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New Inferences on Magma Dynamics in Melilitite-Carbonatite Volcanoes: The Case Study of Mt. Vulture (Southern Italy)

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Abstract This study provides the first micro-thermometric data of fluid inclusions (FIs) in mafic loose (disaggregated) xenocrysts and ultramafic xenoliths in explosive products of the melilitite-carbonatite Mt. Vulture volcano (southern Italy). Pure CO_2 late stage FIs hosted in rock-forming minerals of wehrlite xenoliths and clinopyroxene xenocrysts were trapped at the local crust-mantle boundary (32 km). In contrast, trapping pressures within the loose olivine xenocrysts are from 3.2 to 4.5 kbar (8–13 km). Considering the ongoing degassing of mantle-derived CO_2 rich gases, together with seismic evidences of the presence of low amount of melts at depth, and the tectonic control of the past volcanic activity, our study opens new perspective about the hazardous nature of the "quiescent" melilitite-carbonatite volcanoes.

Plain Language Summary The study of fluid inclusions (FIs) (small amount of fluid trapped within minerals) provides important information on variable environments and magmatological processes in which the host minerals were formed. Investigation of the FIs with respect to their composition, trapping pressure and temperature, allow us to constrain magma ascent history. To understand the last explosive volcanic activity of Mt. Vulture volcano (southern Italy), we investigated FIs in mafic minerals and mantle fragments brought to the surface by a melilitite-carbonatite magma. Our results show the presence of CO_2-rich FIs with trapping pressure corresponding to a depth of 32 km in mantle fragments, and a shallower depth (8–13 km) in mafic mineral. Estimates on magma ascent rate show rapid ascent dynamics to the surface. Our study emphasizes the importance of a multidisciplinary approach that combine geochemistry and petrology to investigate a volcanic system even if the volcano is considered "quiescent," as is the case of Mt. Vulture volcano, where mantle degassing is still ongoing.

Key Points:

- Micro-thermometric analyses show the occurrence of high-density CO_2-rich fluid inclusions hosted by minerals within wehrlite xenoliths
- Ascent rate between melilitite-carbonatite (≈20 m/s) and kimberlite (≈45 m/s) magma is comparable
- Melilitite-carbonatite volcanoes can be hazardous even after long time of quiescence ($>10^5$ years)

Supporting Information:

Supporting Information may be found in the online version of this article.

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1. Introduction

Carbonatite magmatism is mainly associated with intraplate continental tectonic settings characterized by significant extension and even rifting, with a temporal distribution from Archean to the present (e.g., Jones et al., 2013; Woolley & Kjarsgaard, 2008; Yaxley et al., 2022), and currently, Oldoinyo Lengai (Tanzania) represents the only active carbonatite volcano, characterized by a natrocarbonatitic affinity (e.g., Berkesi et al., 2020). The growing number of carbonatite occurrences from unconventional tectonic settings, such as oceanic contexts (e.g., Carnevale et al., 2021; Day, 2022; Doucelance et al., 2010; Mata et al., 2010; Schmidt & Weidendorfer, 2018) or subduction zones (e.g., D'Orazio et al., 2007; Li et al., 2018; Lustrino et al., 2019, 2020), received considerable attention during last two decades, given their importance as source of rare elements such as La, Ce, Pr, and Nd (Anenburg et al., 2021; Verplanck et al., 2016), and, most importantly, because they provide meaningful information on the geochemical cycle of carbon and mantle metasomatism as well (e.g., Bouabdellah et al., 2010; Horton, 2021).

Mt. Vulture (southern Italy) is an isolated volcano located between the Apulia foreland and the eastern side of the Apennine orogenic belt, in correspondence of the geodynamic context of the Apennine subduction zone (D'Orazio et al., 2007; Peccerillo, 2017). This volcano is located along the deep NE-SW lithospheric faults that represent a local vertical tear of the slab (e.g., Rosenbaum et al., 2008), a potential pathway for the ascent of melts (Caracausi, Martelli, et al., 2013; D'Orazio et al., 2007).

The Vulture volcano is a small volcanic complex, with several eccentric eruptive vents, covering an area of approximately 70 km². Its eruptive activity started about 739 ± 12 ka (Villa & Buettner, 2009) and it continued

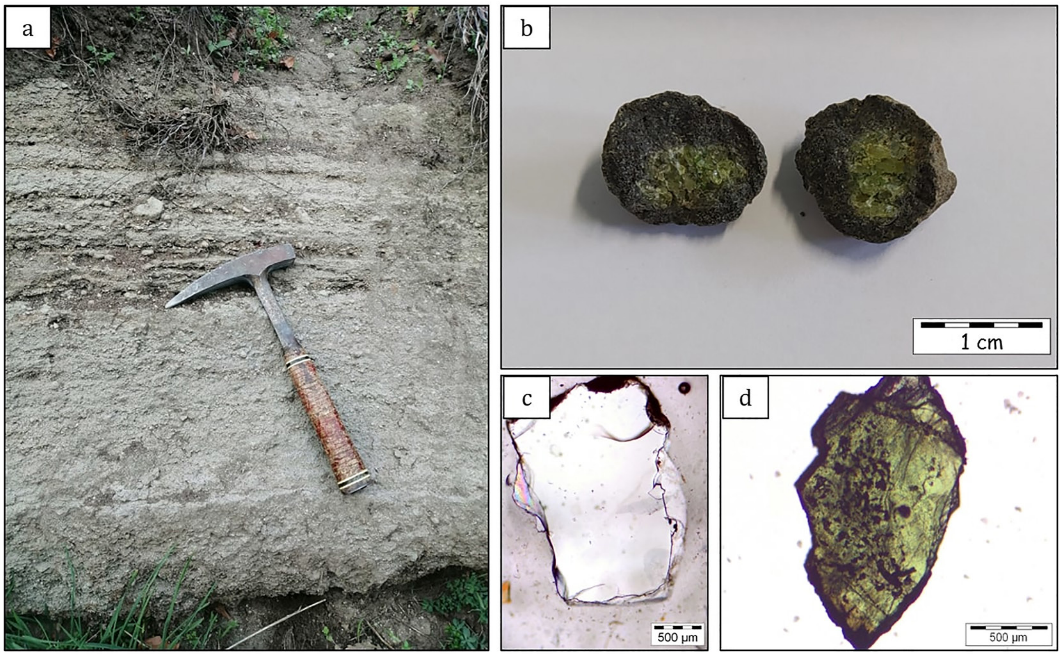

Figure 1. Photomicrographs of the sampling site together with pelletal lapilli and loose crystals. (a) Ash-rich tuff surge deposit of Lago Piccolo Subsynthem. (b) Pelletal lapilli with ultramafic xenolith cores. (c) Olivine xenocryst from the fine-grained matrix (parallel polars). (d) Clinopyroxene (Cr-diopside) xenocryst from the fine-grained matrix (parallel polars).

until to 141 ± 11 ka (Villa & Buettner, 2009), with long inter-eruptive quiescence (>10^5 years, Buettner et al., 2006). The last volcanic event was a maar-forming eruption (Stoppa & Principe, 1997). Water of the two resulting crater lakes (Monticchio Lakes) dissolves CO_2-rich mantle-derived volatiles (Caracausi, Nicolosi, et al., 2013; Caracausi et al., 2009; Paternoster et al., 2016), supporting the active degassing at this volcano (Caracausi et al., 2009, 2015). The last volcanic activity (identified as Monticchio Lakes Formation, Stoppa & Principe, 1997), fed by a melilitite-carbonatite magma, brought to the surface some pelletal lapilli (enclosing abundant ultramafic mantle xenoliths and xenocrysts) considered to be juvenile component, because they represent the interface between the erupting magma and the volatile component (Lloyd & Stoppa, 2003). These products are particularly useful to characterize the mantle source beneath Vulture volcano, providing important information about the melilitite-carbonatite magma ascent path and its mantle source.

To this aim, micro-thermometric data of fluid inclusions (FIs), hosted in the ultramafic xenolith cores of pelletal lapilli and in loose olivine and clinopyroxene xenocrysts, have been used together with mineral chemistry in order to describe the way in which these very particular magmas are transported to the surface and the possible implications in terms of volcanic hazard.

2. Sample Description

Samples were collected from the Lago Piccolo Subsynthem (Giannandrea et al., 2006) (Figure S1 in Supporting Information S1). Twenty-nine pelletal lapilli were sampled from a compact fine-grained carbonate-dominated matrix in an ash-tuff phreatomagmatic deposit. The ultramafic xenoliths (dominantly wehrlitic in modal composition) constitute the core of pelletal lapilli and are surrounded by a 3–10 mm thick rim of micro-phenocrysts (Figures 1a and 1b). We also selected approximately 200 olivine and 100 clinopyroxene (Cr-diopside) xenocrysts (Figures 1c and 1d) from the fine-grained carbonate-rich matrix, where xenocrysts of blackish clinopyroxene, amphibole, mica (phlogopite) and spinel, were also present. To compare the FIs within the xenocrysts with those trapped in the ultramafic xenolith cores of pelletal lapilli, we selected two wehrlite cores, three olivine and two clinopyroxene xenocrysts. We analyzed 171 FIs in olivine xenocrysts, 107 in clinopyroxene xenocrysts, and 184 FIs in the ultramafic cores of studied lapilli, all being <10 µm in size and most of them in the range of 1–5 µm.

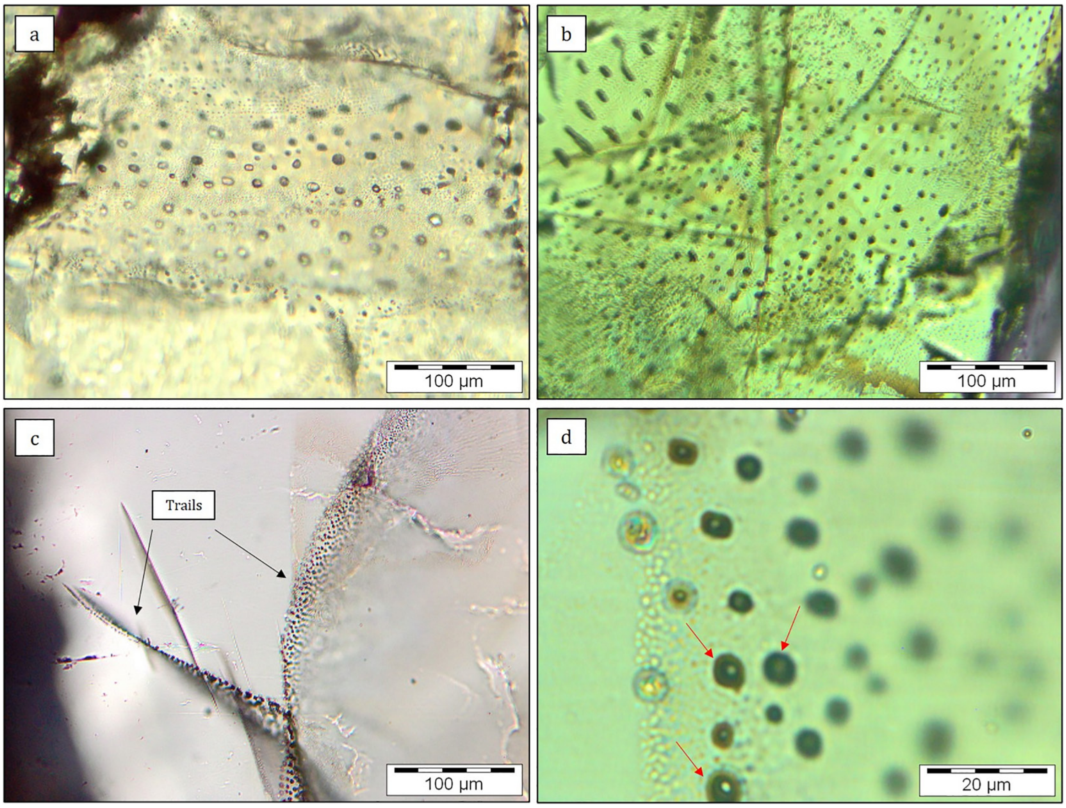

Figure 2. Photomicrographs (parallel polars) of fluid inclusions (FIs) and their textural position. FIs enclosed by (a) olivine and (b) clinopyroxene xenocrysts. (c) Intragranular trails (black arrows) of FIs in olivine xenocryst. (d) FIs with decrepitation features (red arrows) in clinopyroxene from the ultramafic core of a pelletal lapillus.

3. Results: Petrography, Mineral Chemistry, and Fluid Inclusions

The ultramafic xenolith cores of pelletal lapilli (the diameter of enclaves vary from 6 to 17 mm) are characterized by the presence of Mg-rich olivine (Fo_{90-91}, NiO varying from 0.35 to 0.38 wt. %, Table S1 in Supporting Information S1) and diopside (Wo_{46-48}, En_{47-48}, and Fs_{4-5}) with relatively high Cr_2O_3 content (1.3–1.5 wt. %, Table S2 in Supporting Information S1). The Mg# values of olivine and clinopyroxene in the ultramafic xenolith cores are uniform (0.90–0.92). The grain size of the ultramafic xenolith cores is fine-to medium-grained (300–600 µm) with granoblastic texture, interlocking with randomly oriented olivine and elongated clinopyroxene (Figure S2 in Supporting Information S1). The thick rim of fine-grained material surrounding the xenoliths, is composed essentially of häuyne micro-phenocrysts, with xenocrystic debris of olivine and clinopyroxene (Figure S3 in Supporting Information S1).

Olivine xenocrysts show very similar composition ($Fo_{89–92}$, NiO = 0.37–0.41 wt. %, Table S1 in Supporting Information S1) compared to olivine from the ultramafic xenolith cores of pelletal lapilli. Similarly, almost all clinopyroxene xenocrysts show akin composition ($Wo_{46–48}$, $En_{47–48}$, $Fs_{4–6}$, Cr_2O_3 = 0.4–1.3 wt. %) with respect to clinopyroxene from the ultramafic xenolith cores of pelletal lapilli (Table S2 in Supporting Information S1). The Mg# values in olivine and clinopyroxene xenocrysts are also uniform (0.89–0.92) (Figure S4 in Supporting Information S1).

In all studied samples FIs are usually rounded and slightly stretched (Figures 2a–2d), and some of them form trails of variable length (0.1–1 mm), lined in sealed fractures (Figure 2c). Re-equilibration features in FIs are present (i.e., stretching and/or decrepitation process), as evidenced by the occurring of an outer dark halo around the FIs (Figure 2d). In the xenocrysts, secondary FIs (distinguished on the basis of their textural characteristics and distribution within the crystals, such as the presence of trails in sealed fractures) are more abundant than primary FIs and tend to be smaller than the primary ones. On the contrary, in olivine and clinopyroxene in the ultramafic cores of pelletal lapilli, early stage FIs are more abundant than late stage FIs. The studied FIs are

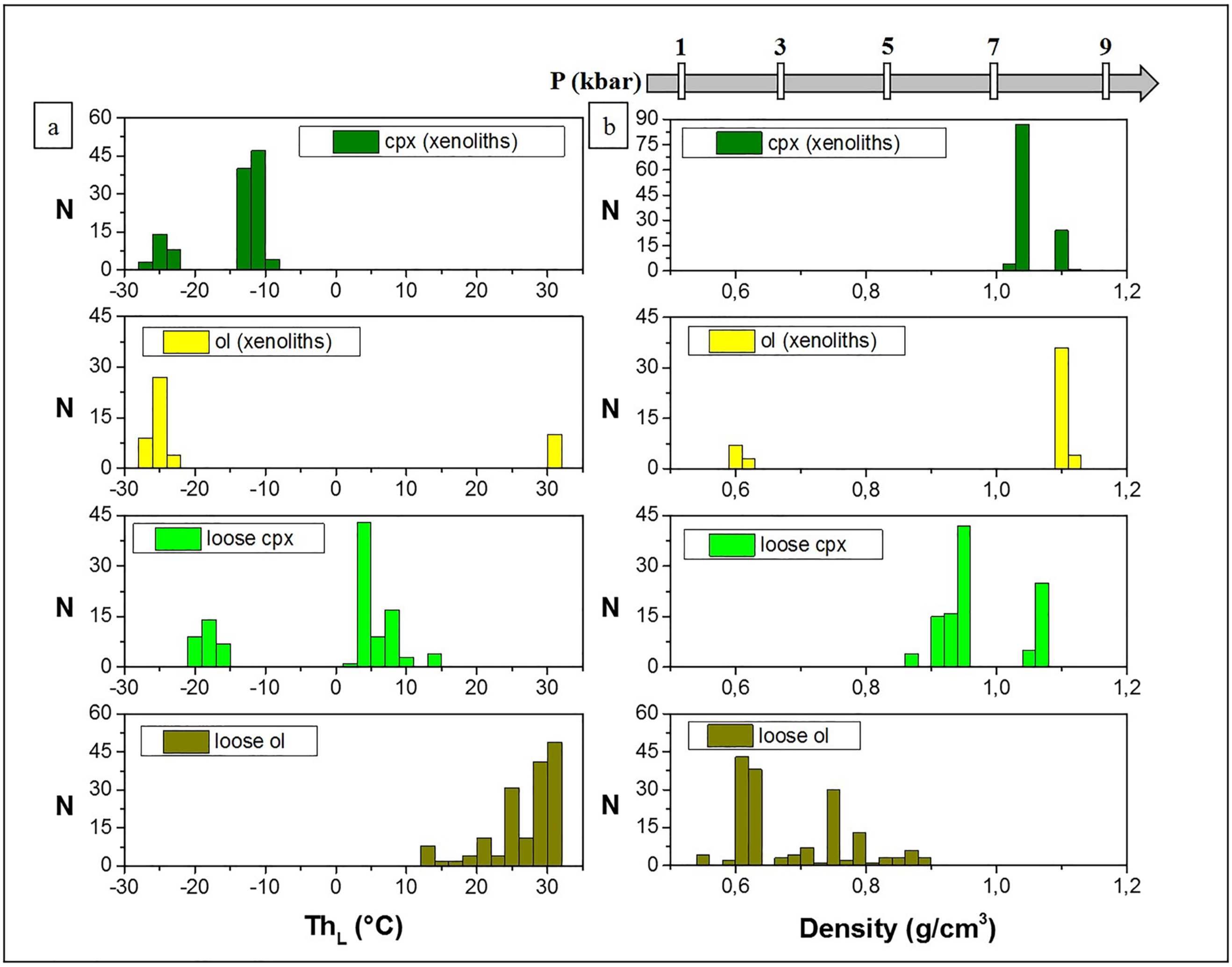

Figure 3. Frequency distribution of (a) homogenization temperatures (Th$_L$) and (b) densities of fluid inclusions hosted in olivine and clinopyroxene xenocrysts and in ultramafic cores of pelletal lapilli from Mt. Vulture. The number of the total measurements (*N*) is reported to the left of each graph.

characterized by pure CO_2, with melting temperatures (T_m) ranging in a very narrow interval between −56.6 (i.e., the triple point of pure CO_2 at 1 bar) and −56.8°C ± 0.1°C. All FIs homogenized to a liquid phase with temperatures of homogenization (Th$_L$) ranging from 11.5°C to 30.2°C and from −20.0°C to 13.2°C, respectively for olivine and clinopyroxene xenocrysts, and corresponding to density range (ρ) of 0.58–0.85 and 0.84–1.03 g/cm^3. In FIs hosted in ultramafic xenoliths, Th$_L$ range from −27.3°C to −8.5°C (ρ = 0.98–1.06 g/cm^3) in clinopyroxene crystals, and from −27.7°C to −6.0°C (ρ = 0.96–1.07 g/cm^3) in olivine crystals. Values of Th$_L$, densities, corrected densities and number of measures are reported in Table S3 in Supporting Information S1. Further details, also about analytical methods, are reported in Supporting Information S1.

4. Significance of Fluid Inclusions Data

Histograms of homogenization temperatures (Figure 3a) and densities (Figure 3b) show polymodal skewed distributions. These distributions are due to fluid trapping episodes and re-equilibration that occur at different depth of the volcanic system. The highest corrected density values of FIs are in the ultramafic xenoliths (1.10–1.11 g/cm^3), corresponding to minimum fluid pressure between 8.5 and 9.0 kbar (≈27–28 km). In the clinopyroxene xenocrysts FIs recorded fluid pressure of 8.2–8.7 (≈26–27 km) and 6.7–7.2 kbar (≈21–22 km). FIs in olivine xenocrysts show fluid pressures of 2.8–3.1 (≈8–9 km) and 4.1–4.5 kbar (≈12–13 km). The low-density peak (ρ = 0.63 g/cm^3) is also present in ultramafic xenoliths. Trapping pressures and densities were estimated at the equilibrium temperature of 1100°C, that is, an intermediate value of the temperature in the range 1050°C–1150°C previously inferred by Jones et al. (2000) on pyroxenes from mantle xenoliths.

All FIs show stretching and, most importantly, partial decrepitation process (Figure 2d), which is evidence of volumetric re-equilibration at high strain rates associated with a short-time scale event (e.g., Bodnar, 2003), and this is supported also by the skewed distribution of histograms. The pristine density of trapped fluid was therefore lowered during crustal ascent. Magma ascending through the lithosphere halted at important discontinuities marked by changes in chemical and physical properties of country rocks (Menand, 2011), the most important being the crust-mantle boundary. This is the case of the common depth registered by FIs in ultramafic xenoliths and clinopyroxene xenocrysts. Olivine composition (Fo% > 0.90, NiO > 0.35 wt.%), clinopyroxene Cr content ($Cr_2O_3 > 1.3$ wt. %) and spinel Cr# (Cr/Cr + Al) (>0.38), strongly suggest that the studied wehrlitic cores are of mantle origin and are not cumulates produced by fractional crystallization in shallow level magma ponding stages (Beccaluva et al., 2002), although cumulates can be also formed by underplating in the vicinity of Moho (e.g., Kovács et al., 2004). Furthermore, the xenoliths have no cumulative or poikilitic textures, they lack plagioclase that often occurs in cumulates, and the chemical composition overlaps well with other xenoliths from previous studies (Figure S4 in Supporting Information S1). Coherently with the crust-mantle boundary inferred beneath the Vulture area with geophysical methods (magnetism and gravimetry) at a depth of about 32 km (Kelemework et al., 2021), the re-equilibration processes lowered by about 15% the fluid density at the time of trapping. Thus, the xenoliths probably represent the shallowest upper mantle, and the FIs in the rock-forming minerals of these xenoliths suggest a minimum trapping pressure of 9–10 kbar, which is around the local Moho. FIs in loose clinopyroxene xenocrysts suggest a minimum trapping pressure of 7–9 kbar. Therefore, if we consider these clinopyroxenes as fragments of the xenoliths (as also witnessed by their similar chemical composition) it is likely that they crystallized nearby the Moho. The crystallization of loose olivine xenocrysts may have taken place at shallower, crustal depths, with FIs suggesting a minimum trapping pressure of 3–4 kbar.

It is worthy of note that the shallowest trapping event in olivine xenocrysts occurring at depth of 8–13 km overlaps the depth (6–15 km) of a mafic body, probably a dense crystal mush, within the Vulture volcano magma system. Petrological investigation located a shallow magma reservoir down to 6 km (Beccaluva et al., 2002). To resume, the micro-thermometric data here presented and obtained from the crystal content from a single eruption, show a very good agreement with both geophysical and petrological data for this volcano. Figure 4 shows a simplified schematic profile view of Vulture volcano with our considerations.

5. Carbonatite Metasomatism and Magma Ascent Dynamics

The study of mantle xenoliths represents a great tool to understand the composition and possible modification of a mantle source influenced by metasomatic fluids. In this framework, the increase of modal clinopyroxene at the expense of orthopyroxene has been interpreted as a result of the interaction between ultramafic material and carbonate melts, and carbonatite metasomatism is accompanied by the formation of secondary clinopyroxene formed during the reaction of carbonatite melts with orthopyroxene (Dalton & Wood, 1993; Russell et al., 2012). Although interaction between peridotite wall rock and alkaline mafic melts normally lead to clinopyroxene enrichment in the mantle, with the consequent formation of wehrlites (e.g., Patkó et al., 2020), in our case study, the process of "*wehrlitization*" in the lithospheric mantle is primary due to carbonate melts instead of mafic silicate melts.

Among the Mt. Vulture mantle products, the presence of wehrlite xenoliths is widely recognized (e.g., Beccaluva et al., 2002; Downes et al., 2002; Jones et al., 2000) and is corroborated by our findings where pelletal lapilli cores are largely wehrlitic. According to Zong and Liu (2018), specific crystallochemical patterns in clinopyroxenes (e.g., Mg# vs. Ca/Al; and Ca/Al vs. $^{87}Sr/^{86}Sr$) fall into the mantle-related carbonate metasomatism field (Figure S5 in Supporting Information S1), and $(La/Yb)_N$ ratios (>3–4), further suggest carbonatite metasomatism (Coltorti et al., 1999). Furthermore, the presence of carbonates and apatites in some wehrlites of the Mt. Vulture (Downes et al., 2002; Jones et al., 2000), reinforce the role of carbonatite melts instead of silicate melts in metasomatizing the wehrlite xenoliths. Rosatelli et al. (2007) also propose carbonatite melts as the main metasomatism agent of Mt. Vulture mantle source region, emphasizing the role of silicate-carbonatite magma immiscibility during the magma evolution at shallower depths (Solovova et al., 2005), supported by a number of experimental constrains underlying melilititic magma (the last erupted at Vulture volcano) as the best candidate to exsolve an immiscible carbonatite melt (Brooker & Kjarsgaard, 2011). Further evidence of metasomatism by carbonatite melts is given

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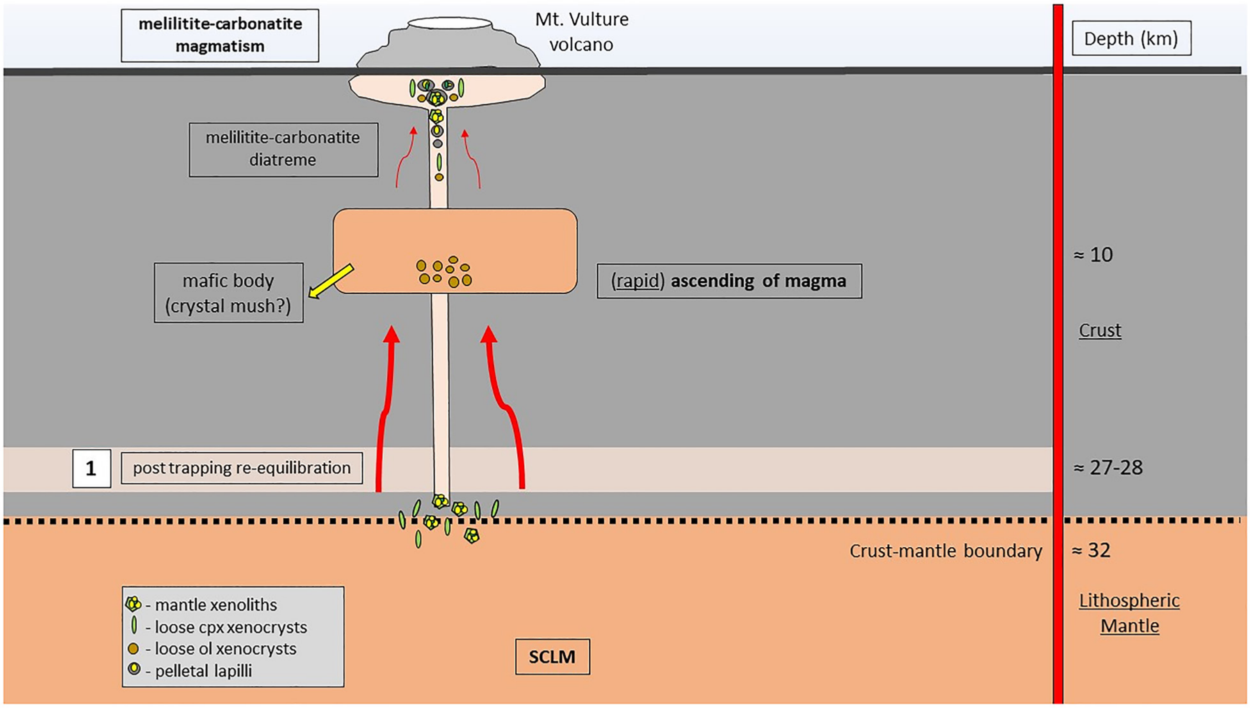

Figure 4. Simplified cross section of Vulture volcano ponding stages. Olivine and clinopyroxene of wehrlite cores of pelletal lapilli, together with clinopyroxene xenocrysts register the same fluid trapping event at the local crust-mantle boundary. Olivine xenocrysts show a shallower signature of fluid entrapment, overlapping the depth of the mafic body within the Mt. Vulture magma system (Improta et al., 2014). The involvement of a carbon-rich subcontinental lithospheric mantle (SCLM) is from Bragagni et al. (2022).

by the presence of interstitial calcite associated with Fe-Ni-sulphides between olivine grains in a mantle xenolith from Mt. Vulture (Blanks et al., 2020).

Despite the last eruptive event of Mt. Vulture dates back to 141 ± 11 ka (Villa & Buettner, 2009), geochemical evidences support that active degassing of mantle-derived volatiles is still ongoing in Mt. Vulture area (Caracausi, Martelli, et al., 2013; Caracausi et al., 2009), showing how the relationship between the deep CO_2 release and the time of its last eruption could be an important tool for evaluating the state of current activity (Caracausi et al., 2015). Moreover, recent studies show how the source of CO_2 degassing in Mt. Vulture area is related to the presence of a subcontinental lithospheric mantle (SCLM), that sequesters large amounts of CO_2 due to the infiltration of fluids and melts during carbonatite-like metasomatism (Bragagni et al., 2022). In this scenario the He isotopic signature in FIs of the Vulture mantle xenoliths (<6.1Ra; Ra is the He isotopic signature in air) overlap the range of the SCLM He end member (6.1 ± 0.9; Gautheron & Moreira, 2002).

Considering, (a) the degassing of mantle-derived fluids in Mt. Vulture area (Caracausi, Martelli, et al., 2013; Caracausi, Nicolosi, et al., 2013; Caracausi et al., 2009, 2015), (b) the explosive behavior associated with a maar-diatreme system of the Monticchio Lakes Synthem (Solovova et al., 2005; Stoppa & Principe, 1997), (c) the occurrence of small amounts of magma at the crust-mantle boundary depth (<1.6%, Tumanian et al., 2012), in absence of mantle upwelling or extensional tectonics that could favor decompression melting (Peccerillo & Frezzotti, 2015), (d) the role of tectonics in the transfer of the mantle-derived magma and volatiles and its control of the Vulture volcanism and outgassing (e.g., Caracausi, Martelli, et al., 2013; D'Orazio et al., 2007; Rosenbaum et al., 2008), (e) the long inter-eruptive periods (>140 ka, Buettner et al., 2006), and (f) the recognized occurring of volatiles rich magmas at the crust-mantle boundary (Section 4, Significance of Fluid Inclusions Data), we computed by using a simplified model the possible melilitite-carbonatite magma ascent rate to figure out fast versus slow uprise of these magmas from the crust-mantle boundary, furnishing new elements to the knowledge of Mt. Vulture activity. In order to constrain the ascent velocity of the melilitite-carbonatite magma, we used the equation from Lister and Kerr (1991) and applied by Sparks et al. (2006) in their physical model, with the same approach also proposed by Moussallam et al. (2016).

Taking into consideration (a) a closed system during the magma ascent with a constant dike width of 1 m, (b) a magma density of 2,500 kg/m^3, (c) a constant viscosity of 0.6 Pa s, and (d) a mean density of the crust of 2,600 kg/m^3, we obtain ascent rate of about 17 m/s (Equation 8 from Sparks et al. (2006)), assuming that the buoyancy is the main driving force. As there are no previous works that can help to fix the dike width in our case study, we assumed the unity (1 m) as a conservative dimension value, with the awareness of the non-linear correlation between the dike width and magma ascent rate, and the effect of different variables on dike propagation (e.g., uneven stress distribution within the crust). Magma viscosity value is taken from experimental studies of a representative melilitite synthetic melt (Stagno et al., 2020). Magma density is calculated using the model of Ochs and Lange (1999) at 1,100°C and 10 kbar, assuming a bulk composition from Stoppa and Principe (1997) with $SiO_2 = 37$ wt. %, and a mean CO_2 value of 7.5 wt. %, obtained from the H_2O-CO_2 solubility model proposed by Moussallam et al. (2016). Indeed, if we consider their model for a low SiO_2- and H_2O-free melts (our FIs study indicates the presence of pure CO_2 as the main volatile phase), at about 30 km depth, we obtain bulk CO_2 concentration between 5 and 10 wt. %. The model of Moussallam et al. (2016) is applied to a kimberlite magmatism (and to basalt magmatism) with 25 wt. % $\leq SiO_2 \leq$ 32 wt. %, and it is comparable to the melilitite-carbonatite magmatism of Monticchio Lakes Syntheme with $SiO_2 <$ 40 wt. % (Stoppa & Principe, 1997).

Our result of the ascent rate of the melilitite-carbonatite magma is in the same order of the ascent rates of kimberlite magmatism (e.g., Kelley & Wartho, 2000; Moussallam et al., 2016) and more than two times faster if compared with ascent rate calculated from other volcanic complexes where CO_2-rich FIs in metasomatized upper mantle xenoliths occur (e.g., 5 m/s, Szabó & Bodnar, 1996). In our simplified modeling the melilitite-carbonatite magmas could reach the surface from the depth of 30 km in less than an hour, considering, however, a single fast event without taking into account possible ponding level at crustal depth. 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; Harangi et al., 2015; Molnár et al., 2018, 2019), 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; Rosenbaum et al., 2008), 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.

6. Conclusion

We analyzed FIs hosted in rock-forming minerals of the wehrlitic cores of pelletal lapilli and in xenocrysts of olivine and clinopyroxene brought to the surface by a melilitite-carbonatite magma from the last eruption of Vulture volcano (Monticchio Lakes Syntheme, Lago Piccolo Subsynthem). We found pure CO_2 FIs with different trapping pressures (from 3.2 to 10.3 kbar) that correspond to magma storage at different depths within the volcano plumbing system. The deepest ponding stage is represented by the crust-mantle boundary (at a 32 km depth), while the shallower corresponds to a solidified magmatic body (former crystal mush) imaged by geophysical investigations (Improta et al., 2014). Modeling magma ascent rate results in quite high velocity (≈20 m/s) for melilitite-carbonatite magma from the crust-mantle boundary to the surface, and it is comparable with ascent rate of kimberlite magmatism (e.g., ≈45 m/s, Moussallam et al., 2016). These evidences, coupled to (a) the outgassing of magmatic volatiles at Mt. Vulture, which isotopic signature correspond to those in the FIs of the last activity of the volcano (Caracausi, Martelli, et al., 2013; Caracausi et al., 2009), and to (b) the presence of small amounts of melt (<1.6%) at the crust-mantle boundary depth, add constraints for magma production and ascent pathways. Therefore, this study confirms that the scientific community must pay attention also to the inactive volcanoes, because they could be still hazardous systems notwithstanding the last volcanic activity occurred hundreds/thousands of years ago.

Data Availability Statement

The complete data set of chemical and micro-thermometric analyses of this study was uploaded to the Zenodo FAIR aligned repository (www.zenodo.org) and will be available for download at the required link: Carnevale et al. (2022).

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