



# What coupled cerium and neodymium isotopes tell us about the deep source of oceanic carbonatites



Régis Doucelance<sup>a,b,c,\*</sup>, Nina Bellot<sup>a,b,c</sup>, Maud Boyet<sup>a,b,c</sup>, Tahar Hammouda<sup>a,b,c</sup>,  
Chantal Bosq<sup>a,b,c</sup>

<sup>a</sup> Clermont Université, Université Blaise Pascal, Laboratoire Magmas et Volcans, BP 10448, F-63000 Clermont-Ferrand, France

<sup>b</sup> CNRS, UMR 6524, LMV, F-63038 Clermont-Ferrand, France

<sup>c</sup> IRD, R 163, LMV, F-63038 Clermont-Ferrand, France

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## ABSTRACT

We present the first Ce isotope compositions together with Nd and Pb isotope ratios measured for whole rocks and mineral separates (apatite, titanite) from oceanic carbonatites and associated silicate samples from the Cape Verde and Canary archipelagos (Atlantic Ocean). We compare them with measurements performed on carbonatites from Tamazert (Morocco) for which a common source has been suggested. We couple the La/Ce and Sm/Nd systems in order to model the rare-earth-element concentrations and the Ce–Nd isotope compositions of marine carbonates and mantle and/or mantle-derived reservoirs, notably the D'' boundary layer. We discuss the two contrasting models that have been previously presented for the origin of the carbon of the oceanic carbonatite source: recycled marine carbonates via subduction vs. primordial carbon. Our measurements favour the recycling of marine carbonates in the source of oceanic carbonatites. We use Pb isotope ratios together with experimental petrology constraints to discuss the age and depth of carbonate recycling in the mantle.

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## 1. Introduction

Carbonatites are rare, magmatic rocks consisting mainly of calcium- (calcite), calcium and magnesium- (dolomite) or iron- and magnesium carbonates (sidero-magnesite), accompanied by accessory minerals (silicates and/or phosphates, oxides, sulphides, sulphates, etc.). They have been found on every continent (Woolley and Kjarsgaard, 2008). Geochemical studies based on stable, radiogenic and noble gas isotopes have led to the general consensus that their parental magmas originate in the mantle (Bell and Blenkinsop, 1987; Bell and Tilton, 2001; Cartigny et al., 1998; Deines, 1989; Marty et al., 1998; Nelson et al., 1988; Sasada et al., 1997). Such an origin is in agreement with petrographical observations, since xenocrysts from the mantle, generally chromite spinels, are found in some extrusive carbonatites (Bailey, 1989; Mourtada et al., 1997). This is also in agreement with results of experimental petrology: carbonated melts can be generated by melting of a mixture of peridotite and CO<sub>2</sub> (Eggler, 1978; Wallace and Green, 1988; Wyllie and Huang, 1976; Wyllie et al., 1983).

Experimental petrology predicts that carbonated melts formed in the mantle should be magnesio-carbonatites (dolomitic), but most sampled carbonatites are calcic (Woolley and Kempe, 1989; Woolley and Kjarsgaard, 2008). Thus it has been suggested that magnesio-carbonatites are enriched in calcium during their ascent by reaction with their surrounding mantle environment (Dalton and Wood, 1993; Hammouda et al., 2014). An alternative solution considers that the source of carbonatites consists instead of recycled oceanic crust + carbonated sediments. Melting experiments using such materials as their starting composition have led to the generation of calcitic, carbonatitic melts in subduction P–T conditions (Hammouda, 2003; Kiseeva et al., 2012). In this scenario all carbonatite sources should contain recycled oceanic crust.

Oceanic carbonatites are less frequent than their continental counterpart. They have been reported only at the Cape Verde and Canary archipelagos, Atlantic Ocean (Woolley and Kjarsgaard, 2008). The mantle origin of their parental magmas is attested (Lancelot and Allègre, 1974). However, contrasting models have been proposed for the origin of the carbon of their source. They involve either primordial carbon or marine carbonates that have been recycled via subduction. The genetic models also cover a wide spectrum, from a deep mantle origin, which might contain recycled carbon (see De Ignacio et al., 2006, 2012; Hoernle et al., 2002; Mata et al., 2010; Mourão et al., 2010, 2012), to a more superficial

\* Corresponding author at: Clermont Université, Université Blaise Pascal, Laboratoire Magmas et Volcans, BP 10448, F-63000 Clermont-Ferrand, France.

E-mail address: R.Doucelance@opgc.univ-bpclermont.fr (R. Doucelance).

one (lithosphere previously metasomatized by carbonatite liquids related to recycling of Devonian carbonates, see Doucelance et al., 2010). Moreover, the role of the plume in the genesis of carbonatites is debated. The melting of the plume gives rise to the carbonatite melt in the case of a deep origin. Its role is mostly limited to its thermal expression in the case of a lithospheric origin. Finally, it was also proposed that carbonatites from the Cape Verde and Canary Islands share a common source with those of Tamazert in Morocco (Bouabdellah et al., 2010).

In this study we present the first cerium isotope ratios, together with Nd and Pb measurements, of calcio-carbonatites from the Cape Verde and Canary archipelagos and from the Tamazert massif in Morocco.  $^{138}\text{La}$  decays to stable  $^{138}\text{Ce}$  (138 Lanthanum;  $\beta$ -decay with a half-life of 292.5 Ga; Tanimizu, 2000) and  $^{138}\text{Ba}$  (138 Barium; electron capture with a half-life of 156.1 Ga; Sato and Hirose, 1981). Coupling the La/Ce and Sm/Nd systems may help to distinguish between the two proposed origins for the carbon of oceanic carbonatites: recycled carbon vs. primordial carbon. Marine carbonates show rare-earth-element (REE) patterns distinct from those assumed or measured for mantle and mantle-derived reservoirs so that old recycled carbonates and mantle-related materials are likely to display very contrasted  $\varepsilon\text{Ce}$  and  $\varepsilon\text{Nd}$  values with time. We will show that the Ce–Nd isotope compositions of oceanic calcio-carbonatites are in agreement with the recycling of marine carbonates in the source of their parental magmas.

## 2. Previous studies of oceanic calcio-carbonatites

Hoernle et al. (2002) have presented the first, radiogenic and stable, multi-isotope study of oceanic carbonatites from both the Cape Verde and Canary archipelagos. For Cape Verde, they proposed that calcio-carbonatites derive from the melting of recycled, 1.6 Ga-old oceanic crust containing secondary calcitic carbonates. The model was developed to explain the Sr–Nd–Pb isotopic compositions measured in samples, but fails to reproduce the low  $^4\text{He}/^3\text{He}$  isotopic ratios (down to 46,700;  $R/R_a$  up to 15.5) recently documented by Mata et al. (2010) for samples from the same outcrops. Neither does the model explain the mantle-like  $\delta^{13}\text{C}$  values ( $-8.0\text{‰}$  to  $-4.2\text{‰}$ ) measured in the samples. This last observation, however, must be balanced as eclogites have  $\delta^{13}\text{C}$  ranging from  $-34\text{‰}$  to  $+4\text{‰}$  (Pearson et al., 1994) with an average value close to that observed in the calcio-carbonatites.

As an alternative, Mata et al. (2010) proposed that the source of Cape Verde carbonatites is deeply seated in the mantle, being located at the D'' boundary layer. Such a source is supposed to be enriched in primordial rare gases and carbon, thus explaining the measured He and C isotopic compositions. Unradiogenic Sr–Nd isotopic ratios and intermediate Pb are not, however, reproduced by such a scenario, as is also the case for trace element patterns (Doucelance et al., 2010).

Mourão et al. (2012) then proposed an intermediate model for calcio-carbonatites from Brava (the western island of the Cape Verde southern group) in which the carbonatites result from liquid immiscibility, being thus genetically related to silicate magmas from the same island. The model relies notably on the similarity of isotope signatures between the calcio-carbonatites and the silicate formations of Brava. The Sr–Nd–Pb–He isotopic compositions of silicate samples from the Cape Verde archipelago are usually interpreted as the contribution of a high- $\mu$  (HIMU)-type end-member, which represents an ancient recycled oceanic crust, with lower mantle material (Doucelance et al., 2003; Holm et al., 2006; Millet et al., 2008; Mourão et al., 2012). Again, such a source can explain the Sr–Nd–Pb–He isotope compositions of carbonatites, but maybe not their  $\delta^{13}\text{C}$ . Furthermore, the model cannot be extended to other islands of the archipelago such as Fogo or Santiago, since silicate samples show Sr–Nd–Pb isotope compositions significantly

distinct from those of carbonatites. In the same way, De Ignacio et al. (2012) proposed that the calcio-carbonatites from São Vicente (Northern Islands) represent fractionation products of the parental magma of nephelinites, whose outcrops are spatially related to those of carbonatites. Although close, the nephelinites and calcio-carbonatites still present significantly distinct Sr–Nd–Pb values, which undermines the model.

Lastly, it has also been proposed that oceanic calcio-carbonatites are related to the recycling of marine carbonates at shallow mantle depth. The latter are the origin of metasomatized oceanic lithosphere, Cape Verde plume-induced partial melting giving rise to carbonatite parental magmas. A small contamination of carbonatitic melts by Cape Verde plume material explains both Sr–Nd–Pb and He isotopic compositions of carbonatites. The model would suggest heavier  $\delta^{13}\text{C}$  ratios than are actually measured, but is in agreement with the  $^{187}\text{Os}/^{188}\text{Os}$  isotopic ratios of carbonatites that present more radiogenic values than mantle end-members (Escrig et al., 2005), as is also the case for the model by Hoernle et al. (2002). Although radiogenic Os ratios could also result from minor seawater contamination during their emplacement, Re/Os whole-rock isochrones give a consistent age of  $3.2 \pm 1.2$  Ma for carbonatites.

Data measured in carbonatite samples from the Canaries are less numerous. Hoernle et al. (2002) have proposed that the calcio-carbonatites from the basal complex of Fuerteventura Island also result from melting of secondary calcitic carbonate in recycled oceanic crust, similar to the Cape Verde calcio-carbonatites. The model relies on the similar geochemical characteristics of calcio-carbonatites from both archipelagos. On the other hand, De Ignacio et al. (2006) have proposed that the calcio-carbonatites derive from a heterogeneous mantle plume with a FOZO-HIMU mixing-like component. Both models would explain HIMU-like initial radiogenic isotopic compositions measured in Fuerteventura samples, with uniform  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{143}\text{Nd}/^{144}\text{Nd}$  and relatively large range in Pb isotopic compositions. Turning to the Tamazert massif in Morocco, calcio-carbonatite samples with low  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios have mantle-type O and C isotopic compositions (Bouabdellah et al., 2010). Their source was interpreted to contain HIMU- and EMI-type components, as for the Cape Verde and Canary Island case (De Ignacio et al., 2012; Hoernle et al., 2002; Mourão et al., 2012). Thus, given the close similarity in carbonatite isotopic composition between the three localities, Bouabdellah et al. (2010) concluded that there was a common source for these rocks.

Two distinct types of models can thus be distinguished from the previous studies: 1) models involving primordial carbon in the oceanic carbonatite source vs. 2) models invoking marine carbonates recycled via subduction. Within the carbonate recycling models, two possible depths of recycling have been considered: the base of the oceanic lithosphere and the lower mantle.

## 3. Analytical procedures

100–200 g of each sample were reduced into chips in a jaw crusher before being powdered in an agate swing mill. Fine crushing was carried out in three rounds: 1) the first crushing run was conducted to pre-contaminate the agate mill; 2) the second run was kept for potential major element determinations; and 3) the third one for trace elements and Ce–Nd–Pb isotope analyses.

Trace-element ratios and cerium, neodymium and lead isotope separations and measurements were carried out at the Laboratoire Magmas et Volcans. 100 mg of sample powders were first acid-digested using HCl. Then, after removing of the supernatant, residues of centrifugation were dissolved with an HF–HNO<sub>3</sub> mixture. Sample solutions were then separated in three aliquots for Ce–Nd, Pb and trace element analyses, respectively.

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