

# Kimberlite melts rich in alkali chlorides and carbonates: A potent metasomatic agent in the mantle

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## ABSTRACT

Kimberlite magmas, as the deepest probe into Earth's mantle (>150 km), can supply unique information about volatile components (hydrogen, carbon, chlorine, sulfur) in mantle-derived melts and fluids. All known kimberlite rocks are not suitable for studies of mantle volatiles because of their pervasive postmagmatic alteration; however, this study discusses an exceptionally fresh group I kimberlites (<0.5 wt% H<sub>2</sub>O) from the Udachnaya-East diamondiferous pipe in Siberia. Kimberlite groundmass, in addition to euhedral olivine and calcite, is extremely enriched (at least 8 wt%) in water-soluble alkali chlorides, alkali carbonates, and sulfates (ratio 5:3:1), and often shows immiscibility textures. A primary magmatic origin of alkali chlorides and alkali carbonates is confirmed by the study of strontium isotopes in the water- and dilute acid-leachates of the groundmass (<sup>87</sup>Sr/<sup>86</sup>Sr = 0.7069 and 0.7050) that contrast with much more radiogenic isotope composition of the Cambrian platform sedimentary rocks and the Udachnaya-East mine-site brines. Melt inclusions in groundmass olivine, composed of halite, sylvite, alkali-Ca carbonates, phlogopite, olivine, and CO<sub>2</sub> fluid, were used to determine the composition and evolution of the kimberlite melt prior to emplacement. Melt inclusions show immiscibility between chloride and carbonate liquids at <600 °C in heating stage experiments. The chloride and carbonate enrichment in the kimberlite parental magma suggests the presence of a powerful agent for chemical modifications (metasomatism) in the mantle and crust.

**Keywords:** kimberlite, mantle volatiles, chlorine, carbonate, metasomatism, melt inclusions, immiscibility.

## INTRODUCTION

Major volatiles (water, carbon dioxide, and chlorine) are the key components in crustal hydrothermal processes and in the formation of many magma-related ore deposits. Although these volatiles were originally delivered from the mantle, only C-H-O volatile species have so far been considered important in mantle processes, such as mantle metasomatism (e.g., transfer of elements, melt-peridotite reactions, and formation of chemical heterogeneities) and melt production, extraction, and transport. Hydrated and carbonated peridotites are thought to be essential components of the continental lithosphere and shallow convecting mantle (e.g., Dautria et al., 1992; Green, 1973; Ionov et al., 1996; Yaxley et al., 1998), and they may also influence lower-mantle compositions through deep recycling in subduction zones. By contrast, Cl-

rich peridotites are not recognized, and very little is known about storage and transport of Cl in the mantle, its influence on partial melting, and its association with other volatiles in deep processes. To date, the only reliable evidence for mantle Cl comes from brine inclusions in diamonds from kimberlites (e.g., Bulanova et al., 1998; Izraeli et al., 2001; Navon et al., 1988; Turner et al., 1990). Kimberlite magmas, derived from Earth's mantle at >150 km (e.g., Dawson, 1980; Haggerty, 1999; Mitchell, 1989), deeper than any other known terrestrial magma type, can be the best available probe into behavior of mantle-derived fluids en route to the surface.

Typical kimberlites are composed of xenocrysts and xenoliths of mantle and crustal origin set in a fine- to coarse-grained groundmass of serpentinized euhedral olivine, calcite, and minor silicate and oxide minerals (e.g., phlogopite, perovskite, Cr-spinel, magnetite). This groundmass, although invariably altered

by secondary processes in almost all kimberlite occurrences (Mitchell, 1989), is thought to represent crystallized kimberlite melt. Here we report on remarkably fresh kimberlites from the Udachnaya-East pipe in Yakutia, Russia, in which the complete absence of alteration has made possible the discovery of significant enrichment of deep-mantle melts in alkali chlorides and alkali carbonates.

## COMPOSITION OF UDACHNAYA-EAST KIMBERLITE

The diamond-productive Udachnaya-East pipe was emplaced ca. 365 Ma (Kinny et al., 1997) into Ordovician sedimentary rocks of the Daldyn-Alakit region of the Siberian craton. The studied kimberlite rocks, obtained from deep levels (~500 m) of the pipe, are exceptionally fresh and show no secondary serpentinization. Their petrographic features, mineral assemblages, key chemical parameters (Table 1; Appendix 1), and initial Sr-Nd isotope compositions (<sup>87</sup>Sr/<sup>86</sup>Sr ≈ 0.7050; ε<sub>Nd</sub> ≈ +4) are typical of most common type I (Mitchell, 1989) or group I (Clement and Skinner, 1985; Smith et al., 1985) kimberlites.

The Udachnaya-East kimberlite groundmass consists of euhedral silicates (olivine, phlogopite, monticellite, and sodalite), oxides (perovskite, spinel, ilmenite, rutile, and titanomagnetite), sulfides (pyrrhotite, djferisherite), chlorides (halite and sylvite), apatite, Na-Ca sulfates, and different carbonates (Table 1; Appendix 1). Among carbonates, Sr-bearing (0.4–0.7 wt% SrO) calcite is dominant, showing intimate association with olivine, phlogopite, sodalite, chlorides, and other carbonates (Fig. 1). Alkali-Ca carbonates have variable compositions in terms of CaO, Na<sub>2</sub>O, and K<sub>2</sub>O (31–43 wt%, 11–21 wt%, and 6–8 wt%, respectively) and significant enrichment in S (≤6.6 wt% SO<sub>3</sub>), F (≤2 wt%), and P (≤6 wt% P<sub>2</sub>O<sub>5</sub>). Another carbonate type is more homogeneous in composition and enriched principally in Na<sub>2</sub>O (10–16 wt%), CaO (40–50 wt%), Cl (≤1.3 wt%), and SrO (0.4–0.6

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TABLE 1. COMPOSITIONS OF BULK GROUNDMASS, GROUNDMASS MINERALS, AND MELT INCLUSIONS

	1	2	3	4	5	6	7	8	9
SiO <sub>2</sub>	26.1	n.d.	0.1	0.2	39.1	0.0	1.1	36.0	17.3
TiO <sub>2</sub>	1.3	n.d.	0.0	0.0	0.0	n.d.	0.0	3.4	0.6
Al <sub>2</sub> O <sub>3</sub>	2.0	n.d.	0.0	0.0	31.0	n.d.	0.0	14.7	2.6
FeO	7.2	n.d.	0.1	0.5	1.2	48.2*	0.9	7.3	4.6
MgO	28.9	0.1*	0.1	0.2	1.8	n.d.	1.5	20.8	16.6
CaO	12.4	0.2*	32.7	42.3	0.3	0.0	33.1	0.0	8.1
Na <sub>2</sub> O	4.3	30.9*	15.5	11.9	19.0	n.d.	13.4	0.5	9.1*
K <sub>2</sub> O	2.1	11.6*	6.5	1.0	0.6	9.4*	8.7	10.3	12.1*
P <sub>2</sub> O <sub>5</sub>	0.4	n.d.	0.9	0.4	n.d.	n.d.	0.2	0.0	0.1
Cl	2.8	33.8	0.0	0.3	7.2	1.5	0.1	0.2	18.5
SO <sub>3</sub>	0.7	6.8	4.3	1.2	0.5	34.1*	3.4	0.1	0.8
CO <sub>2</sub>	10.3	16.6	39.7**	42.1**	n.d.	n.d.	37.7**	n.d.	8.3**
H <sub>2</sub> O	0.5	n.a.	n.d.	n.d.	n.d.	n.d.	n.d.	4.0**	0.7

Note: Columns: (1) Average groundmass composition ( $n = 3$ ) of the Udachnaya-East pipe kimberlites. (2) Average composition ( $n = 3$ ) of H<sub>2</sub>O-soluble component in the kimberlite groundmass, recalculated to 100 wt%. (3–6) Average compositions of some groundmass minerals, associated with chlorides: (3) Alkali-Ca carbonate. (4) Sodium-calcium carbonate. (5) Sodalite. (6) Djerfisherite (5.8 wt% Ni, 0.6 wt% Cu). (7–9) Daughter minerals (7—alkali-Ca carbonate and 8—phlogopite) and bulk composition of (9) the melt inclusion shown in Figures 2F and 2G (calculated by using relative abundances and average compositions of daughter phases).

\*Weight percent of element.

\*\*CO<sub>2</sub> and H<sub>2</sub>O are calculated on the basis of the carbonate and phlogopite (OH + F + Cl = 2.0 atoms per formula unit) stoichiometry; n.d.—not determined, n.a.—not applicable.

wt%), whereas K<sub>2</sub>O, SO<sub>3</sub>, F, and P<sub>2</sub>O<sub>5</sub> (0.3–2.0 wt%, 0.1–0.4 wt%, <0.1 wt%, and <0.06 wt%, respectively) are relatively minor components.

Although chloride and carbonate intergrowths commonly form “cement” for silicates and oxides throughout the kimberlite groundmass, carbonate and chloride are also present in large ( $\leq 1$  cm) segregations (ocelli), often with textures reminiscent of liquid immiscibility. The segregation shown in Figure 1A comprises a central area of halite with droplets of sylvite, calcite, and small sodalite crystals. Adjacent to it is a distinct lens-shaped mass of Na-Ca carbonate that contains numerous blebs of calcite. Minute chloride grains are sprinkled throughout the carbonate mass. The mass of alkali-Ca carbonate together

with calcite and groundmass olivine and phlogopite crystals envelops this segregation of chlorides and Na-Ca carbonates.

The abundance of carbonate, chloride, and sulfate minerals in the groundmass is reflected in high concentrations of CO<sub>2</sub> (10–11 wt%), Cl (2.3–3.2 wt%), and SO<sub>4</sub> (0.5–0.9 wt%), whereas the lack of serpentinization is supported by low H<sub>2</sub>O (<0.5 wt%). Water-soluble components are modally significant (7.3–10.8 wt% of the kimberlite groundmass) and include alkali chlorides, alkali carbonates, and sulfates (~5:3:1, respectively, Table 1; Appendix 1). Other Cl-bearing minerals in the kimberlite groundmass are sodalite (7 wt% Cl) and potassium-nickel-iron sulfide (djerfisherite, 1.5 wt% Cl), always present in close intergrowths with chlorides (Fig. 1B).

## MELT-INCLUSION STUDIES

Euhedral groundmass olivines, thought to have crystallized from the kimberlite melt (Mitchell, 1989; Sobolev et al., 1989), have resorbed cores of variable composition (Fo<sub>86–93</sub>), overgrown by olivine with constant Fo content (89 mol%). Melt inclusions are trapped either individually within the olivine cores, or in healed fractures terminated within the inner rims of olivine Fo<sub>89</sub> (Fig. 2). Only inclusions of these two types are considered truly magmatic, whereas inclusions in fractures connected to the groundmass and inclusions that have undergone decrepitation were excluded from this study. Melt inclusions are often interconnected by thin channels, which suggests possible modification of original melt compositions by necking down. That process could explain why the proportions of fluid and mineral phases in the studied melt inclusions show considerable variability. Fluids are represented mainly by low-density CO<sub>2</sub> bubbles, whereas solid phases are mainly olivine, phlogopite, alkali-Ca carbonates, halite, and sylvite (Figs. 2F, 2G). Calcite, nahcolite (NaHCO<sub>3</sub>), alkali-Ca sulfates, iron-nickel-potassium sulfides, iron-titanium-chromium oxides, apatite, and sodalite are less common. The presence of abundant nonsilicate components in melt inclusions is confirmed by optical heating-stage experiments (Figs. 2C, 2E). Melting in the inclusions starts at 250–300 °C, and two separate melt phases, identified as immiscible carbonate and chloride liquids (according to laser Raman spectroscopy in GeoForschungsZentrum, Potsdam, and proton-induced X-ray emission [PIXE] imaging by a Commonwealth Scientific and Industrial Research Organisation [CSIRO]-GEMOC Nuclear Microprobe, Sydney), and vapor bubbles

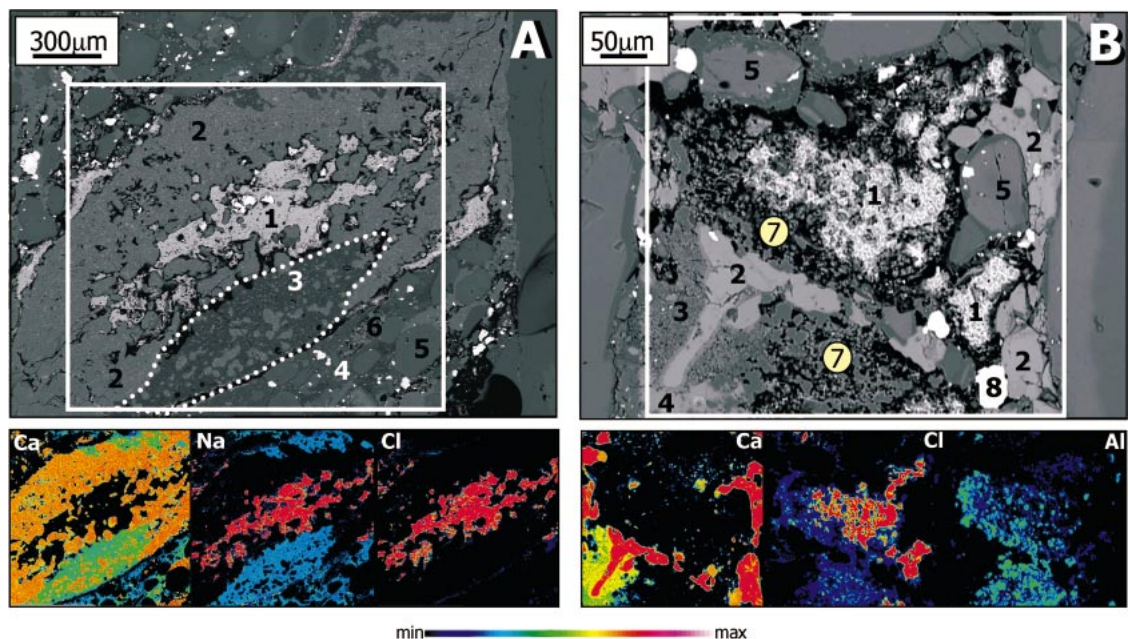


Figure 1. Associated alkali chlorides, alkali-Ca carbonates, and silicates in kimberlite groundmass. Minerals: 1—halite with rounded blebs of sylvite (white), calcite (in A), and sodalite (in B); 2—calcite; 3—sodium-calcium carbonate; 4—alkali-Ca carbonate; 5—olivine; 6—phlogopite; 7—sodalite; 8—perovskite. Small color panels below backscattered electron images are X-ray maps of boxed areas. Note coexistence of three carbonates in lens-shaped segregation (dotted outline).

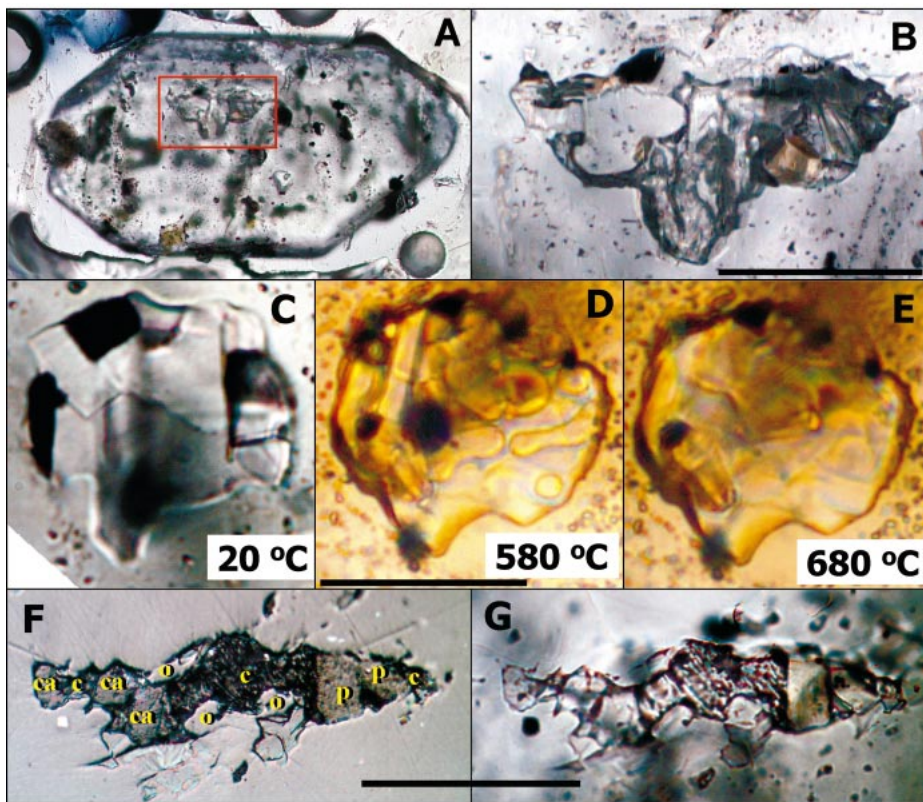
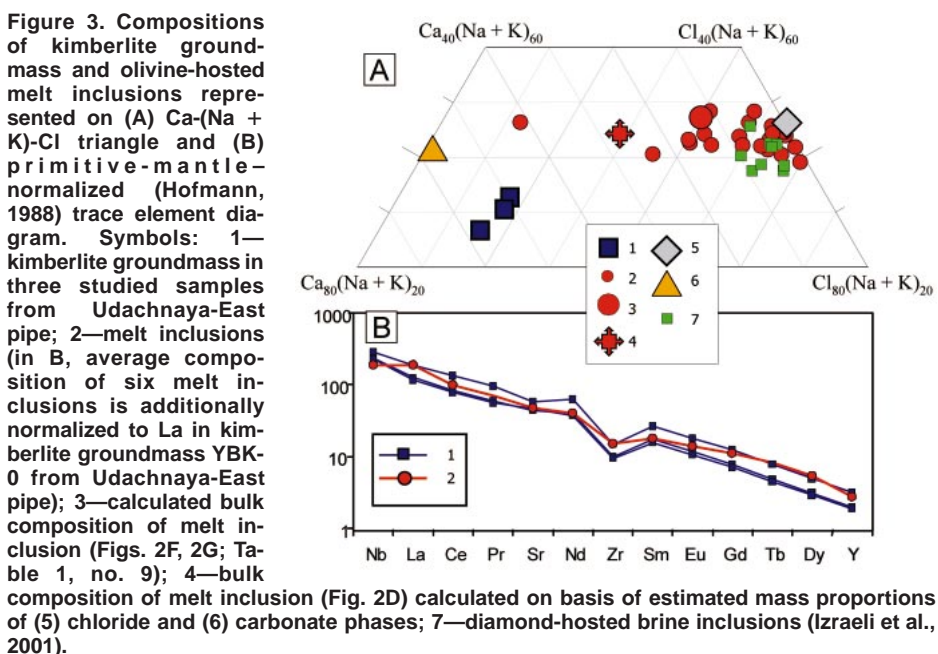


Figure 2. Photomicrographs of (A) groundmass olivine and (B–G) olivine-hosted melt inclusions. Scale bars represent 50  $\mu\text{m}$ . B: Multiphase melt inclusion hosted in core of olivine (boxed in A). C: Typical melt inclusion at room temperature. D: Same inclusion at 580  $^{\circ}\text{C}$  shows immiscibility between carbonate (matrix) and chloride (globules) melt. E: Same inclusion at 680  $^{\circ}\text{C}$  shows complete miscibility and homogenization (transmitted light). Note sculptured surface of melt inclusion at temperature of homogenization. F, G: Multiphase melt inclusion in transmitted and reflected light, respectively. Principal daughter phases: c—sodium-potassium chloride; o—olivine; p—phlogopite; ca—sodium-potassium-calcium carbonate.



are clearly visible at  $\sim 550$ – $600$   $^{\circ}\text{C}$  (Fig. 2D). Bubble dissolution and miscibility of the liquids occur almost simultaneously at  $660$ – $760$   $^{\circ}\text{C}$  (Fig. 2E).

Melt-inclusion compositions, when recast in terms of Ca, Cl, and Na + K, are enriched in Cl and alkalis compared to the kimberlite groundmass (Fig. 3A). Variation in major melt components, tentatively defined as carbonate and chloride, could be a result of postentrapment processes (e.g., necking down), but despite such complications, the melt inclusions share the enriched trace element signature of the kimberlite groundmass (Fig. 3B; Appendix 1).

#### MAGMATIC SOURCE OF CHLORINE

Although the petrographic evidence indicates that the enrichment in alkali chlorides and alkali carbonates in the kimberlite groundmass is of magmatic origin, a possibility of a crustal source of chlorine needs investigation given that the Udachnaya-East kimberlite was emplaced through Cambrian platform sedimentary rocks. Furthermore, Ca-Na-K-Mg-Sr-rich brines are present at the mine site and could be a source of additional contamination. However, despite the very high Sr concentrations in the potential contaminants (e.g., 1000 ppm in the brine), the Sr isotopes in water and dilute-acid leachates of the kimberlite groundmass ( $^{87}\text{Sr}/^{86}\text{Sr} = 0.7069$  and  $0.7050$  before age correction) are much less radiogenic than those in the Cambrian seawater and Siberian platform sedimentary rocks ( $>0.7080$ – $0.7095$ ) (e.g., Montanez et al., 1996; Nicholas, 1996) and in the mine-site brines ( $0.7089$ ). A magmatic source for the Cl is also evidenced by the intergrowths of chlorides and the Cl-bearing minerals (sodalite and djerfisherite) of apparently high-temperature origin (Fig. 1B). Further support for a primary magmatic origin of the alkali chlorides and carbonates is provided by the evidence from melt inclusions in groundmass olivines.

#### DISCUSSION AND CONCLUSIONS

Our analyses of the Udachnaya-East pipe kimberlite differ from typical group I kimberlites in having significant enrichment in alkali chlorides, carbonates, and sulfates (at least 8 wt% in total, Table 1). The amount of alkali chlorides and carbonates in the kimberlite parental melt prior to emplacement could have been even higher than that measured in the groundmass (Table 1). This possibility can be inferred from the composition of pristine melt inclusions (Table 1; Figs. 2 and 3). On the basis of mineral assemblages, liquid-immiscibility textures, and Sr isotope data, we argue for the mantle origin of this unusual enrichment of the otherwise normal kimberlite rocks. The reason that such a significant constituent of the Udachnaya-East pipe kimberlite has not been found in other kimberlites or other mantle-

derived silicate rocks is most likely related to the segregation and flushing out of alkali chlorides and carbonates in the magmatic stage and/or destruction by postemplacement leaching and alteration. By contrast, some nonsilicate, mantle-derived rocks show broadly comparable components. For example, historic natrocarbonatite lavas from the Oldoinyo Lengai volcano (Dawson, 1962) are enriched in chloride, sulfate, and phosphate (Gittins and McKie, 1980; Keller and Krafft, 1990; Mitchell, 1997) and show immiscibility between chloride and carbonate (Mitchell, 1997). We also anticipate that carbonate- and chloride-enriched melt and fluid inclusions in diamonds from kimberlites (e.g., Izraeli et al., 2001; Navon et al., 1988; Schrauder and Navon, 1994), including the Udachnaya-East kimberlite (Bulanova et al., 1998), can match the chloride- and carbonate-enriched kimberlite described here, both compositionally (Fig. 3A) and genetically.

Metasomatic modification of lithospheric mantle by carbon-bearing melts and fluids has been demonstrated in peridotite xenoliths (e.g., Dautria et al., 1992; Ionov et al., 1996; Yaxley et al., 1991, 1998) and experimentally (e.g., Dalton and Wood, 1993; Wallace and Green, 1988; Thibault et al., 1992). Under mantle conditions, the chloride- and carbonate-rich kimberlite, like the one identified in our study, would be highly volatile, mobile, reactive, and capable of pervasively percolating and wetting ambient peridotite (Hammouda and Laporte, 2000; Hunter and McKenzie, 1989). We envisage that such kimberlites, exceptionally enriched in Cl, CO<sub>2</sub>, and alkalis, and the kimberlite-derived chloride- and carbonate-bearing fluids play a previously unrecognized role as a potent metasomatic agent in both mantle and crust.

#### APPENDIX 1. ANALYTICAL METHODS

The Sr-Nd isotope compositions were determined on an NU Instruments multiple-collector inductively coupled plasma-mass spectrometer (ICP-MS) (University of Melbourne). Analytical details and precisions were given in Kamenetsky and Maas (2002). Mineral and melt-inclusion compositions were analyzed by electron microprobe (JEOL JXA-8200, Max Planck Institut für Chemie, Mainz; Cameca SX50, University of Tasmania). Analyses of major elements on fused glass discs (X-ray fluorescence, Phillips 1400) and trace elements (solution ICP-MS, HP 4500) were performed at the University of Tasmania. Soluble components in the groundmass were analyzed by the Analytical Services Tasmania, by ICP-atomic emission spectroscopy (for cations) and ion chromatography (for anions) using standard methods for the examination of water and wastewater proposed by the American Public Health Association (APHA methods). Carbon in the soluble component of the groundmass was determined by Carlo Erba CHNS-O elemental analyzer (University of Tasmania). Unexposed melt inclusions were analyzed by laser-ablation ICP-MS (University of Tasmania). The setup includes a UP213 ultraviolet laser with a custom-built small-volume (~4 cm<sup>3</sup>) ablation cell, coupled to an HP 4500 ICP-MS. Samples

were ablated at 30–60 μm spots, a repetition rate of 5 Hz, and laser energy of 14 J/cm<sup>2</sup>. The measurements were calibrated by using the NIST612 standard.

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#### Notes