Petrology and geochemistry of Peri-Mediterranean carbonatite magmatism: case studies from Fuerteventura (Canary Islands) and Mt. Vulture Volcano (Southern Italy)

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DOI:10.19276/plinius.2023.01.003

INTRODUCTION AND AIM OF THE WORK

Carbonatites are rare magmatic rocks but of great importance for several reasons, among which: (i) scientific importance, given their petrogenesis that involves modified carbonated mantle, liquid immiscibility, geodynamic setting (e.g., Yaxley et al., 2022); (ii) economic relevance, given that they are considered the main natural hosts of critical metals for key economic sectors in heavily industrialized countries, due to their great abundance in Rare Earth Elements (REEs) and trace elements (such as Nb, Hf, U, Th, etc), essential in many high-tech applications (e.g., Anenburg et al., 2021); (iii) environmental, due their strict ties with CO_2 uptake and exsolution. Despite their importance, many aspects of carbonatites petrogenesis and evolution processes remain still poorly constrained. One of the principal purposes of the scientific community is to refine the existing models for carbonatite magma petrogenesis, because they provide meaningful information on the deep geochemical cycle of carbon and mantle metasomatism. Then, it is fundamental to understand not only the genesis of carbonatite magmatism investigating its source, but also any possible post-genetic process during the evolutionary history of carbonatite melt.

This work adopts a multidisciplinary approach to define petrology and isotope geochemistry of carbonatites in different geodynamic settings. The goal is to characterize the source of carbonatite magmatism and identify any post-genetic processes such as crustal contamination, mixing processes, isotopic exchanges, hydrothermal mineralization, that were able to modify the primary signature and the origin composition of carbonatite melts. Fuerteventura (Canary Islands) was taken representative of oceanic carbonatites, while Mt. Vulture (southern Italy) was taken representative of intra-continental carbonatites, within the particular geodynamic context of Apennine subduction zone (Fig. 1). The comparison of oceanic carbonatites with intra-continental ones was aimed to provide information on the mantle source and the relationship of carbonatite magmatism with the various geodynamic settings, trying to understand why oceanic carbonatites are so rare if compared with continental counterpart. Principal inferences regard also the role of the carbonatite melts in metasomatiz-

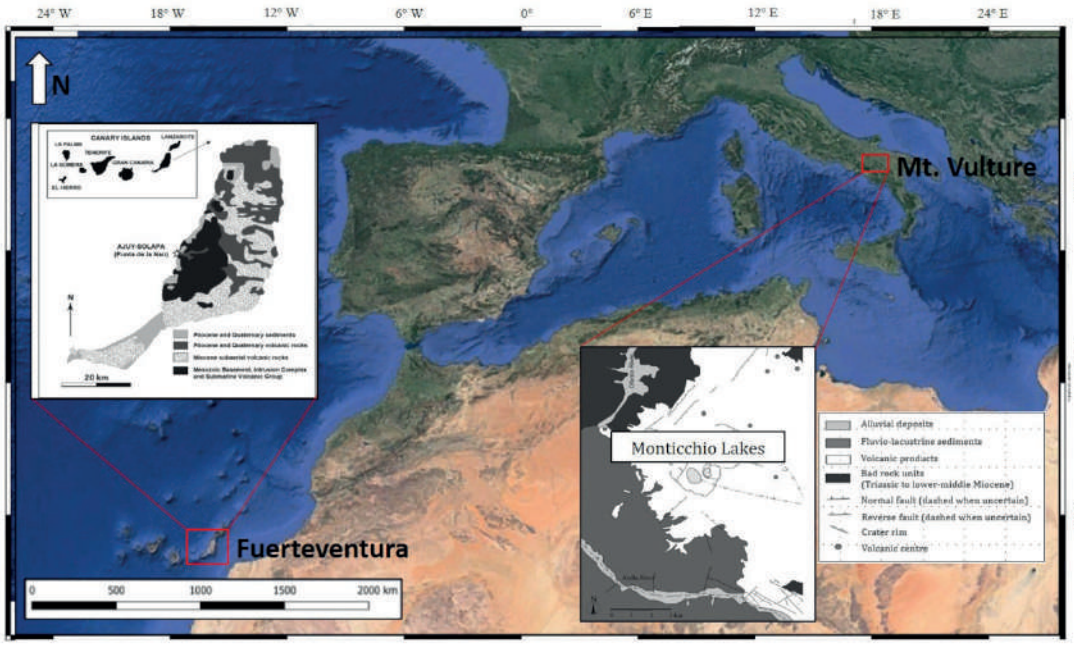

Figure 1 *Geographic location of the two studied areas.*

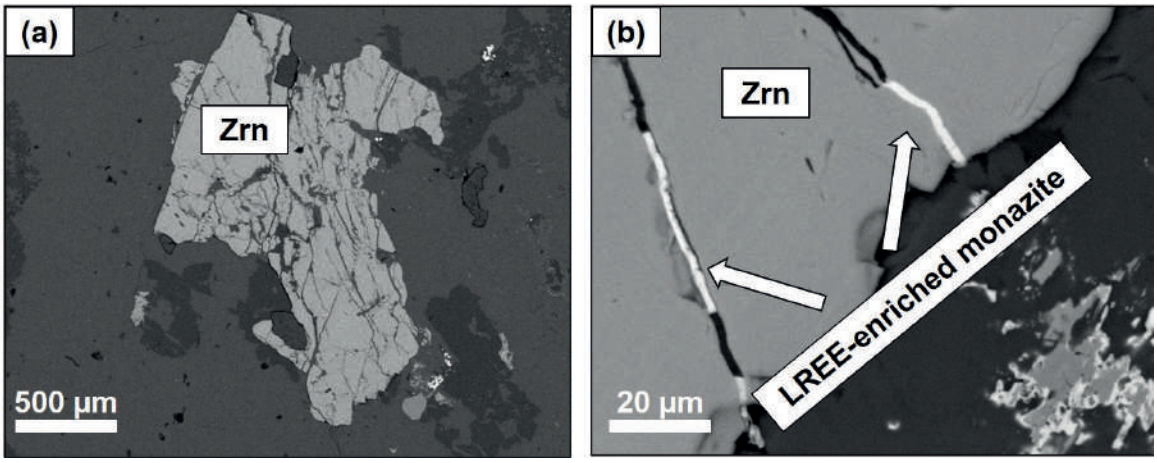

Figure 2 Backscattered electron images (BSE) of **(a)** zircon (Zrn) crystal and **(b)** filled microfractures in zircon crystal with monazite enriched in LREE (La, Ce, Nd).

ing the mantle source, and also on the importance of post-magmatic processes in which the infiltrating fluids play an important role in the transport and concentration of REEs.

To achieve these objectives, for each case study, a detailed minero-petrographic and minero-chemical characterization was firstly carried out. In particular, a detailed micro-thermometric study was performed in Fuerteventura carbonatites to understand the processes involved in carbonatite-related hydrothermal mineralization and formation condition (temperature, pressure and composition of fluid inclusions), and in Mt. Vulture products to provide important information about the melilitite-carbonatite magma ascent path and its mantle source, describing the volcano plumbing system and the way in which these very particular magmas are transported to the surface, with considerations about carbonatite mantle metasomatism and possible implications in terms of volcanic hazard. Subsequently, a characterization of noble gases (He, Ne and Ar), together with literature data of stable isotopes ($\delta^{13}C$ and $\delta^{18}O$), and Sr-Nd-Pb systematics, applied on studied carbonatite products (or closely related to them), was implemented.

RESULTS AND DISCUSSION

The case study of Fuerteventura (Canary Islands)

Hydrothermal fluids and formation conditions of REE enrichment processes

Carbonatitic dykes that crop out in the central western part of Fuerteventura Basal Complex were affected by the Miocene intrusive event responsible for contact metamorphism and partial melting of part of the alkaline-carbonatitic complex (Casillas et al., 2011). Indeed, the original igneous mineralogy disappeared completely in the anatectic zone, and a metamorphic association composed of wollastonite, monticellite, diopside, vesuvianite, garnet, calcite, perovskite, alabandite, pyrrhotite and Nb–Zr–Ca silicates (cuspidine–niocalite–baghdadite series) was formed (Casillas et al., 2011). The absence of metamorphic minerals typical of the contact zone, together with the presence of primary magmatic minerals such as apatite, biotite, and aegirine-augite, indicate that carbonatite outcrops in Punta de La Nao (Ajuy-Solapa sector) escaped this thermal overprint and preserve their original igneous assemblage and textures (Carnevale et al., 2020). Furthermore, the presence of monazite enriched in LREE (La, Ce, Nd) formation processes in microfractures that affect zircon crystals (Fig. 2), suggests carbonatite interactions with hydrothermal fluids and partial REE remobilization processes. Indeed, at sub-solidus temperatures (T > 600°C) and the brine-melt stage (600°C ≤ T ≤ 400°C), where REEs are sufficiently concentrated in the residual brine-melt to form REE-minerals, the infiltrating fluids during the hydrothermal stage play an important role in the transport and concentration of REEs. The identification of chlorite (d_{001}=14 Å), vermiculite (d_{001}=16,6 Å) and interstratified illite-smectite (d_{001}=13 Å) as hydrothermal alteration minerals, allows to frame the hydrothermal event in the metamorphic facies of zeolites, and the formation of these hydrothermal alteration minerals is due to a metasomatism process caused by the exsolution of a hydrothermal fluid phase from the magma that gave rise to carbonatite in the last moments of cooling (i.e. autometasomatism, a common process in carbonatitic intrusive rocks) (Carnevale et al., 2020).

In order to further constrain the formation conditions and characterize the late-stage hydrothermal fluids, the study of fluid inclusions (FIs) in carbonatite rocks is essential, and micro-thermometric studies represent the most straightforward method to obtain information about fluid temperature, pressure and composition (e.g., Carnevale et al., 2022a). Fuerteventura carbonatites show the presence of isolated FIs in host calcite minerals, suggesting a primary origin. The size of the inclusions is < 10 μm, and they mostly are in the range of 1-5 μm. Their shape is generally irregular, with negative-crystal forms and square shapes, according to the host calcite mineral. Few rounded isolated FIs in apatite are also present. Some FIs show textural characteristics of partial decrepitation, such as haloes of tiny fluid inclusions (diameter < 0.1 μm) surrounding the inclusion cavity, revealing that some events of partial density re-equilibration have occurred. Some intragranular trails confined within cal-

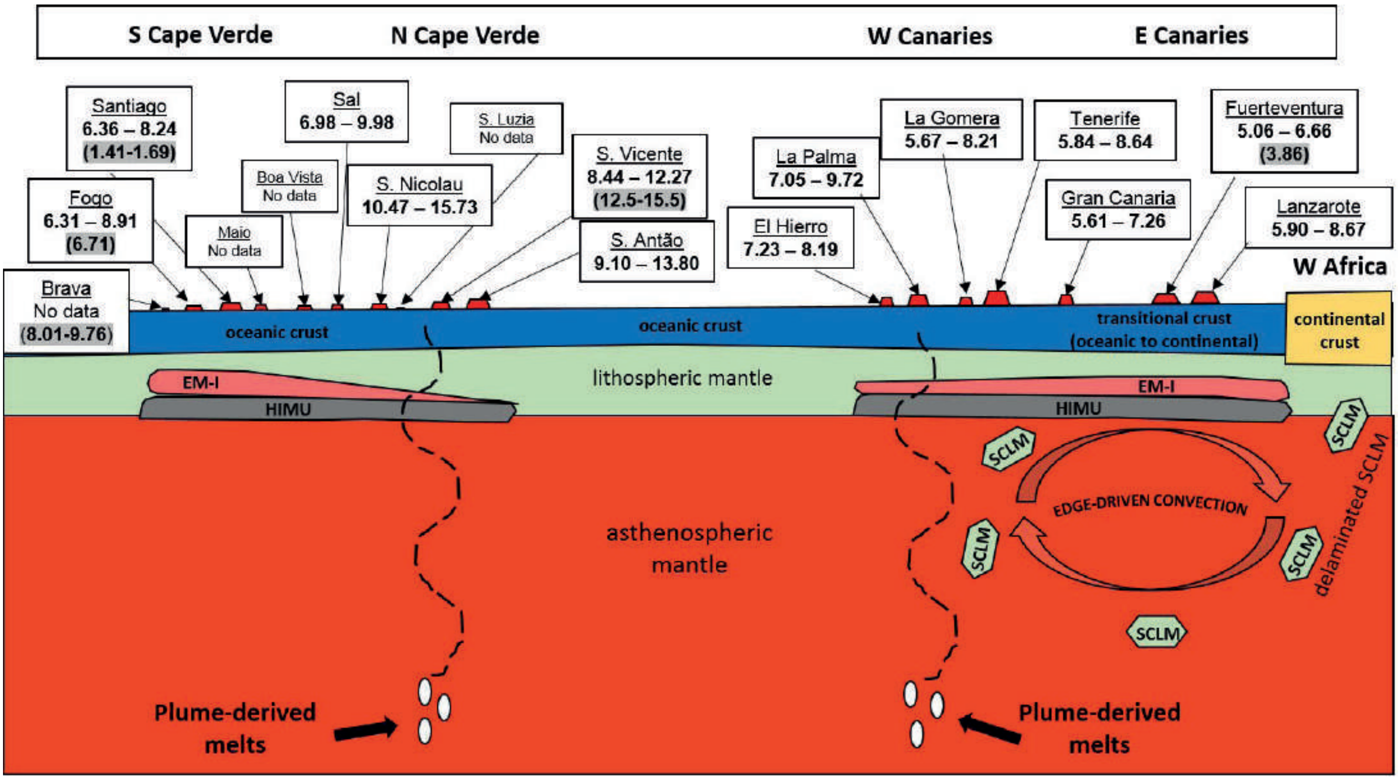

Figure 3 *Hypothetical schematic profile view of the upper mantle/crust under Cape Verde and Canary Archipelago (not in scale) with the involvement of the proposed mantle components and the mechanism of physical detachment of the sub-continental lithospheric mantle (SCLM) caused by edge-driven convection during breakup of the Pangea Supercontinent and incorporated into the oceanic lithospheric mantle beneath at least some Canary Islands.*

cite grain-boundary and crystal interior are also found. Observing the temperatures of phase changes in FIs during heating/cooling analyses, it is possible to know the composition of the inclusions for the appropriate chemical system, because each chemical system has its own eutectic temperature (T_e). Results show apparent eutectic temperatures ranging from -21.7 to -21.0°C, although the small dimension of FIs did not allow the correct determination of T_{e*} for almost all inclusions. Thus, the eutectic behavior and the narrow range of measured T_e suggest the well know binary NaCl-H_2O system (T_e = -21.2°C) as a characterizing chemical system in studied FIs. No CO_2 was detected (the eutectic temperature of pure CO_2 is -56.6°C). With the T_e range and the chemical system, the final ice melting temperatures (T_{im}) during heating was also measured ranging between -3.2 and -2.1°C and corresponding to salinities of 5.3 and 3.6 wt% $NaCl_{eq}$, respectively. All studied fluid inclusions homogenize to liquid phase (ThL; L + V = L), with homogenization temperature ranging from 142.7 to 208.7°C (corresponding to densities of 0.95 and 0.88 g/cm^3, respectively) and homogenization pressures ranging from 4.6 to 15.3 bars. To resume, micro-thermometric studies on Fuerteventura carbonatites shows the presence of low salinity (≅ 5 wt% $NaCl_{eq}$) and low temperatures (< 300°C) fluids trapped within primary calcite crystals, corroborating that processes of fluid/rock interaction occurred. Thus, REEs can be mobilised and concentrated on the local scale by low-temperature hydrothermal fluids with a process known as autometasomatism, accordingly with the more recent classification proposed by Mitchell & Gittings (2022) using the term carbo-hydrothermalite, where a carbonatite can precipitate at sub-solidus temperatures from a mixed CO_2-H_2O fluid during late-stage processes.

$^3He/^4He$ isotopic ratios

Even though calcite $^3He/^4He$ ratios on Fuerteventura carbonatites (up to 3.86Ra) are a little higher than clinopyroxenes (≈ 2.23Ra) and higher than apatites (up to 0.08Ra), they present values significantly lower than typical MORB mantle values (R/Ra = 8 ± 1), while clinopyroxenes from clinopyroxenite show $^3He/^4He$ ratios (≈ 6.66Ra) slightly lower than typical MORB mantle values and in the range of sub-continental lithospheric mantle values (SCLM, 6.1 Ra ± 0.9). The lower values on mineral separates from Fuerteventura carbonatites might be caused by "magma aging" (i.e. internal production of radiogenic $^4He^*$ from U and Th content that lower the original R/Ra ratio of the magmatic source). However, considering (i) ^3He and ^4He concentration, (ii) Th and U content measured on calcite and clinopyroxene separates and (iii) contribution of radiogenic $^4He^*$, using a simple magma aging model, initial R/Ra ratios of calcite and clinopyroxene from carbonatite and clinopyroxene from clinopyroxenite are not significantly modified. Indeed, initial R/Ra ratios continue to present values lower than typical MORB mantle values (≈5.10Ra and ≈5.24Ra for calcite and clinopyroxene from carbonatite and ≈6.82Ra for clinopyroxene from clinopyroxenite), and they are in the range of SCLM. An age of 53.5 Ma would be required

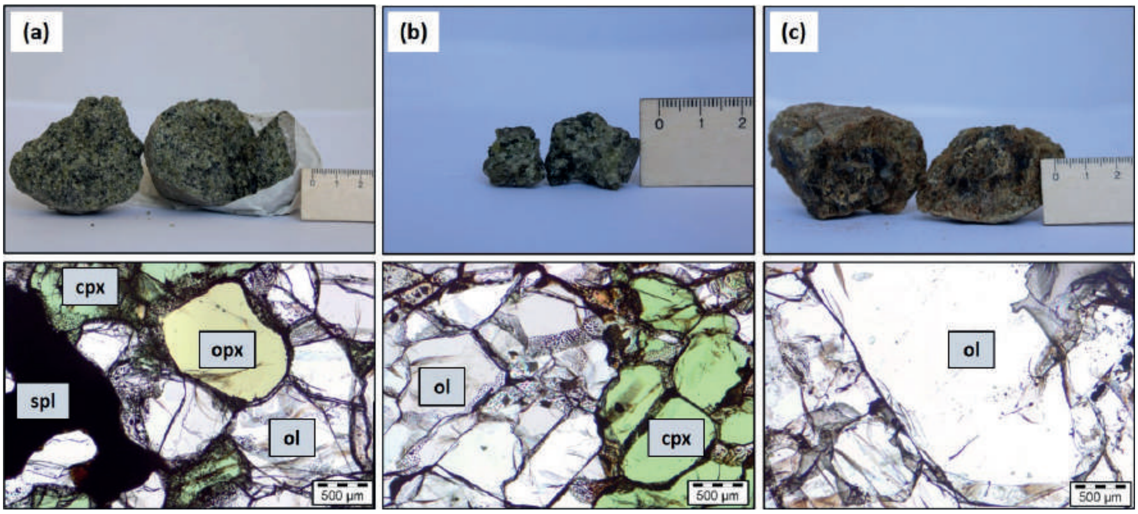

Figure 4 *Mantle xenoliths hand specimens and related parallel polars (PPL) photomicrographs of (a) fine-grained lherzolite, (b) medium-grained wehrlite, and (c) coarse-grained dunite, respectively. ol = olivine; opx = orthopyroxene; cpx = clinopyroxene (emerald green Cr-diopside); spl = spinel.*

for Fuerteventura carbonatites to bring the initial R/Ra ratio of 8 on calcite, and this is not realistic as have ages close to 25 Ma (Muñoz et al., 2005). We therefore consider the He isotopic ratio from the calcite to be reflective of a sub-continental lithospheric mantle signature.

Figure 3 shows a summary of the proposed mantle components for Canary and Cape Verde primitive magmas and shows also a clear regional distribution and an isotopic dichotomy of ^{3}He/^{4}He ratios in both silicate rocks and Cape Verde carbonatites. This regional distribution appears to be slight on Canary Islands, with an apparent increase of R/Ra from eastern (oldest) to western (youngest) Islands, and all islands except for Fuerteventura, present a MORB-like values. Interestingly, both Fuerteventura carbonatites and silicate rocks, present R/Ra values lower to slightly lower than typical MORB mantle values, and they are in the range of SCLM (Carnevale et al., 2021).

The case study of Mt. Vulture (southern Italy)

Carbonatites and xenoliths: a look into the mantle

The carbonatite magmatism from Mt. Vulture is essentially related to a phreatomagmatic activity of the last melilitite-carbonatite eruption (141 ± 11 ka, Villa & Buettner, 2009). The presence of ash-rich tuff deposits with a melilitite-carbonatite matrix and carbonatite-rich layers within the matrix, reflects the direct evidence of carbonatite magmatism. Furthermore, a robust amount of pelletal lapilli (enclosing mantle xenoliths) and mantle xenoliths from the melilitite-carbonatite matrix, represents a great tool to understand the composition and possible modification of a mantle source influenced by carbonatite metasomatic fluids. Mantle xenoliths are rounded, of an average 4-5 cm in diameter, with some rarely samples up to 10 cm. The grain size of the mantle xenoliths is variable, fine- to medium-grained (0.5-1 mm) in the case of the lherzolite and wehrlite, medium- to coarse-grained (2-5 mm) in the case of the wehrlite, ol-clinopyroxenite and dunite (Fig. 4). The texture is usually equigranular and sub-idioblastic, with the exception of dunite xenolith that show an inequigranular texture. All xenoliths show a granoblastic-decussate structure, where the interlocking and randomly oriented grains are variably elongated. Well-defined triple junctions are common. Olivine usually shows undulose extinction and deformation structures such as kink bands, while orthopyroxene and clinopyroxene (emerald green Cr-diopside) occur as subhedral/anhedral with curvilinear boundaries. Spinel is also present as subhedral/anhedral, with colour varying from brown to dark brown. Noteworthy, is the presence in the xenoliths of phlogopite and reaction rim in orthopyroxene replaced by a very fine-grained assemblage of olivine and clinopyroxene microcrysts, suggesting that metasomatism processes occurred. Among the Mt. Vulture mantle products, the presence of wehrlite xenoliths is widely recognized and is corroborated by our findings where pelletal lapilli cores are largely wehrlitic. (La/Yb)N ratios (> 3–4), the presence of carbonates and apatites in some wehrlites, suggest carbonatite metasomatism and the role of carbonatite melts instead of silicate melts in metasomatizing the wehrlite xenoliths.

The chemical composition of olivine, orthopyroxene, and clinopyroxene minerals from the mantle xenoliths is similar to those from the ultramafic core of pelletal lapilli and from the loose xenocrysts within the melilitite-carbonatite matrix. Olivines show Mg-rich composition (Fo$_{87\text{-}92}$) and relatively high NiO (0.2 to 0.4 wt%), with some exception in dunite xenolith with low NiO content (0.08 wt%). Orthopyroxenes show also high values of Mg (Mg# = 0.89-0.90), and compositionally, the proportion of En (Wo$_{2\text{-}3}$, En$_{86\text{-}87}$, Fs$_{10\text{-}11}$) shows a positive correlation with the Fo content of the coexisting olivine. Clinopyroxenes are essentially diopside in composition (Wo$_{47\text{-}51}$, En$_{44\text{-}48}$, Fs$_{5\text{-}7}$) with some Na-rich and Ca-poor exceptions, and they generally show high Cr_2O_3 (0.6-1.4 wt%) and high Mg# values (0.87 to 0.91).

Pelletal lapilli are considered the juvenile component of the last Mt. Vulture volcano activity, representing the interface between the erupting silica undersaturated melilititic-carbonatitic magma and the volatile component, this latter considered very important in the evolu-

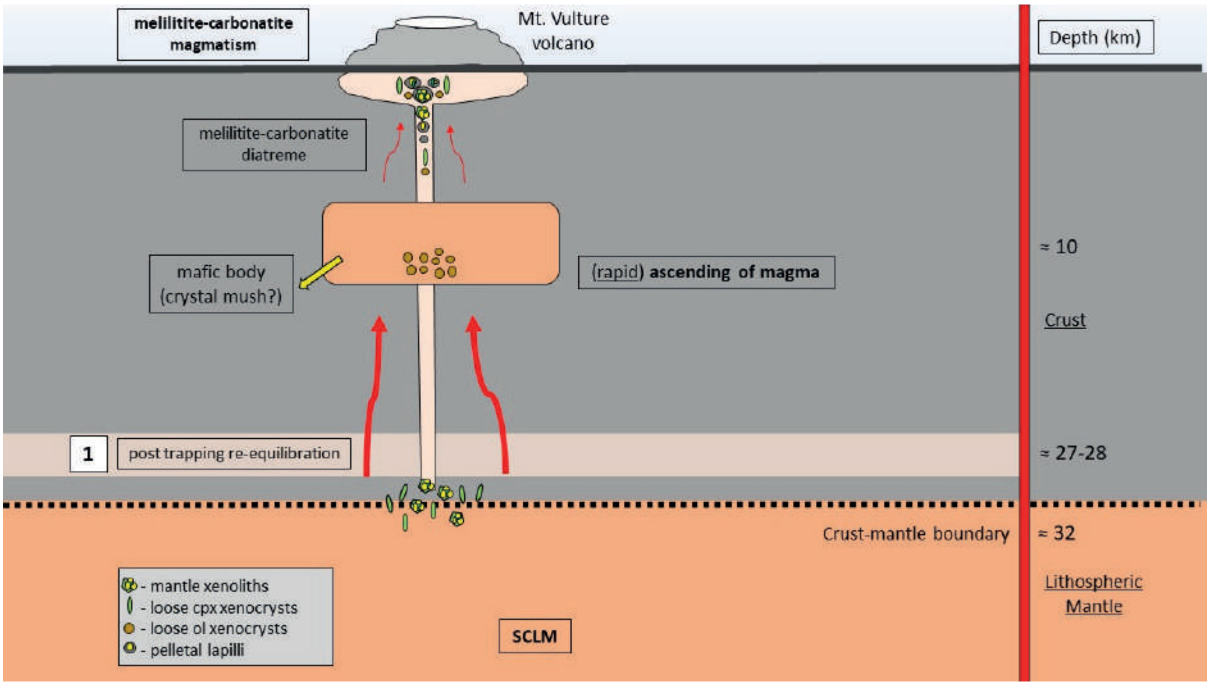

Figure 5 *Simplified cross section of Vulture volcano ponding stages.*

tionary history of the erupting magma (Lloyd & Stoppa, 2003). Thus, the whole rock chemistry of the external portion of Mt. Vulture pelletal lapilli represents a good tracer to distinguish the literature data of sedimentary calcrete-carbonate portion from the melilitite-carbonatite one. Indeed, the external portion of pelletal lapilli shows concentrations (e.g., Ba = 2430 ppm; Sr = 1250 ppm; ∑REE = 527 ppm) comparable with those of the average values of Ca-carbonatite and plotting onto the average range of extrusive carbonatites, suggesting a possible contribution of a carbonatite melt in the trace elements enrichment processes.

The study of chemical composition together with trapping temperature and pressure of fluid inclusions trapped in rock-forming minerals of mantle xenoliths provides insights into the PT history of the mantle and the mineral-melt-fluid interaction processes in the mantle and within the volcano magma storage system. Fluid inclusions are present in all studied samples, in both ultramafic cores of pelletal lapilli and in mantle xenoliths. The histograms of densities of FIs in mantle xenoliths show a similar modal distribution if compared with those of mantle xenolith cores of pelletal lapilli (Carnevale et al., 2022b). FIs trapped in lherzolite and wehrlite xenoliths registered magma ponding at different depths within the Mt. Vulture magma system (Fig. 5). The first ponding stage corresponds to the local crust-mantle boundary (at about 32 km depth), while the second corresponds to a shallow ponding stage (at about 12-14 km depth), as corroborated by geophysical evidences of solidified magmatic body at the same depth below the Vulture volcano (Improta et al., 2014). In order to constrain the ascent velocity of the melilitite-carbonatite magma, we used the equation from Lister & Kerr (1991). Our result of the ascent rate (≈ 20 m/s) of the melilitite-carbonatite magma is in the same order of the ascent rates of kimberlite magmatism. If we consider also recent studies showing how volcanic systems where activity has remained dormant for protracted periods (> 100 ka) still have the potential for reactivation (e.g., Giordano & Caricchi, 2022), and in Mt. Vulture there is a possible link between the development of tear faults, magmatism and related magma ascent along these tectonic pathways (Peccerillo, 2017), our study highlight that the volcanological community should pose great attention to volcanic hazard in melilitite-carbonatite volcanoes, and it should be carefully evaluated even after long time of quiescence.

Oceanic carbonatites: petrogenetic model

Carbonatite magmatism is essentially related to intraplate continental tectonic settings, with only two occurrences in oceanic tectonic setting: Cape Verde and Canary (Fuerteventura) Archipelago. Several studies were proposed trying to explain why oceanic carbonatites are so rare and are not found in all HIMU-OIB-like archipelagos (e.g., Doucelance et al., 2010), but currently there is no petrogenetic model that can constrain the issue in detail. Although it is a simple model, here is presented a carbonatite petrogenetic model in oceanic environment (Fig. 6) based essentially on the most important geochemical and experimental petrology studies and on the main results of this thesis. Since the pioneering studies of experimental petrology about carbonatite petrogenesis it is clear that all petrogenetic processes are related to the formation of a primary carbonate melt derived from a carbonated mantle, with processes of metasomatism with carbonatitic melts, notwithstanding the oceanic or intracontinental context. Hammouda & Keshav (2015) show how the region located in the depth range of 90 to 150 km deep is where carbonatitic melts can most likely

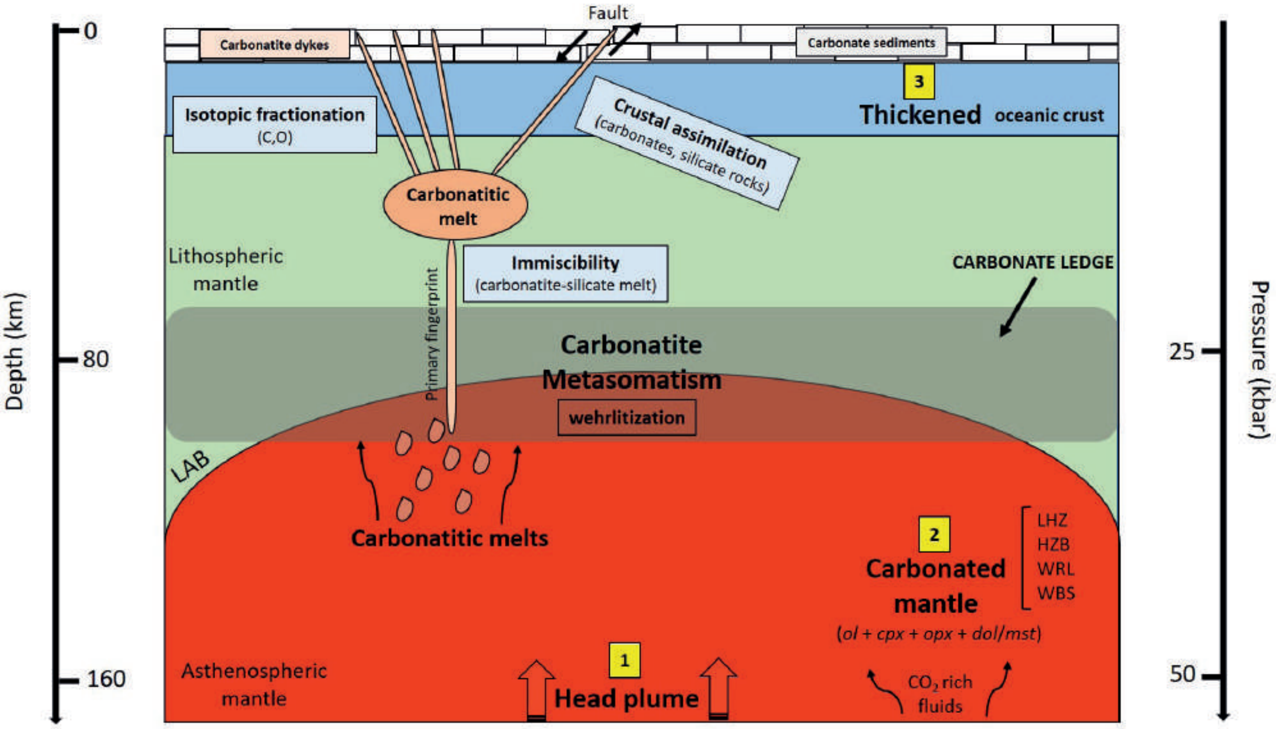

Figure 6 *Proposed simple petrogenetic model for oceanic carbonatites with the PluMeTh condition.*

be produced and impregnate the surrounding mantle through metasomatism, and the upper bound of this region is called the carbonate ledge. This latter represents the shallowest depth at which carbonate melt is stable before degassing, and in the carbonated peridotite system is located between 20 and 30 kbar (Hammouda & Keshav, 2015). Therefore, the presence of the carbonate ledge prevents carbonatite magmas to reach the surface, and only very fast ascent rates can prevent primary carbonatites from degassing by reaction at the ledge through the ascent to the surface. However, Hammouda et al. (2014) experimentally demonstrated that in hot thermal environments the ledge could be avoided, reacting with an harzburgitic mantle and becoming more calcic upon ascent. Moreover, according to them, Schmidt & Weidendorfer (2018) proposed that oceanic carbonatites develop in hotspots where primitive silicate melts have the lowest SiO$_2$ and highest total alkalis, characteristics that originate from deep and low-degree melting, and this would be responsible for more abundant carbonatites in continental hotspots, where the lithosphere is a priori thicker than below oceanic crust. Thus, in oceanic tectonic settings it is fundamental the thickness of the lithosphere, and this is perfectly in agreement with the thickness of the lithosphere under Fuerteventura and Cape Verde Archipelago, with an anomalously high transitional oceanic to continental crust. To resume, according also with Doucelance et al. (2010), here it is proposed a petrogenetic model that can be described as the "PluMeTh" condition: 1) there must be a mantle plume to preserve the required hot environment and acting as a trigger for low-degree partial melting at the base of the oceanic lithosphere; 2) processes of mantle Metasomatism by carbonatitic melts in a carbonated mantle with the involvement of a SCLM are needed; 3) the oceanic lithosphere must be Thick in order to favor plume-induced partial melting at the base, with the production of primitive silicate melts with the lowest SiO$_2$ and highest total alkalis and the subsequent processes that drive evolving melts into the carbonatite–silicate miscibility gap.

CONCLUSIONS

The multidisciplinary approach of this thesis on petrology and geochemistry of carbonatites permitted characterize the source of carbonatite magmatism in different geodynamic settings, identifying the role of the infiltrating fluids in the transport and concentration of Rare Earth Elements (REEs), and the role of the carbonatite primary melts in metasomatizing the mantle source. To resume,

(i) regarding the case study of Fuerteventura:
   (1) isotope geochemistry (noble gases) gives an important contribution to the oceanic carbonatite petrogenesis;
   (2) combining our results with the most important geochemical and experimental petrology studies a general petrogenetic model for oceanic carbonatites is proposed;
   (3) petrography, mineral and whole-rock chemistry show how the low temperature fluids play an important role in the transport and REEs concentration processes in carbonatites after the magmatic stage, with direct implications of economic interest.

(ii) regarding the case study of Mt. Vulture:
   (1) fluid inclusions micro-thermometry shows the presence of CO$_2$-rich fluids in mantle xenoliths, and two

different magma storage at different depth in Mt. Vulture volcanic system;

(2) modelling magma ascent rate results in quite high velocity (≈ 20 m/s) for melilitite-carbonatite magmas;

(3) detailed petrographic studies show evidences of wehrlitization processes as a consequence of carbonatite metasomatism in lithospheric mantle;

(4) the evidences of carbonatite-like metasomatism and mantle degassing, and the presence of mantle xenoliths in volcanic systems related to diatreme explosive eruptions with long quiescence period and high velocity magma ascent rate, are considered important in the evaluation of volcanic hazard.

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