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Valentin Casola, Lyderic France, Albert Galy, Nordine Bouden, Johan Villeneuve. No evidence for carbon enrichment in the mantle source of carbonatites in eastern Africa. *Geology*, 2020, 48 (10), pp.971-975. <10.1130/G47629.1>. <hal-02950427>

HAL Id: hal-02950427

<https://hal.science/hal-02950427v1>

Submitted on 27 Sep 2020

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No evidence for carbon enrichment in the mantle source of carbonatites in eastern Africa

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ABSTRACT

Carbonatites are unusual, carbon-rich magmas thought to form either by the melting of a carbon-rich mantle source or by low-degree partial melting of a carbon-poor (<80 ppm C) mantle followed by protracted differentiation and/or immiscibility. Carbonate-bearing mantle xenoliths from Oldoinyo Lengai (East African Rift), the only active volcano erupting carbonatites, have provided key support for a C-rich mantle source. Here, we report unique microscale O and C isotopic analyses of those carbonates, which are present as interstitial grains in the silicate host lava, veins in the xenoliths, and pseudo-inclusions in olivine xenoliths. The $\delta^{18}\text{O}$ values vary little, from 19‰ to 29‰, whereas $\delta^{13}\text{C}$ values are more variable, ranging from -23‰ to +0.5‰. We show that such carbonate $\delta^{18}\text{O}$ values result from the low-temperature precipitation of carbonate in equilibrium with meteoric water, rather than under mantle conditions. In this framework, the observed $\delta^{13}\text{C}$ values can be reproduced by Rayleigh distillation driven by carbonate precipitation and associated degassing. Together with petrological evidence of a physical connection between the three types of carbonates, our isotopic data support the pedogenic formation of carbonates in the studied xenoliths by soil-water percolation and protracted crystallization along xenolith cracks. Our results refute a mechanism of C enrichment in the form of mantle carbonates in the mantle beneath the Natron Lake magmatic province and instead support carbonatite formation by low-degree partial melting of a C-poor mantle and subsequent protracted differentiation of alkaline magmas.

INTRODUCTION

Carbonatites, which are unusual magmas containing 50% magmatic carbonate and less than 20% SiO_2 (Le Maitre, 2002), have been emitted throughout Earth's history and on all continents (Woolley and Kjarsgaard, 2008), but they are generally observed in rift zones and associated with a metasomatized mantle. As carbonatites are enriched in volatile and incompatible elements, generally interpreted as evidence of low degrees of melting, they have been associated with restricted geodynamic conditions (e.g., Kogarko and Veselovskiy, 2019). The carbon content of their parental metasomatized mantle has important implications for Earth's carbon cycle, yet it remains unconstrained (Kjarsgaard and Hamilton, 1989; Ling et al., 2013). Carbonatites and associated alkaline magmas also represent the world's main rare earth element (REE)

deposits (e.g., Verplanck et al., 2014). Despite their scientific and economic importance, the processes generating carbonatitic melts and the possible C enrichment of their mantle source remain debated. The melting of a C-rich mantle source was experimentally shown to directly produce magmas of carbonatitic nature (e.g., Hammouda et al., 2014). Alternatively, a C-poor mantle source would require the silicate parental melts to either be affected by crustal contamination or undergo extreme differentiation to eventually obtain immiscible silicate and carbonatite melts (Peterson, 1989; Dawson, 2012). Indeed, the strongly incompatible nature of CO_2 implies that the very first partial melts produced from a C-poor source should be enriched in C (e.g., Hirschmann, 2010; Stamm and Schmidt, 2017).

Oldoinyo Lengai (OL), in the East African Rift (EAR), is the only active volcano erupting carbonatites, thus representing an invaluable natural laboratory for studying the genesis and evolution of carbonatitic melts. The amount of

carbon in the OL mantle source remains controversial. As an example, CO_2 is present as a volatile phase in mantle xenoliths from Ethiopia (Frezza et al., 2010), suggesting, at least locally, the mobilization of significant amounts of C in the EAR lithosphere. Nevertheless, similar observations are lacking in Tanzania. A striking observation is that mantle xenoliths collected around OL bear carbonate inclusions and veins, apparently suggesting strong C enrichment in the local mantle (Lee et al., 2000). This is a remarkable feature because carbonates have only rarely been reported in mantle materials (e.g., Ionov et al., 1996; Lee et al., 2000), although this may in part be due to decomposition of carbonates during entrainment, as experimentally demonstrated (Canil, 1990). The presence of solid carbonates in a spinel-bearing peridotite field is also remarkable. At such mantle conditions, Mg-rich carbonates are unstable, and C is rather expected as a fluid phase (Wallace and Green, 1988). However, this may not be suitable in the EAR, where calcites are reported (Lee et al., 2000; Mattsson et al., 2013). For those various reasons, carbonatite formation from a carbonate- or C-rich source is still seriously discussed. On the other hand, modeling of the volatile contents in the mantle source, based on the noble gas and CO_2 contents of OL summit fumaroles, suggests that the mantle C content is low (<80 ppm C; as reassessed from Fischer et al., 2009). Moreover, the $^3\text{He}/^4\text{He}$ ratios of igneous minerals imply that the parental melts of the OL carbonatites were derived from the depleted convective mantle and interacted with the subcontinental lithospheric mantle without any crustal contamination (Mollex et al., 2018). These results suggest that OL carbonatites form by low-degree partial melting of a C-poor mantle, associated with protracted differentiation and liquid immiscibility.

In light of these contradictory results, and because outcrops containing mantle xenoliths generally show posteruption pedogenic

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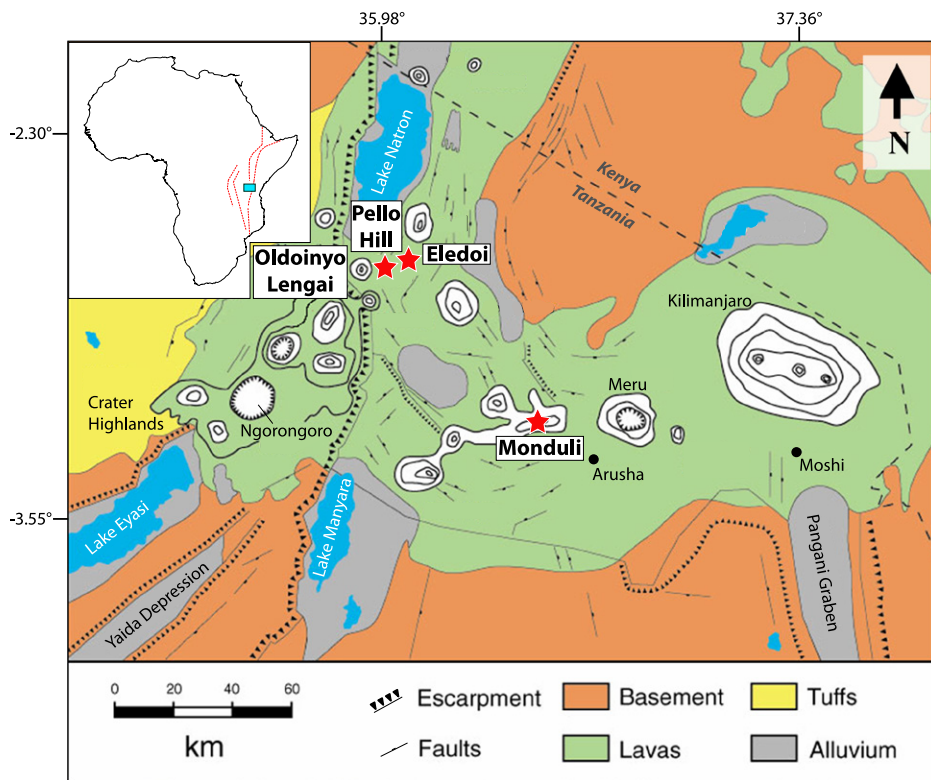


Figure 1. Regional geological map of the Natron Lake magmatic province (from Kervyn et al., 2008). Stars—sampling locations. Inset: Location of study area (blue box) in Africa; red lines—main structures of the East African Rift.

carbonates (Fig. S1 in the Supplemental Material¹), the mantle origin of carbonate inclusions and veins hosted in northern Tanzanian mantle xenoliths is questionable. Here, we employed petrological and in situ geochemical and isotopic analyses of carbonates in five Tanzanian mantle xenoliths to determine the origin(s)

¹Supplemental Material. Additional information on sample location, sample description, carbon and oxygen analytical methods, and the Rayleigh distillation model. Please visit <https://doi.org/10.1130/GEOL.S.12417671> to access the supplemental material, and contact editing@geosociety.org with any questions.

of the carbonates and whether they could be present in the mantle source of OL carbonatites.

METHODS AND RESULTS

Xenolith Samples

Among 110 peridotites that we collected in the Natron Lake magmatic province (<10 km from OL; northern Tanzania, EAR; Fig. 1), 10 hosted carbonate phases. Clinopyroxene grains in protogranular and porphyroclastic peridotites were anhedral diopside containing up to 4.5 wt% Cr₂O₃, indicating their metasomatic origin (O'Reilly and Griffin, 1988; Table S3). A marked hydrous metaso-

matic stage was recorded by both intergranular grains and veins of phlogopite ± amphibole, consistent with previous studies of the regional mantle (e.g., Dawson and Smith, 1988). Hydrous minerals were free of textural deformation. We focused on three representative samples and two samples from Lee et al. (2000), collected in the same area. Detailed sample petrography is presented in the Supplemental Material.

Carbonate Petrography

Carbonates were observed in all mantle lithologies (lherzolite, wehrlite, harzburgite, and dunite), whether strongly metasomatized or free of metasomatic evidence. Carbonates were mostly present as inclusions in olivine, but also as veins crosscutting the xenolith, and locally as patches in the host silicate lava (Fig. 2). The latter were linked to external pedogenic carbonates locally invading the outcrops (Fig. S1). Round carbonate inclusions commonly occurred in large olivines, previously interpreted as evidence for C-rich melt percolation at mantle levels (Lee et al., 2000). Carbonate inclusions were observed to be monocrystalline to polycrystalline, and single grains reached up to 200 μm. Numerous cracks and fractures extending to the exteriors of host olivines imply that none of the observed carbonate inclusions were isolated from external grain boundaries. The analyzed carbonates were calcite in composition (see Supplemental Material for further details).

Carbonate Isotopic Geochemistry

Carbonate C and O isotopic compositions were determined in situ using a Cameca IMS-1270 E7 at the Centre de Recherches Pétrographiques et Géo-chimiques (Nancy, France). Typical internal errors for C and O isotopes measurements were ~0.30‰ and ~0.15‰ (2σ), respectively; see Supplemental Material for analytical details. We performed 76 analyses across the five xenoliths: 8 on carbonates in the

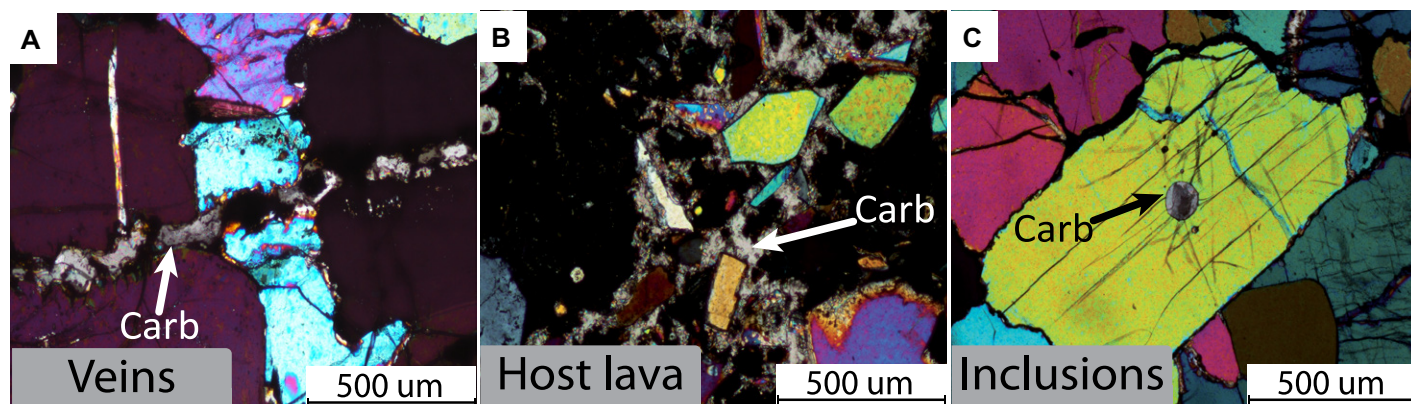


Figure 2. Cross-polarized light microphotographs showing three types of carbonates (Carb). (A) Carbonate vein crosscutting olivines in dunite 19B. (B) Patches of microcrystalline carbonate grains in host lava of wehrlite 24C4. (C) Monomineralic carbonate pseudo-inclusions in olivine in lherzolite 24A4. Note the multitude of cracks connecting pseudo-inclusion to grain boundaries.

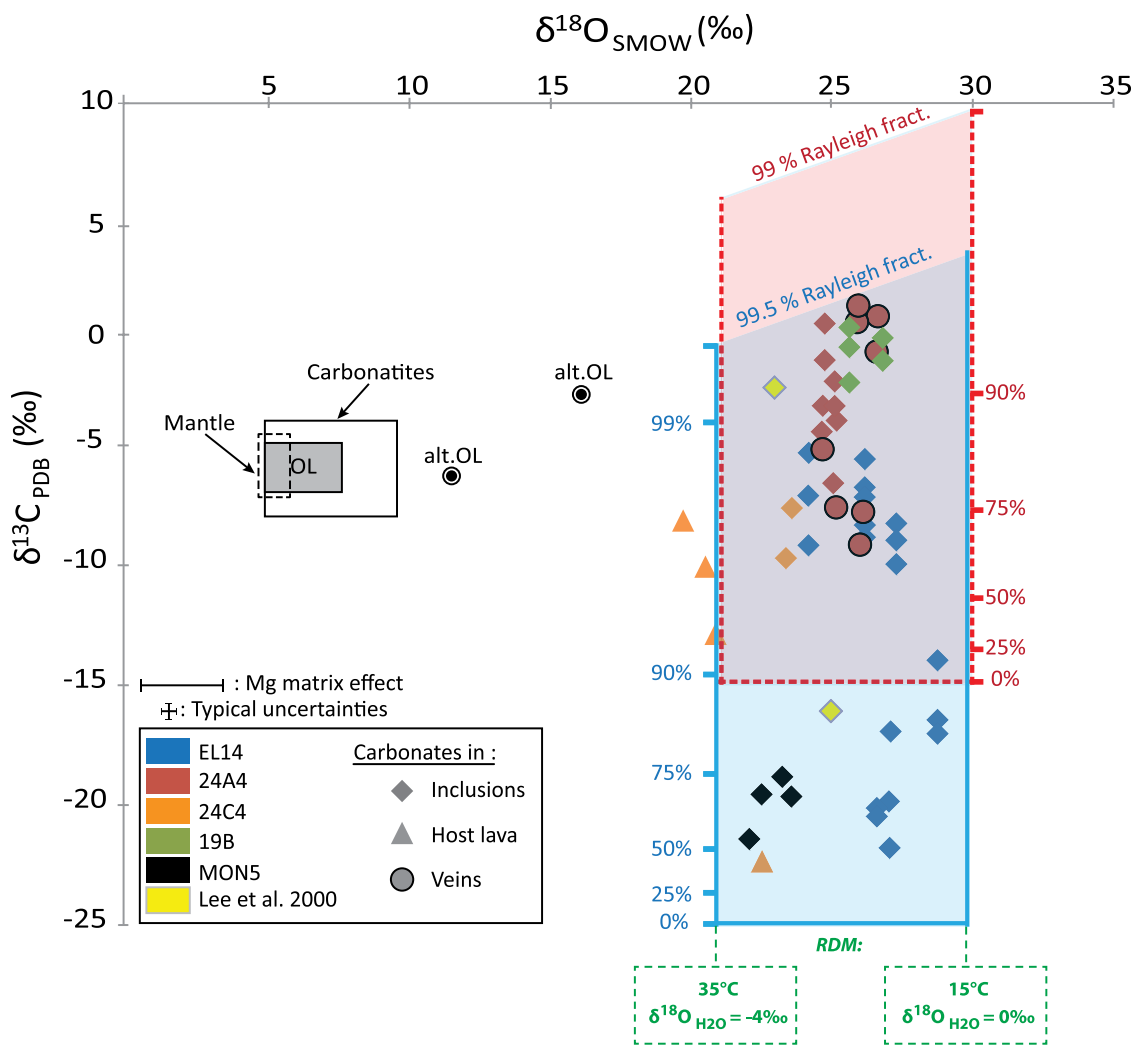


Figure 3. Carbon and oxygen isotopic compositions of carbonates in mantle xenoliths (typical uncertainties shown by cross). OL—fresh natrocarbonatites from Oldoinyo Lengai; alt.OL—altered Oldoinyo Lengai natrocarbonatites; blue and red shaded areas correspond to Rayleigh distillation model (RDM) presented herein (see related references in the Supplemental Material [see footnote 1]). PDB—Pee Dee belemnite; SMOW—standard mean ocean water.

host lava (one sample, four zones; $\delta^{18}\text{O}$, $n = 4$; $\delta^{13}\text{C}$, $n = 4$), 13 on carbonate veins (one sample, two veins; $\delta^{18}\text{O}$, $n = 5$; $\delta^{13}\text{C}$, $n = 8$), and 55 on carbonate inclusions in olivine (five samples, seven inclusions; $\delta^{18}\text{O}$, $n = 16$; $\delta^{13}\text{C}$, $n = 39$) (Fig. 3; Fig. S2; Tables S1 and S2). Carbonates in the host lava were analyzed to compare the isotopic compositions of surface carbonates with veins and inclusions inside the xenoliths.

Overall, carbonate $\delta^{18}\text{O}$ values varied little, from 19‰ to 29‰. Veins and inclusions had $\delta^{18}\text{O}$ values between 22‰ and 29‰, i.e., clearly higher than the mantle O isotopic signature ($5.5\text{‰} \pm 0.4\text{‰}$; e.g., Matthey et al., 1994) and the primary carbonatite field (Fig. 3). Carbonate $\delta^{13}\text{C}$ values were more variable, ranging from -23‰ to $+0.5\text{‰}$. Samples EL14 and 24A4 showed large variations from $-22.3\text{‰} \pm 0.4\text{‰}$ to $-9.1\text{‰} \pm 0.3\text{‰}$ and from $-9.3\text{‰} \pm 0.5\text{‰}$ to $0.1\text{‰} \pm 0.5\text{‰}$, respectively. The two carbonate inclusions analyzed by Lee et al. (2000) had O and C isotopic compositions consistent with our measurements, and these were included in the general trend, which displays a strong $\delta^{13}\text{C}$ variation over relatively restricted $\delta^{18}\text{O}$ values ($24\text{‰} \pm 4\text{‰}$; Fig. 3).

DISCUSSION

Origin of the Carbonate Isotopic Compositions

The C isotopic signature of the mantle is recognized as $\delta^{13}\text{C} \approx -5\text{‰}$, although $\delta^{13}\text{C}$ values as low as -38.5‰ have been observed in mantelic diamonds (e.g., Cartigny, 2005). Nevertheless, $\delta^{13}\text{C}$ values as low as -23‰ (e.g., samples EL14, MON5; Fig. 3) have only been observed in reduced mantle C (diamonds, graphite, and C dissolved in mantle minerals; Deines, 2002) and have never been reported in mantelic carbonates.

Although the presence of round carbonate inclusions in primary minerals strongly suggests a mantle origin (e.g., Lee et al., 2000), their C and O isotopic compositions are radically different from any known carbonate mantle signature, possibly suggesting a surficial origin. Below 100 °C, O diffuses too slowly in calcite (Cherniak, 2010) to explain such high $\delta^{18}\text{O}$ values ($\sim 25\text{‰}$) by the re-equilibration of mantle carbonates with surface fluids. In addition, the presence of pedogenic carbonates locally invading the volcanic outcrops (Fig. S1) opens the possibility of carbonate precipitation from fluids percolating throughout fractures in the mantle

xenoliths after their eruption at the surface. Next, we modeled surficial carbonate formation from groundwater-rock interaction and compared our model with the measured compositions.

The carbonate O isotopic compositions reported herein ($24\text{‰} \pm 4\text{‰}$; Fig. 3) can be reproduced by low-temperature equilibrium carbonate precipitation from groundwater. Given the average $\delta^{18}\text{O}$ value of rainfall in Tanzania (-4‰ to 0‰ ; e.g., Bowen, 2010) and temperatures of 15–35 °C, modeled carbonate $\delta^{18}\text{O}$ compositions vary between 22‰ and 30‰, perfectly matching the compositions of the carbonates analyzed herein.

In volcanic settings, the source of C in groundwater is generally buffered by organic matter (OM) degradation during its path to shallow soil layers within the volcano slope, where the xenoliths were sampled (e.g., Lloret et al., 2011). C3 and C4 plants (average $\delta^{13}\text{C} \approx -25\text{‰}$ and -15‰ , respectively) dominate the African ground surface (Fig. 4; e.g., Cerling et al., 2011), and their degradation in soils is the ^{13}C -depleted source responsible for the low $\delta^{13}\text{C}$ value of groundwater. However, near-surface groundwaters rapidly saturate in dissolved inorganic

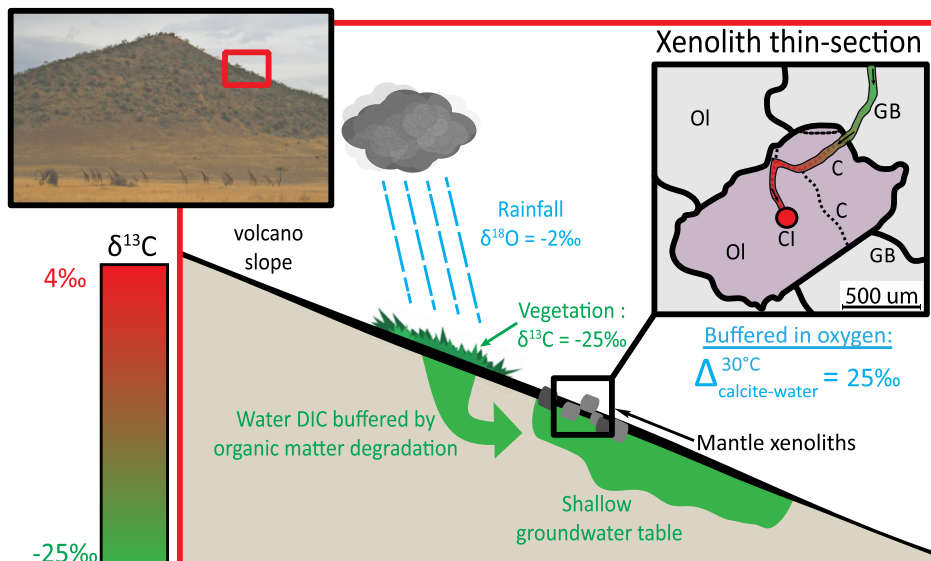


Figure 4. Conceptual model for the formation of carbonates in northern Tanzanian mantle xenoliths. Dissolved inorganic carbon (DIC) in rainfall-fed groundwater is buffered by organic matter degradation. Our Rayleigh distillation model (RDM) corresponds to percolating fluids that progressively fractionate $\delta^{13}\text{C}$ during carbonate crystallization in xenoliths' host lavas, veins, and eventually in pseudo-inclusions. Insert picture is from Pello Hill, with giraffes for scale. Ol—olivine, C—crack, Ci—carbonate inclusion, GB—grain boundary.

carbon (DIC), reaching calcite saturation. Carbonate precipitation from low-C fluids in soils can sustain OM degradation, buffering the C isotopic composition of the fluid, but once the fluid moves out of the soil layers, protracted carbonate crystallization will lower the DIC concentration of the fluid. We thus modeled the concurrent C isotopic evolution of the fluids and carbonates using a Rayleigh distillation model (RDM), with the O isotopic composition of the fluid buffered by the water molecule (typical groundwater O/C ratio $>5 \times 10^3$). Our distillation model considered the reaction $\text{Ca}^{2+} + 2\text{HCO}_3^- \leftrightarrow \text{CaCO}_3 + \text{H}_2\text{O} + \text{CO}_2$, accounting for the precipitation of calcite associated with CO_2 degassing, and it used a temperature-dependent isotopic fractionation factor of 0.9946 at 15 °C and 0.9955 at 35 °C (see the Supplemental Material for details on RDM).

Carbonate Formation Model

Our model considered that meteoric water (with no significant C content) infiltrates the soil and then is stored in shallow groundwater tables where xenoliths are present; the $\delta^{13}\text{C}$ value of DIC in the infiltrating water is buffered by OM. Groundwater circulation can therefore precipitate carbonate throughout the outcrops, and thus within cracks and other pathways in the mantle xenoliths. The percolating fluids progressively precipitate carbonate, and Rayleigh distillation triggers the strong C isotopic variability observed in the carbonates (Fig. 3). This model eventually reproduces the isotopic compositions of the carbonates present in the mantle xenoliths. The amount of DIC remaining in the fluid after carbonate precipitation is nevertheless rather small, since the highest observed carbonate $\delta^{13}\text{C}$ value

would require that $>99\%$ of the DIC is removed by the combined effects of carbonate precipitation and degassing in the case of C3 plants ("RDM" in Fig. 3). C-rich surface continental waters in exchange with the atmosphere tend to be undersaturated with respect to carbonate at DIC concentrations $<10^{-3}$ mol/L (e.g., Appelo et al., 2014). A 99% distillation induced by precipitation of carbonate and associated degassing would require a DIC concentration in excess of 10^{-1} mol/L in the soil aquifer prior to precipitation. Such high values have never been reported when DIC is sourced from the degradation of C3 OM, and 90% distillation (Fig. 3) is likely a maximum value. However, if the OM is dominated by C4 plants, the extreme carbonate $\delta^{13}\text{C}$ values can be achieved with $\sim 90\%$ distillation (Fig. 3). Our model thus shows that the carbonates formed following several episodes of soil-water percolation (with periods dominated alternatively by C3 or C4 plants) associated with protracted crystallization along cracks and in successive pseudo-inclusions within a given xenolith. Therefore, the observed carbonates do not require any primary mantle carbonate in the Natron Lake magmatic province.

Origin of Carbonatites

The origin of the C enrichment in carbonatite magmas, either from the melting of a C-rich source (e.g., Hammouda et al., 2014) or from low-degree partial melting of a C-poor mantle followed by protracted differentiation and/or immiscibility (e.g., Fischer et al., 2009), if not crustal contamination (Jyotiranjana et al., 1999), remains controversial. Crustal contamination at OL has been examined and recently rejected based on the He isotopic compositions of fuma-

roles, crustal cumulates, and mantle xenoliths (Mollex et al., 2018). Our results find no evidence for C enrichment in the form of mantle carbonates in the mantle beneath the Natron Lake magmatic province. Although the mantle domain is carbonate free, C-rich melts may alternatively form by carbonation during redox melting if graphite or diamond is present in the mantle (Stagno et al., 2013). However, as only one unique report of diamond exists in the EAR area (Stachel et al., 1998), it seems very unlikely that abundant carbonatite volcanism could have been generated by redox melting. Consequently, and because carbonates in mantle xenoliths at OL (previously a key argument for a C-rich source) formed after xenolith emplacement at surface conditions, and thus do not require a C-rich mantle, we can now, and for the first time, rule out both C enrichment of the mantle source and crustal contamination at the single active carbonatite province on Earth.

Although C poor, the mantle domain beneath the EAR, and therefore the mantle source of OL carbonatite magmas, is strongly metasomatized by silicate, hydrous, and carbonatite melts, giving it its fertile character (e.g., Dawson and Smith, 1988; Aulbach et al., 2011; Baptiste et al., 2015; Mollex et al., 2018). After their production by low-degree partial melting of a metasomatized (i.e., fertile) mantle domain, carbonatite parental silicate melts undergo protracted differentiation, enriching the magmas in C and other incompatible elements (e.g., Dawson and Smith, 1988; Kjarsgaard and Hamilton, 1989). This enrichment stage is also likely at least partly responsible for the high REE concentrations of carbonatites, the world's main REE deposits.

ACKNOWLEDGMENTS

We thank E. Deloule for constructive discussions, R. Rudnick for providing samples from Lee et al. (2000), and S. Aulbach and two anonymous reviewers. We thank the Tanzania Commission for Science and Technology (COSTECH) for field permits. This work was supported by the Région Grand-Est and the French National Research Agency through the national program "Investissements d'Avenir" (ANR-10-LABX-21-01/LABEX RESSOURCES21) and project GECO-REE, "Genesis and evolution of carbonatites, the main rare earth element deposits: Quantification of the concentration processes from source to metal-rich magma" (ANR-16-CE01-0003-01). This is Centre de Recherches Pétrographiques et Géochimiques contribution 2996, and GECO-REE contribution 4.

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