



Stable isotope (C, O, S) compositions of volatile-rich minerals in kimberlites: A review

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ABSTRACT

The composition of primary kimberlite melts and, in particular, the absolute and relative abundances of volatile components (mainly CO₂ and H₂O) are controversial issues, because kimberlite melts entrain and interact with abundant mantle and crustal xenoliths during ascent, react with wall rocks during emplacement, and lose some of their volatile inventory during pre- and syn-emplacement degassing. Compositional constraints are further complicated by the common alteration of kimberlitic rocks by post-emplacement fluids of various origin (e.g., deuteric, meteoric, hydrothermal). Consequently, the compositions of kimberlitic rocks may not be entirely representative of their parental melts. In kimberlitic rocks, CO₂ is concentrated in carbonate minerals, whereas H₂O is mainly stored in the secondary minerals serpentine and, to a lesser extent, chlorite and brucite, with minor contribution by primary magmatic phlogopite. This review focuses on utility of carbon, oxygen and sulphur stable isotopes to constrain the source of volatiles (i.e. magmatic vs non-magmatic) for carbonate, serpentine, sulphide and sulphate formation and the origin of fluids altering kimberlitic rocks.

A global compilation of kimberlite carbonate data ($\delta^{13}\text{C} = -11.9$ to $+0.2\text{‰}$, median $\delta^{13}\text{C} = -5.0\text{‰}$, relative to VPDB; $\delta^{18}\text{O} = 1.2$ – 26.6‰ , median $\delta^{18}\text{O} = 13.2\text{‰}$, relative to VSMOW) reveals that the majority of results (86%) plot within a range of $\delta^{13}\text{C} \sim -2$ to -8‰ , which is considered representative of mantle carbon, but only 15% of analyses are in the field of oxygen isotopic values for mantle carbonates ($\delta^{18}\text{O} \sim 6$ – 9‰). Variations in kimberlite carbon isotopic compositions occur on regional scales, implying widespread mantle heterogeneity, possibly related to input of carbon from recycled crustal material and/or partial overprinting by secondary processes at the local scale. Carbonates in southern African Group I (or archetype) and Group II kimberlites (or orangeites) show different $\delta^{13}\text{C}$ distributions (median values of -5.3‰ and -6.5‰ , respectively). This is consistent with distinct mantle sources, as demonstrated previously by radiogenic isotope studies. Kimberlite breccia carbonates commonly have higher $\delta^{18}\text{O}$ values than carbonates in massive and hypabyssal kimberlites, which suggests more extensive interaction of kimberlite rocks with hydrous fluids in the brecciated parts of kimberlite pipes. Modelling of the stable isotope compositions of carbonates from the Kimberley, Lac de Gras and Udachnaya-East kimberlites reveals that several processes are capable of modifying these compositions, including interaction with H₂O-rich deuteric (i.e. late-stage magmatic) fluids, meteoric waters and/or hydrothermal fluids, and incorporation of sedimentary material. However, these processes can produce similar variations of the carbonate C–O isotopic compositions, which means that carbonate isotopes alone cannot provide tight constraints on the alteration of kimberlite rocks. Only few carbonates in hypabyssal kimberlites show isotopic compositions consistent with abundant CO₂ degassing (i.e. increasing $\delta^{18}\text{O}$ with decreasing $\delta^{13}\text{C}$ values), thus implying that kimberlite magmas that are not emplaced explosively retain most of their CO₂ concentrations prior to carbonate crystallisation.

In kimberlitic rocks early-formed serpentine exhibits higher $\delta^{18}\text{O}$ values ($\sim +4$ – $+6\text{‰}$) than later serpentine rims and segregations ($\delta^{18}\text{O}$ values as low as $\sim -2\text{‰}$). These variations are consistent with serpentine crystallisation from hydrous fluids derived from mixing between deuteric fluids and meteoric/hydrothermal fluids, with progressive enrichment in the latter component. Serpentine is considered to have formed under hydrothermal conditions when externally derived hydrous fluids infiltrated the cooling kimberlite volcanic system.

Only limited sulphur isotopic data are available for kimberlitic bulk rocks and sulphide and sulphate phases. Of these, relatively few sulphur isotopic ratios approach the $\delta^{34}\text{S}$ values considered representative of the mantle

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($0 \pm 2\%$, relative to VCDT). Elevated $\delta^{34}\text{S}$ values ($\sim 14\%$) characteristic of sulphates in the Udachnaya-East kimberlite are consistent with equilibration with sulphides ($\delta^{34}\text{S} \sim 1\text{--}2\%$) at temperatures of $\sim 500\text{--}550^\circ\text{C}$, after kimberlite melt outgassing under oxidising conditions. Conversely, the large $\delta^{34}\text{S}$ range shown by some southern African and Yakutian kimberlites ($-3\text{--}+12\%$ and $+15\text{--}+53\%$, respectively) may be largely due to alteration and crustal contamination.

In conclusion, the stable isotopic compositions of carbonates, serpentine and S-rich minerals in kimberlites, can be used in conjunction with detailed petrographic and geochemical analyses, to constrain processes affecting kimberlite magmas prior to, during, and subsequent to crystallisation. The available stable isotopic data indicate that externally derived (i.e. non-magmatic) hydrothermal fluids have affected the compositions of most kimberlites, including the hypabyssal varieties often used to reconstruct the compositions of primary kimberlite melts. This discrepancy remains a major obstacle in the quest for the primary composition of kimberlite melts.

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1. Introduction

Kimberlites are enigmatic, rare, small volume igneous rocks that are important because they are the primary host rock to diamonds and because kimberlite parental melts originate from deep within the Earth ($>150\text{ km}$, i.e. within the diamond stability field – e.g., Haggerty, 1994; Ringwood et al., 1992; Tainton and McKenzie, 1994; Torsvik et al., 2010). In addition, kimberlite magmas have entrained abundant fragments of mantle and deep crust wall rocks en route to the surface, thus providing the major source of information about the petrology and geochemistry of the deep lithosphere in continental areas (e.g., Dawson, 1980; Menzies and Hawkesworth, 1987; Nixon, 1987; Pearson et al., 2003; Schmitz and Bowring, 2003a,b; Zartman et al., 2013; Giuliani et al., 2014). Due to their hybrid and volatile-rich nature and widespread alteration by deuteric (i.e. late-stage magmatic), meteoric and hydrothermal fluids, the primary composition of kimberlites has proven difficult to constrain. A number of studies have employed stable isotopes to provide insights into the evolution of kimberlite magmas upon emplacement, with particular emphasis on the source of fluids involved during alteration of kimberlite rocks (e.g., Vinogradov and Ilupin, 1972; Sheppard and Dawson, 1975; Kobelski et al., 1979; Ukhonov et al., 1986; Kirkley et al., 1989; Ustinov et al., 1994; Wilson et al., 2007; Mitchell, 2013). Carbon and oxygen stable isotopes have been widely used to constrain the source of CO_2 and H_2O , i.e. the most abundant volatile species, in kimberlites.

This review critically evaluates the existing data for C and O isotopes in kimberlite carbonates, the O isotopes of serpentine and the S isotopes

of sulphides and bulk kimberlitic rocks. A large number of analyses (>500) exist for carbonates, but limited data are available for serpentine and sulphide/sulphate minerals in kimberlites. After introducing the main geological, mineralogical and geochemical features of kimberlites, we will define the range of O–C–S stable isotopic compositions shown by mantle rocks. Detailed examination of carbonate data for kimberlite pipes and clusters reveal that kimberlite carbonates are ubiquitously affected by interaction with mixtures of deuteric and meteoric fluids and that the C and O isotopic compositions of only some carbonates reflect CO_2 degassing. Conversely, the O isotopes of serpentine provide evidence that most of the water in kimberlites is of external derivation. Finally, we will summarise the most important processes that affect the S isotopes of magmatic rocks and discuss existing data for kimberlite rocks and the implications for kimberlite petrology.

1.1. Geology, mineralogy and geochemistry of kimberlites

Kimberlites are silica-poor, volatile-rich igneous rocks of variable but broadly ultrabasic composition that mainly occur as volcanic pipes and hypabyssal intrusions in cratonic areas. Two distinct groups of kimberlites have been recognised based on mineralogy, major and minor elements and radiogenic isotope compositions. Kimberlites *sensu stricto* or Group I kimberlites (Smith, 1983) are hybrid olivine-rich rocks consisting of fragments of rocks and minerals of mantle and crustal origin intermixed in a matrix of carbonates, olivine, phlogopite, spinel, perovskite, apatite, serpentine and other minor phases, which crystallised from volatile-rich melts and fluids (Dawson, 1980; Mitchell, 1986,

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