



## Olivine zoning and the evolution of kimberlite systems

Andrea Giuliani<sup>1,2</sup>, Ashton Soltys<sup>2</sup>, Emilie Lim<sup>2</sup>, Henrietta Farr<sup>2</sup>, David Phillips<sup>2</sup>,  
Karsten Goemann<sup>3</sup>, William L. Griffin<sup>1</sup>

<sup>1</sup> ARC Centre of Excellence for Core to Crust Fluid Systems (CCFS) and GEMOC, Department of Earth and Planetary Sciences, Macquarie University, Australia  
*andrea.giuliani@mq.edu.au, andrea.giuliani@unimelb.edu.au*

<sup>2</sup> KiDs (Kimberlites and Diamonds), School of Earth Sciences, The University of Melbourne, Australia

<sup>3</sup> Central Science Laboratory, University of Tasmania, Tasmania, Australia

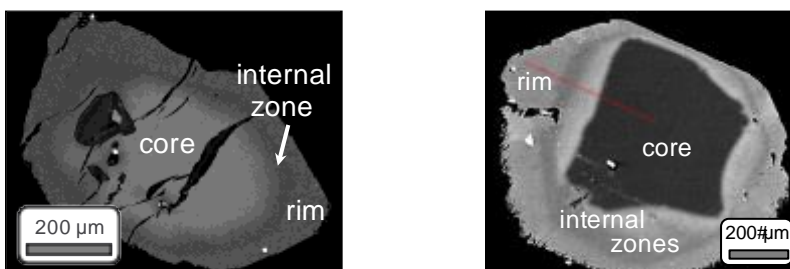
### Introduction

Olivine is the most abundant mineral in kimberlite rocks and generally occurs as macrocrysts (i.e. anhedral grains larger than ~1 mm in size), and (micro-)phenocrysts (i.e. subhedral to euhedral crystals commonly less than 1 mm in size). Recent advances in imaging and microanalytical techniques have confirmed that olivine macrocrysts and the vast majority of phenocrysts contain cores and a rims with distinct compositions (e.g., Kamenetsky et al., 2008; Brett et al., 2009; Giuliani et al., 2017), as per the observations of Boyd and Clement (1977) for olivine grains in the De Beers kimberlite (South Africa). The rims of macrocrysts and phenocrysts typically contain primary inclusions of groundmass phases (e.g., chromite, MUM spinel, Mg-ilmenite), and exhibit homogeneous Mg# composition coupled with decreasing Ni and increasing Ca and Mn concentrations. These features are consistent with a magmatic origin for the rims. Conversely, the olivine cores host inclusions of typical mantle phases (e.g., clinopyroxene, garnet, Cr-spinel), which are not stable in kimberlite magmas. The cores show widely variable compositions extending from those of olivine in lithospheric mantle peridotites (i.e. Mg# ~ 91-94) to compositions richer in Fe. Although there is broad consensus that the olivine cores derive from disaggregation of peridotite wall rocks, the following features have been invoked to suggest that some olivine cores originate from mantle wall rocks formed by earlier kimberlite metasomatism (Arndt et al., 2010; Sobolev et al., 2015; Howarth and Taylor, 2016): 1) low-Mg# cores display unusual compositions unlike typical mantle peridotites; 2) clinopyroxene and garnet inclusions with variable compositions occur in individual cores; and 3) the textural features of some macrocrysts indicate deformation and recrystallisation immediately prior to kimberlite entrainment. The occurrence of phlogopite antecrysts in kimberlites and entrainment of mantle polymict breccia xenoliths (which represent failed kimberlite intrusions at mantle depths), are additional indications that earlier pulses of kimberlite melt 'prime' the mantle before kimberlite eruptions (e.g., Giuliani et al., 2016). It is however unclear if there is any relationship between these kimberlite-related metasomatic processes and the composition of kimberlite magmas that are emplaced at surface. Furthermore, as olivine is a liquidus mineral in kimberlite melts and olivine formation can persist throughout most of the kimberlite crystallisation sequence, deciphering the processes that control the compositional evolution of olivine can provide enhanced constraints on kimberlite petrogenesis.

### Samples, methods and results

We have examined the zoning (SEM-EDS imaging) and major-element compositions (electron microprobe (EMP) analyses) of olivine in archetypal kimberlites from South Africa, Botswana, Lesotho, Canada and Brazil, and compared these results with new and existing data for kimberlites, orangeites and ultramafic lamprophyres from South Africa, Canada, Russia and Greenland. In addition, we have assessed compositional variations across kimberlite grains from the Bultfontein (Kimberley, South Africa) and Udachnaya-East (Russia) kimberlites using detailed EMP traverses.

Olivine macrocrysts and phenocrysts from the kimberlites examined in this study are typically zoned, with a core, rim and one or more internal zone(s) (i.e. intermediate between cores and rims; Fig. 1). The core is often partly resorbed, particularly when enriched in Fe. The (outermost) internal zone hosts chromite inclusions. The compositional features of cores and rims are consistent with previous studies, e.g., variable core Mg# vs constant rim Mg#. Different kimberlite pipes from individual clusters (e.g., Kimberley in South Africa, Ekati in Canada) contain olivine with very similar compositional features (e.g., restricted range of rim Mg#). However, large compositional variations are evident for olivine grains from kimberlite clusters on the same craton and worldwide.



**Figure 1.** SEM-EDS images of zoned olivine grains from the Bultfontein (left panel) and Udachnaya-East kimberlites (right panel; red line: EMP traverse).

The most remarkable finding of this study is the linear correlation between the compositions of olivine cores and rims in kimberlites worldwide, which extends to South African orangeites and carbonate-rich ultramafic lamprophyres (i.e. aillikites) from Greenland. The correlation between average core Mg# (or NiO concentrations) and average rim Mg# values of olivine in kimberlites (orangeites and aillikites) is statistically significant ( $R^2 = 0.85$ ), despite our dataset including samples from five continents and different carbonate-rich magma types. Conversely, olivine grains from the olivine melilitite sample included in this study (Sutherland, South Africa) plot outside this trend.

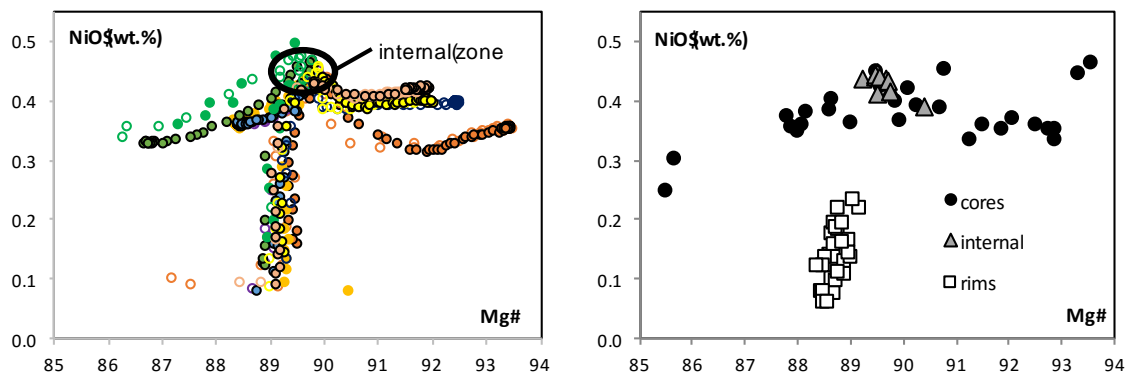
Multiple core-to-rim EMP traverses across 10 olivine grains from the Bultfontein kimberlite exhibit very consistent patterns in Mg# vs Ni, Mn, Ca, or Cr charts, despite significantly different core compositions (Mg# = 86.3-93.5; Fig. 2, left panel). The patterns show a moderately to strongly curved segment, which extends from compositions of the core to the individual internal zone, and is present in every measured grain (e.g., Fig. 1). This internal zone shows remarkably consistent composition (Mg# =  $89.7 \pm 0.3$ ; NiO =  $0.43 \pm 0.02$  wt.%; CaO =  $0.08 \pm 0.01$  wt.%; 1sd) across the ~60 grains measured from this kimberlite. The traverses include a second segment, where Mg# initially decreases to values typical of the rims and then becomes constant, while Ni and Cr decrease, and Ca and Mn increase throughout this segment. Olivine Mg# vs Ni, Mn or Ca charts where cores, rims and internal zones were measured by spot analysis only provide the endmembers of the patterns illustrated by the traverses (Fig. 2, right panel).

SEM-EDS images and EMP traverses across 8 representative grains of the Udachnaya-East kimberlite show considerably more complexity than the Bultfontein olivine (e.g., Fig. 1), including up to 3 internal zones. However, similar to Bultfontein, the outermost (or single) internal zone has consistent composition across the sample suite at higher Mg# (89.5-90.0) and NiO contents (0.33-0.35 wt.%) than the rims.

## Discussion

Olivine in kimberlites worldwide exhibit remarkably consistent geochemical features, i.e. 1) distinct zoning between core, internal zone(s) and rim (plus Mg-enriched outer rinds preserved in some localities; e.g., Kamenetsky et al., 2008; Howarth and Taylor, 2016); 2) variable core compositions; and 3) rims with very restricted Mg# ranges. Our work demonstrates that all zoned grains contain an internal zone (the outermost when more internal zones occur), where compositions do not vary amongst olivine grains

from the same kimberlite. This feature, coupled with inclusions of magmatic chromite, suggests that the (outermost) internal zone represents the liquidus phase of the host kimberlite magmas. Partial resorption of olivine cores and Ni-Cr enrichment of the internal zones (e.g., Fig. 2) indicate that liquidus olivine crystallises after significant assimilation of wall rock material. The correlation between Mg# of (xenocrystic) cores and magmatic rims also suggests that the composition of wall rocks along the kimberlite magma conduit exerts a fundamental control on the composition of the olivine rims and, therefore, kimberlite magmas. This process also applies to other mantle-derived carbonate-rich magmas (orangeites and aillikites). Low-Mg# olivine cores have compositions resembling olivine belonging to the megacryst suite or sheared peridotites, for which a genetic relationship with early kimberlite metasomatism is commonly advocated. We therefore propose that more intense metasomatism of mantle magma conduits by early kimberlitic fluids results in olivine rims and, therefore, kimberlite magmas more enriched in Fe and Mn, and depleted in Mg and Ni.



**Figure 2.** Mg# vs NiO compositions of olivine grains in the Bultfontein kimberlite from electron microprobe traverses (left panel) and spot analyses (right panel). In the left panel, the various traverses are shown using different colours. Internal: internal zones.

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