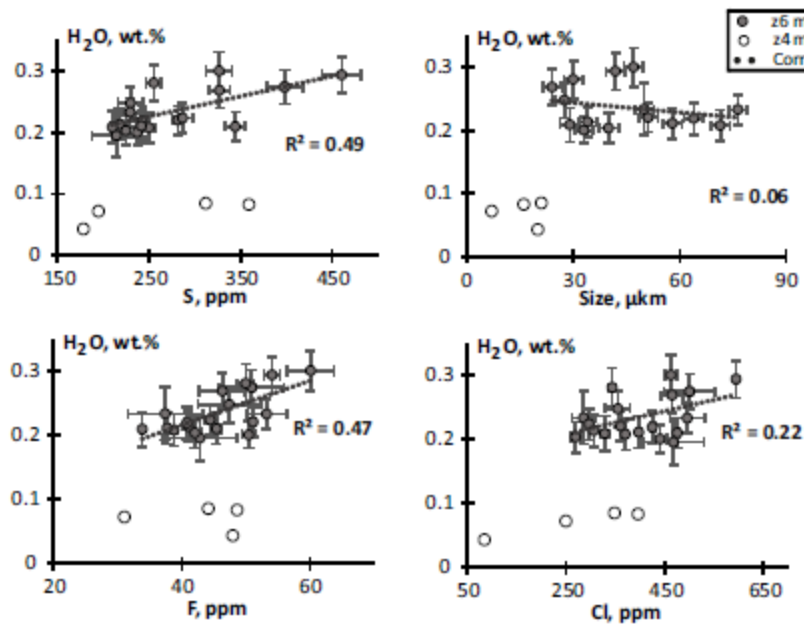


Fig. 9.

Fig. 10. Pressure calculated using the measured and corrected. Dashed line – average pressure- is given for the corrected  $\text{CO}_2$  contents of the trapped melts (Newman and Lowenstern, 2002) refer to the depth of crystallization of the host olivine.

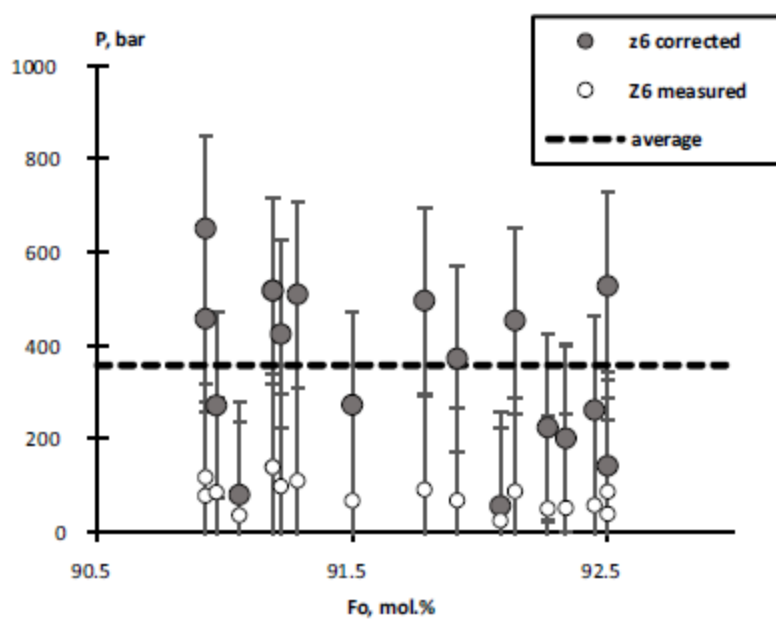


Fig. 10.

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Fig. 11. A. Crystallization temperatures of Belingwe komatiites versus olivine composition. Temperatures are estimated from Sc/Y olivine-melt, Mg-Fe olivine-melt and Al olivine-spinel geothermometers (See Methods). Dashed lines show liquidus temperatures of the komatiite melt with various water contents. B. Oxygen fugacity, estimated from V partitioning between olivine and melt, follows a closed-system trend.

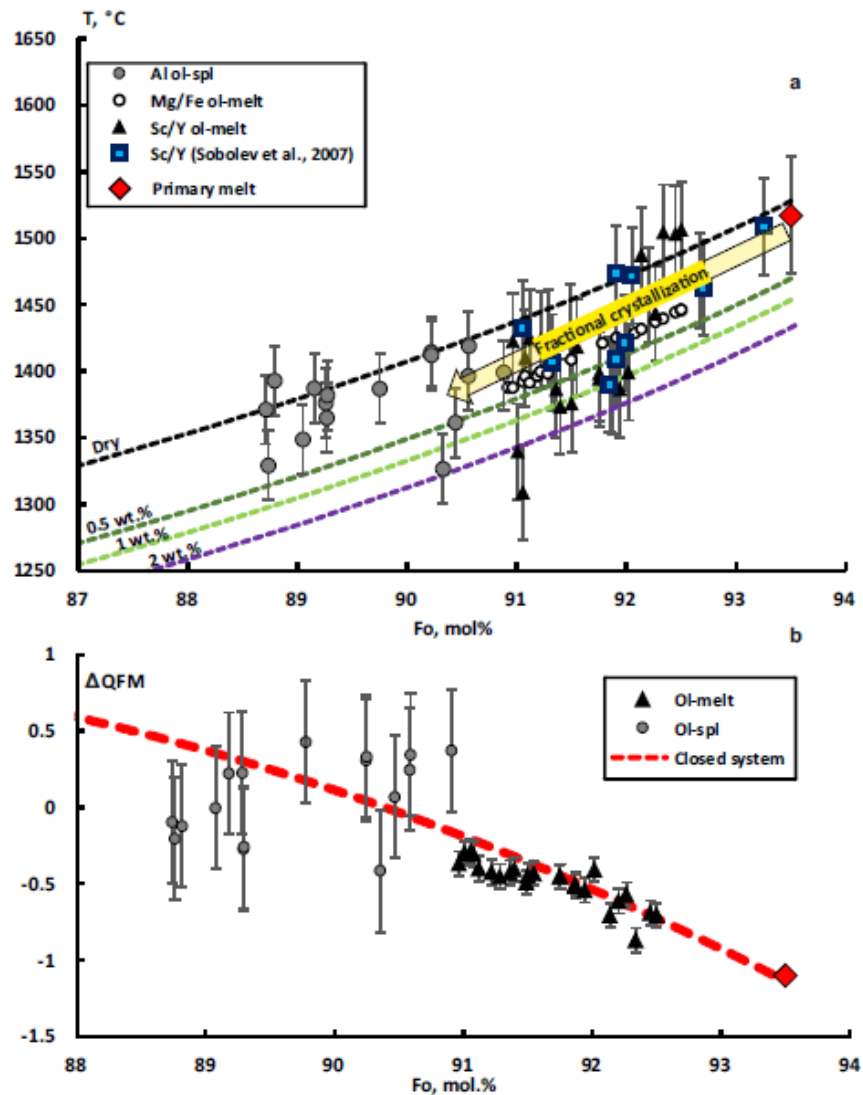


Fig. 11.

Fig. 12.  $H_2O/Cl$  versus  $K/Cl$  with melt inclusions data for Belingwe komatiites (this study), Abitibi komatiites (Sobolev et al., 2016) and Gorgona komatiites (Gurenko and Kamenetsky, 2011; Gurenko et al., 2016). Arc lavas – the primitive magma compositions compilation by Ruscitto et al. (2012). MORB and OIB glasses compilation by Kendrick et al. (2017). "Contaminated" identifies samples that may have assimilated seawater Cl. AOC – altered oceanic crust (Michael and Cornell, 1998). Serpentine data from (Kodolányi et al., 2011)

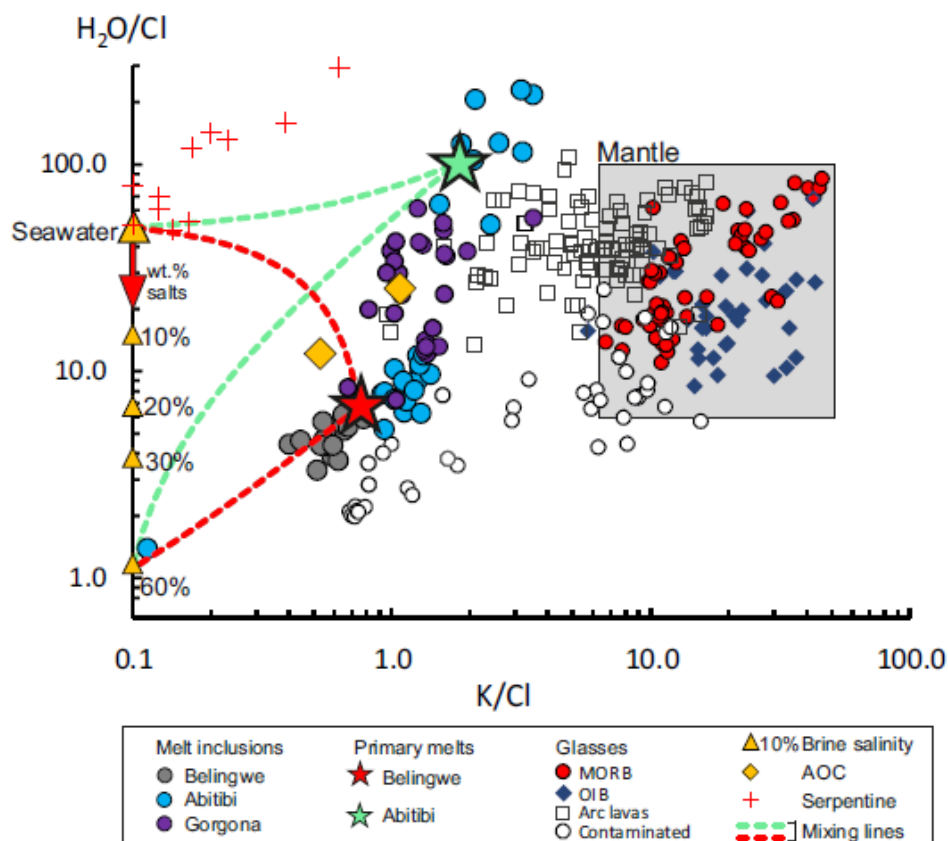


Fig. 12.

Fig. 13. MgO and SiO<sub>2</sub> contents of komatiites from Belingwe (this study) and Abitibi (Sobolev et al., 2016) komatiites compared with the model volatile-free primary magmas of peridotite KR4003 produced by batch melting (a) and accumulated fractional melting (b) after Herzberg (2016).

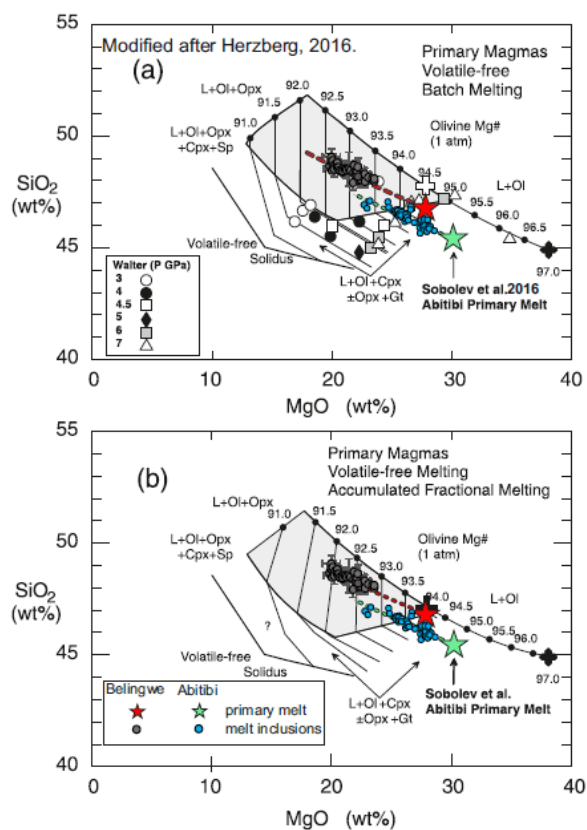


Fig. 13.

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Fig. 14. Spider diagram of the calculated parental melt for z6 komatiites and parental melt for the Abitibi komatiites inferred from the melt inclusions (Sobolev et al., 2016). Knowing the approximate degree of partial melting (60%) and assuming olivine with minor orthopyroxene to be the only residual phases in the restite allows to reconstruct the source peridotite trace element composition by roughly multiplying the initial komatiite melt composition by 0.6. All data normalized for the incompatible lithophile elements in the PM after Hofmann (1988). Water, fluorine and chlorine were estimated from the ratios of  $H_2O/Ce=166$ ,  $Cl/K=0.10$ ,  $F/Nd=22.7$ . Average ratios for uncontaminated OIB and MORB glasses after Kendrick et al. (2017).

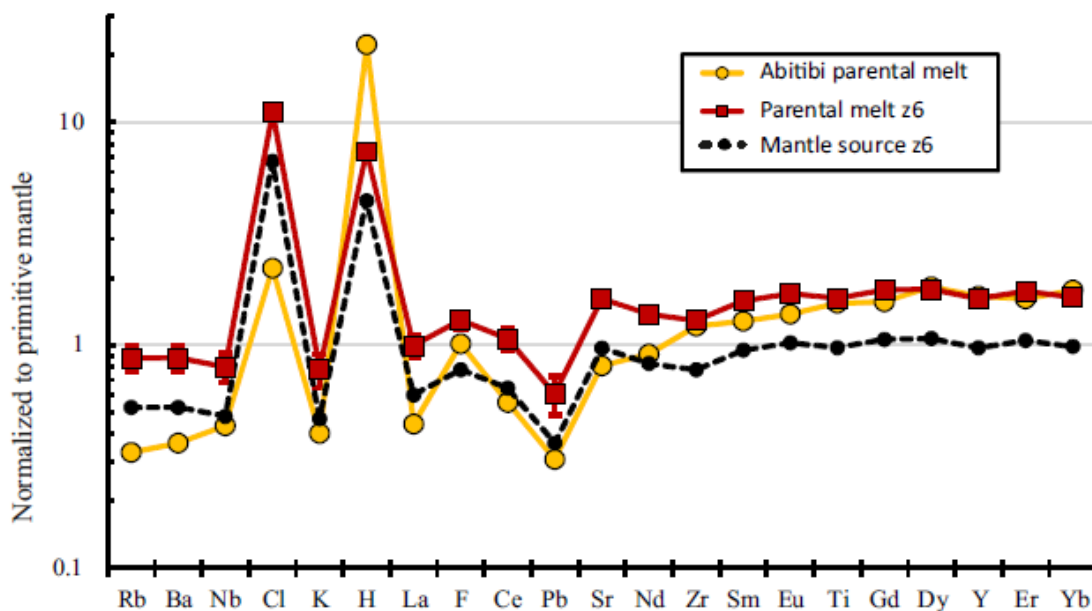


Fig. 14.

Table captions

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Table 2. Melt inclusions corrected data on the major and trace element concentrations and volatile contents and calculated parental melt compositions.

SAMP NO	Melt Inclusions												Parental Melt											
	MZ	MZ	MZ	MZ	MZ	MZ	MZ	MZ	MZ	MZ	MZ	MZ	Al	Ca	Fe	Na	Si	SO	Ti	Zr	2	3	4	
1	4	4	4	4	4	4	4	4	4	4	4	4	2	4	2	3	7	4	4	4	4	4	4	4
2	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
3	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
5	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
6	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
7	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
8	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
9	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
10	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
11	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
12	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
13	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
14	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
15	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
16	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
17	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
18	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
19	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
20	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
21	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
22	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
23	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
24	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
25	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
26	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
27	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
28	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
29	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
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45	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
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\*Mz-4 series melt compositions were calculated as a function of Fo contents of the measured olivine. Major element contents of the olivine by Sobolev et al., 2007.

<sup>1</sup>Z6-Cl- extremely Cl rich melt inclusion

<sup>2</sup>Sc/Y\* melt is calculated as a function of MgO - see Methods) and host olivine major and trace element compositions.