MAGMA MIXING EXPERIMENTS: A NEW TOOL TO UNRAVEL THE TIMING OF VOLCANIC ERUPTIONS

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INTRODUCTION

The physical and chemical interactions between compositionally different magmas (*i.e.*, *magma mixing* in the broadest interpretation including both *mingling* and *mixing*, in agreement with Perugini & Poli, 2012) is considered a major petrogenetic process concurring in the generation of the wide compositional diversity of igneous rocks on Earth, in both plutonic and volcanic environments (*e.g.*, Anderson, 1976; Sparks *et al.*, 1977; Bateman, 1995; Perugini & Poli, 2012). The first definition referred to magma mixing as "the process whereby two or more compositionally distinct magmas are mixed together such as the melt of each is blended into a compositionally uniform single magma" (Anderson, 1976), and the most accepted general scenario consists of a magmatic storage region in the mid-upper crust containing an evolved magma (*e.g.*, dacite or rhyolite) that is intruded by a batch (or batches) of more primitive magma (*e.g.*, basalt) ascending from a deeper reservoir (*e.g.*, the lower crust or the upper mantle). The evidence of magma mixing processes remains recorded by a range of structural and textural features in igneous and volcanic rocks, including the occurrence thermo-chemical disequilibria in mineral phases (*e.g.*, Didier & Barbarin, 1991; Hibbard, 1981; Wallace & Bergantz, 2002), magmatic enclaves dispersed into compositionally different host rocks (Bacon, 1986; Blake & Fink, 2000), and bandings of different magma compositions (*e.g.*, Flinders & Clemens, 1996). Also, magma mixing is witnessed by extreme geochemical variations of major and trace elements and isotopes, which can occur in the rocks even at very short length scales, down to millimeters or micrometers (*e.g.*, Perugini & Poli, 2012; Wiesmaier *et al.*, 2015). Furthermore, magma mixing has been suggested as an effective eruption trigger (*e.g.*, Druitt *et al.*, 2012; Leonard *et al.*, 2002; Sparks *et al.*, 1977), and recently, the combination of experimental investigations and the study of volcanic rocks produced by the interaction between different magmas has been proposed to be used as a volcanic chronometer (Perugini *et al.*, 2015). The main idea behind the use of magma mixing as a volcanic chronometer assumes the injection of hotter, more primitive, and potentially volatile-rich magma into a more evolved magmatic reservoir as among the leading causes of mechanical and thermo-barometric destabilization of the system, possibly triggering the eruption (*e.g.*, Sparks *et al.*, 1977; Eichelberger, 1980; Laumonier *et al.*, 2014; Perugini *et al.*, 2015; Rossi *et al.*, 2019).

The observation of pyroclastic rocks and lava flows strongly supports the hypothesis that magma mixing is often involved in the volcanic plumbing system dynamics before an eruption. However, even if the occurrence of a direct link between the injection of new magmas in a shallow crust magmatic reservoir and a subsequent eruption has been already established, the timescales of the process remain unclear and deserve further investigation. An important point emerging from magma mixing studies is that the latter is a time dependent process: this means that the longer is the mixing time, the more homogeneous is the mixture. For this reason, recent studies have suggested, on the basis of high-temperature experiments performed with natural silicate melts, that the compositional variability caused by magma mixing might be used to define new geo-chronometers to estimate the mixing-to-eruption timescales (Perugini *et al.*, 2015). In order to derive empirical relationships to constrain eruption timescales, an efficient assessment of the reliability of magma mixing experiments in terms of robustness and reproducibility of the results is determinant, also due to the considerable difficulties arising from the study of such a complex process through experimental procedures conducted at high temperatures and with high viscosities melts.

Motivated by these considerations, in this work we performed different sets of magma mixing experiments using natural compositions aiming to develop a robust statistical framework for the construction of

mixing-to-eruption geo-chronometers, also evaluating the degree of reproducibility of the experiments, as well as the reliability of results. In particular, we performed experiments at different mixing times and at condition relevant for natural magmatic systems. The degree of reproducibility of experimental results has been tested repeating one experiment using the same starting conditions and comparing the compositional variability of major elements. This allowed us to evaluate the robustness of the empirical relationships relating the degree of homogeneity of the samples against the mixing time.

We then applied our experimental procedure and results to a selected case study showing clear evidence of magmatic interaction. In this light, significant evidence of magma mixing has been recognized in different volcanic products (both pyroclastic and effusive) derived from the eruptive activity of the Aeolian Islands volcanoes (Southern Tyrrhenian Sea, Italy), making this area of great interest to study the interaction between compositionally different magmas prior to the eruptions. As a first point, we made a comprehensive review of the evidence supporting the hypothesis of mixing-triggered eruptions in the entire Aeolian archipelago and then we moved our attention on the experimental investigation of the AD 1739 eruption at Vulcano Island, chosen as a case study because it represents one of the most active and dangerous volcanoes in the Aeolian region, posing the highest risk in the area (De Astis *et al*., 2013). For the selected eruptive event, we provided an estimate of the mixing-to-eruption time (*i.e.*, the time elapsing between the beginning of the mixing process in the magmatic reservoir and the subsequent eruption) by comparing the results of magma mixing experiments to natural samples. Also, we estimated the averaged magma ascent rates, and discussed the implications of the results for an improved definition of the pre-eruptive dynamics occurring in the shallow plumbing system below Vulcano Island.

In the end, to further constrain magma mixing dynamics, we experimentally evaluated the space and time evolution of the mixing process for a system characterized by the presence of a mafic crystal-bearing end-member. In fact, among others, the possible presence of crystals may greatly increase the complexity of the mixing system due to the occurrence of regrowth/resorption processes in minerals that can be induced by mixing dynamics (*e.g.*, Kouchi & Sunagawa, 1985; Laumonier *et al*., 2014). The latter may lead to a significant compositional heterogeneity of the magmatic mass, causing the formation of complex zoning patterns that reflect the intensity of chemical exchanges between the interacting magmas. Preliminary results indicate a considerable enhancement of the process due to the mechanical action of the crystals in the mixing flow field with increasing mixing time.

Despite its great complexity, that makes difficult to study the phenomenon taking into account all the variables that can affect it at the same time, our findings represent an advancement in the knowledge of the mixing process and may have significant implications in the context of volcano monitoring activities and civil protection purposes since the knowledge of the dynamics of the plumbing systems of active volcanoes and their monitoring is essential for the assessment of the risks associated with potential future eruptions.

MATERIALS AND METHODS

Volcanological setting of the Aeolian Islands and the AD 1739 and 1888-90 eruptions at Vulcano Island

The Aeolian Islands are a volcanic archipelago in the Southern Tyrrhenian Sea, north of the coast of Sicily (Fig. 1A). The erupted products range between 270 and 250 ka, with the oldest exposed on Lipari (ca. 270 ka), Salina (ca. 250 ka) and Filicudi (ca. 250 ka), respectively. Compositionally, they range from calc-alkaline, high-K calc-alkaline, to shoshonitic and alkaline (De Astis *et al*., 2013). The mixing process, together with fractional crystallization, magma–fluid interactions, and different degrees of host rock assimilation, has often been invoked as a recurring event in the petrological evolution of magmatic feeding systems throughout the Aeolian arc (*e.g.*, De Astis *et al*., 2013). At Vulcano Island (Fig. 1B), the volcanic activity started ca. 120 ka ago (Clocchiatti *et al*., 1994; De Astis *et al*., 2013) and the last eruptions occurred in AD 1739 and 1888–90. The AD 1739 eruption was characterized by a first explosive phase with the production of fallout deposits, banded pumices and minor pyroclastic density currents (*e.g.,* PDCs), and a second effusive phase with the emplacement

of the Pietre Cotte lava flow. The 1888-90 eruption was characterized by a predominantly explosive style producing pyroclastic and fallout deposits, trachytic to latitic in composition. Magma mixing has been then investigated for the AD 1739 eruption, whose products show evidence of magmatic interaction revealed by reaction rims in the different phases, reversely zoned clinopyroxenes, banded pyroclastic products, and the presence of magmatic enclaves (latitic in composition) dispersed within a rhyolitic lava flow (*e.g.*, Pietre Cotte outcrop).

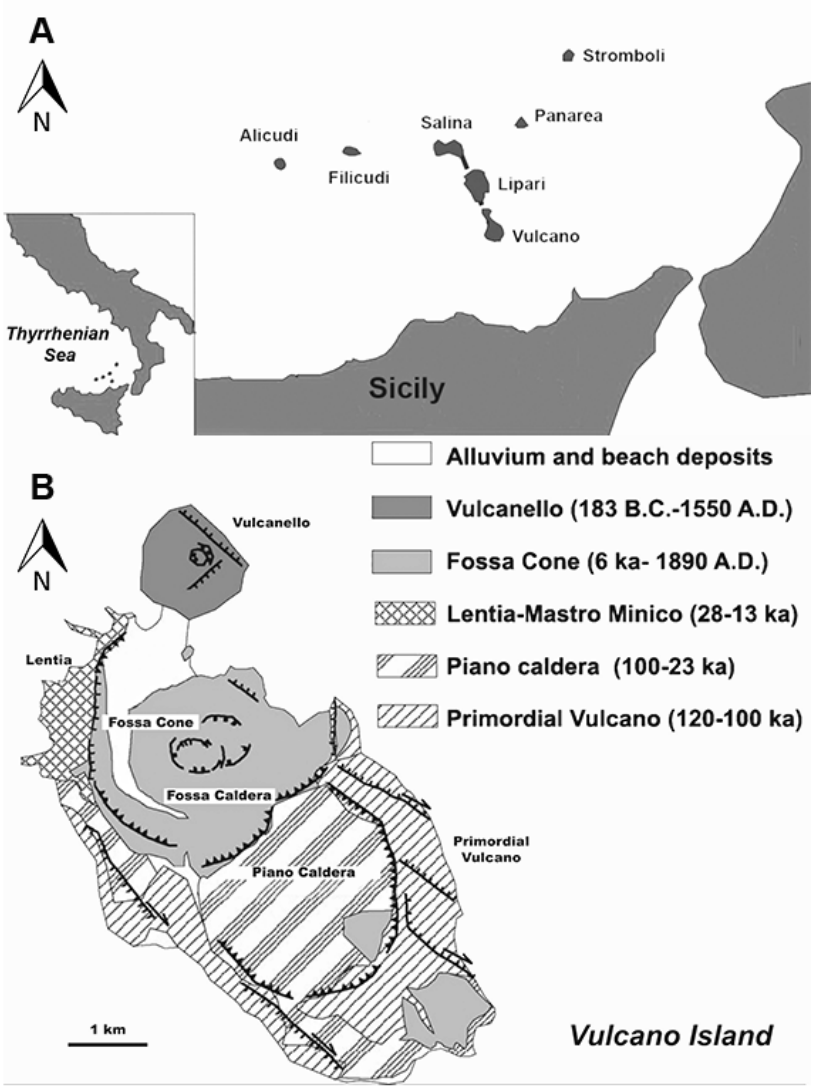

Fig. 1. - A) Schematic map of the Aeolian Islands and B) schematic geological map of Vulcano Island (modified from Peccerillo *et al*., 2006) showing the main volcanic units and the location of the Pietre Cotte lava flow generated during the effusive phase of the AD 1739 eruption.

Sample selection for the experimental procedure

Three rock samples have been used as end-members in the experiments. A shoshonite sampled at the Vulcanello lava platform and a latite from the top part of La Fossa cone have been used as mafic end-members in the first and second set of mixing experiments, respectively. The most evolved composition, used in both series of experiments, is a high-K rhyolitic obsidian from the Pietre Cotte lava flow, belonging to La Fossa cone. The chemical compositions of the three end-members are listed in Table 1. Further details on sample preparation procedure for mixing experiments is reported in Morgavi *et al.* (2015) and Rossi *et al.* (2017).

Banded pumices, utilized to compare the experimental results to natural samples, derive from the fallout events of the explosive phase of the AD 1739 eruption. Macroscopically, they show a high degree of vesiculation

along with the presence of several filament-like structures. Optical analysis showed a glassy matrix with a few occurrences of small Cpx, Plg and minor Ol phenocrysts. The collected samples did not highlight significant inter-sample textural and compositional variations. Due to these similarities, we selected one representative sample to be utilized in this study.

Table 1 - Concentrations (in weight %) of major elements in the end-member glasses used in the mixing experiments.

	Shoshonitic end-member (wt.%)	Latitic end-member (wt.%)	Rhyolitic end-member (wt.%)
SiO_2	52.22	58.81	73.90
Al_2O_3	16.38	16.36	13.73
TiO_2	0.78	0.58	0.10
FeO_t	8.84	6.31	1.81
MgO	4.61	3.20	0.22
CaO	8.84	6.12	1.05
Na_2O	5.39	4.77	3.69
K_2O	2.94	3.85	5.49
Total	100.00	100.00	100.00

Experimental apparatus and method

Magma mixing experiments were performed using the Chaotic Magma Mixing Apparatus (COMMA, Morgavi *et al*., 2015; Fig. 2), installed at the Department of Physics and Geology (University of Perugia), and designed to conduct experiments at high temperature with high-viscosity natural and synthetic magmas.

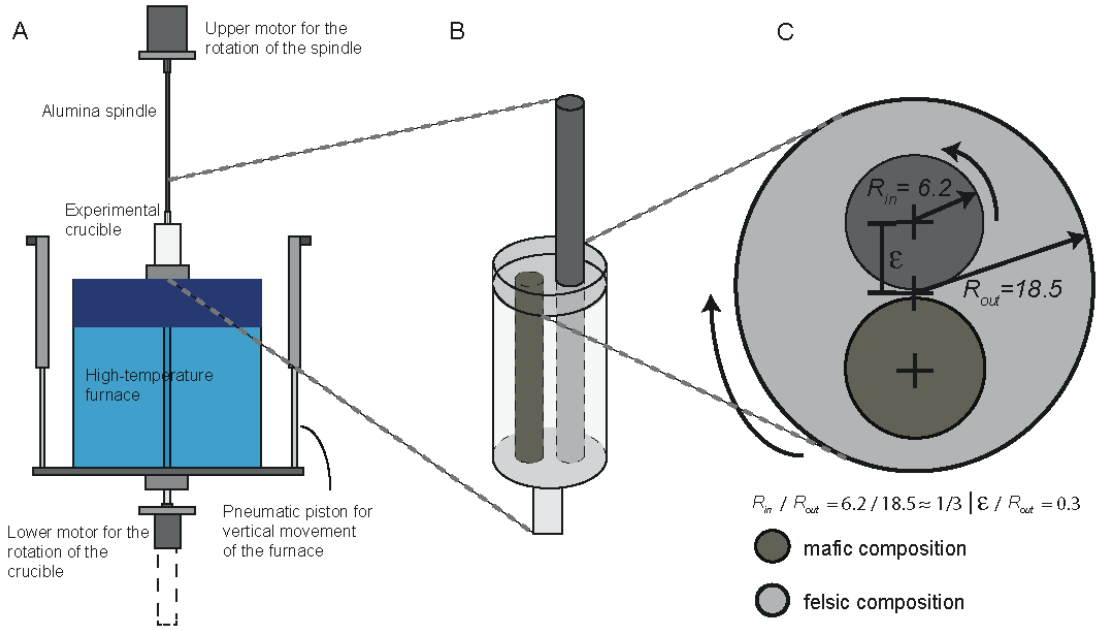

Fig. 2 - Schematic representation (not to scale) of the COMMA (reproduced and slightly modified from Morgavi *et al*., 2015). A) Complete experimental setup showing the high-temperature furnace, the upper and lower motors for the rotation of the spindle and the crucible hosting the end-member melts, and the positioning of the experimental crucible; B) enlargement of the experimental crucible filled with the end-member compositions; C) section of the crucible perpendicular to its vertical axis showing the relative position of the end-members and the spindle. Geometrical parameters of the used experimental protocol are also shown (see Morgavi *et al*., 2015 for further details).

We performed two main sets of mixing experiments using a specific mixing protocol able to produce efficient mixing dynamics in the experimental products (Fig. 3; see also Morgavi *et al*., 2015 and Rossi *et al*.,

2017 for further details). The experiments were performed with different run durations, progressively increasing, to observe the space-time evolution of the system due to the application of the mixing protocol (Fig. 3).

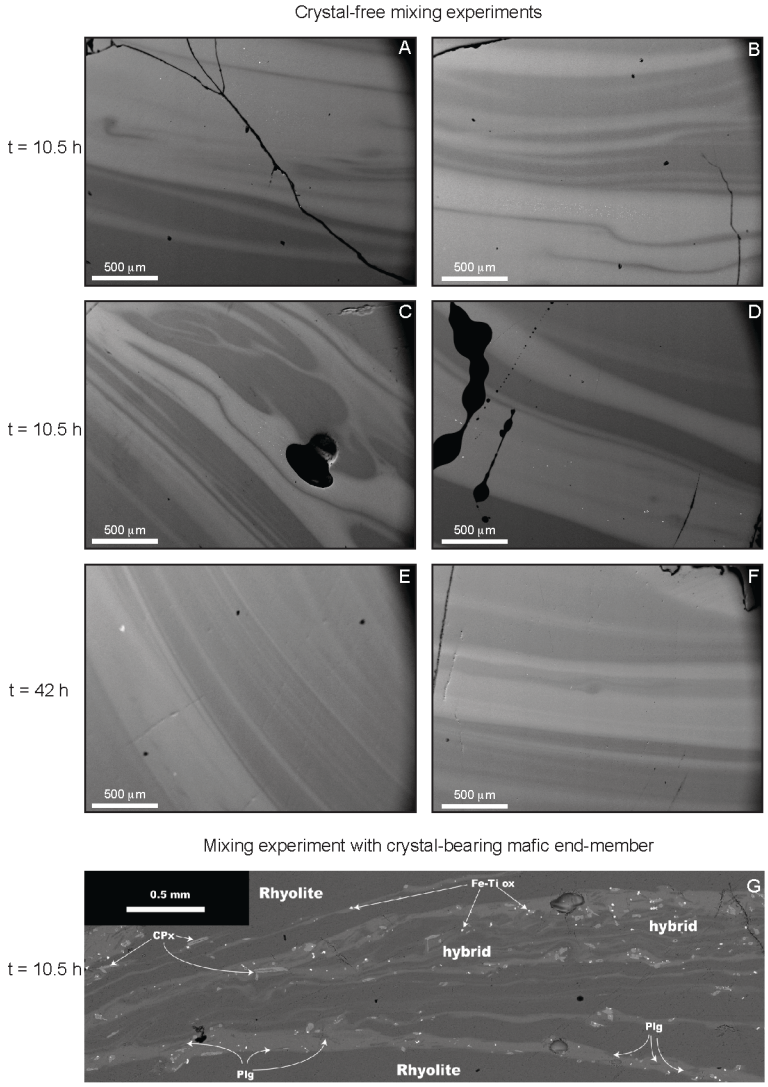

Fig. 3.- A-F): Representative back-scattered electron images (BSE) of selected areas of the experimental samples showing the intricate morphologies produced during the first set of mixing experiments. Mixing times (*t*) of each experiment are also reported. G): BSE collage showing the intricate filament-like and melt + crystals structures produced after the mixing experiment with the crystal-bearing shoshonitic end-member. Abbreviations: Cpx: clinopyroxene, Plg: plagioclase, Fe-Ti ox.: Fe-Ti oxides.

The first set of experiments, using a rhyolite and a shoshonite as felsic and mafic end-members, was made in order to test the reproducibility of results, as well as the robustness of the experimental method. The second set, using a rhyolite and a latite as felsic and mafic end-members, was made to compare the experimental results to natural samples produced by the AD 1739 eruption at Vulcano Island. More detailed and specific information on the experimental procedure are reported in Rossi *et al*. (2017, 2019) for the first and second set of experiments, respectively. Finally, last experiment was made in order to investigate the influence of a crystal-bearing shoshonitic end-member on the rhyolite-shoshonite mixing system. A summary of the experimental conditions adopted in the experiments is indicated in Table 2.

Major element concentrations and back-scattered electron images (Fig. 3) were acquired for the experimental products of each experiment using a Cameca SX100 electron microprobe at the Institut für

Mineralogie, Leibniz Universität Hannover, Germany. Additional details about the analytical conditions, as well as data reduction procedures, are reported in Rossi *et al.* (2017, 2019).

Table 2 - Experimental conditions adopted during magma mixing experiments. Detailed information are reported in Rossi *et al.* (2017, 2019). Abbreviations: RHY: rhyolite; SHO: shoshonite; LAT: latite.

	MAGMA MIXING EXPERIMENTS (COMMA; University of Perugia)					
	1° set			**2° set**		**3° set**
	Experiment A	***Experiment B***	***Experiment C***	***Experiment A***	***Experiment B***	***Experiment A***
Felsic end-member	RHY	RHY	RHY	RHY	RHY	RHY
Mafic end-member	SHO	SHO	SHO	LAT	LAT	SHO
Duration of the mixing protocol (hours)	10.50	10.50	42.00	10.50	31.50	10.50
Temperature (°C)	1200.00	1200.00	1200.00	1200.00	1200.00	1125.00
Pressure (atm)	1.00	1.00	1.00	1.00	1.00	1.00
H_2O (wt.%)	0.00	0.00	0.00	0.00	0.00	0.00
Volume of felsic end-member in the experiments (vol. %)	88.00	88.00	88.00	88.00	88.00	88.00
Volume of mafic end-member in the experiments (vol. %)	12.00	12.00	12.00	12.00	12.00	12.00
Crystal content in the felsic end-member (area %)	0.00	0.00	0.00	0.00	0.00	0.00
Crystal content in the mafic end-member (area %)	0.00	0.00	0.00	0.00	0.00	20.00

Concentration Variance Decay (CVD) method for the Mixing-to-Eruption timescales estimates

Mixing-to-eruption timescales estimates were developed using the method of Concentration Variance Decay (CVD, Perugini *et al.*, 2015; Fig. 4).

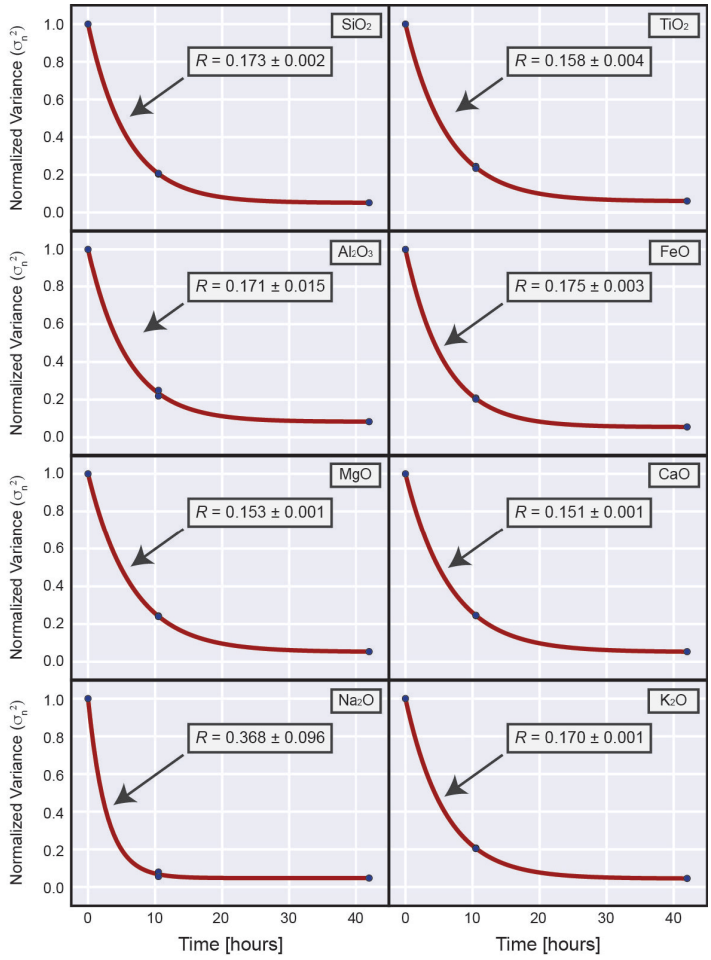

Fig. 4 - Variation of concentration variance as a function of mixing time for the analyzed chemical elements fitted using Eq. (3) (see text). The value of R (*i.e.*, the rate at which concentration variance decays with time) is reported for each element. Uncertainty on R is reported considering the uncertainty related to both the fitting and chemical analyses.

It was utilized to capture the time elapsing between the beginning of the mixing process (*i.e.*, the injection of new mafic magma in the reservoir) and the eruption. It consisted of experimentally evaluating the rate at which the concentration variance of the different chemical elements decayed with time during the mixing process (*e.g.*, Morgavi *et al*., 2013; Perugini *et al*., 2015). For a given chemical element, the concentration variance is defined as follows:

$$\sigma^2(C^i) = \frac{\sum_{n=1}^{N}(C_n^i - \mu^i)}{N} \qquad Eq.\ 1$$

where N is the number of the analytical determinations, C_n^i is the concentration of a given chemical element i in the analytical determination n, and μ^i is the average composition for the element i. The concentration variance decreases as mixing time increases (and the system moves towards homogeneity) and depends on the concentrations of each element i.

In this work, considering the concentration of chemical elements in the experimental sample for the rhyolite-latite mixing system (Table 1) and following the method of Morgavi *et al*. (2013) and Perugini *et al*. (2015), we normalized he variance values to the initial variance utilizing the equation:

$$\sigma_n^2(C^i) = \frac{\sigma^2(C^i)_t}{\sigma^2(C^i)_{t=0}} \qquad Eq.\ 2$$

where $\sigma^2(C^i)_t$ and $\sigma^2(C^i)_{t=0}$ are, respectively, the variance in the chemical element (C^i) at time t and $t = 0$ (i.e. before the beginning of the experiment or mixing event in nature).

The successive step consisted of the derivation of empirical relations, typically exponential, linking the variance decay with time and expressed by the following equation:

$$\sigma_n^2(C^i) = C_0 exp(-Rt) + C_1 \qquad Eq.\ 3$$

where C_0 and C_1 represent the value of the concentration variance $[\sigma_n^2(C^i)]$ at $t = 0$ and $t = \infty$, respectively, and *t* is the mixing time (Fig. 4). *R* ("relaxation of concentration variance"; RCV) quantifies the rate at which the concentration variance decays with mixing time and the mobility of the chemical elements in the system (Fig. 4).

We applied the CVD method to the experimental products and then, comparing the concentration variances calculated also for the natural sample, i.e. the banded pumice generated by the fallout events of the early explosive phase of the AD 1739 eruption, to the parametrization derived from the experiments of the rhyolite-latite mixing system, we provided an estimate of the mixing-to-eruption timescales.

RESULTS AND DISCUSSIONS

Magma mixing in the AD 1739 and 1888-90 eruptions at Vulcano Island

Magma mixing has been indicated as having a primary role in the pre-eruptive dynamics of the plumbing system at Vulcano Island, also directly related to the eruptive events. In the case of the AD 1739 eruption, it has been suggested that the most plausible scenario was a shallow reservoir (*i.e.*, 3-5 km; De Astis *et al*., 2013) in which a latitic magma from a deeper reservoir (*i.e.*, 20-25 km; De Astis *et al*., 2013) intruded a rhyolitic magma, finally leading to eruptive event (Vetere *et al*., 2015). Part of the volume of the latite remained in the reservoir after the conclusion of the eruptive event, to be erupted during the last cycle of AD 1888–90. In the case of the AD 1888–90 eruptive cycle, Clocchiatti *et al*. (1994) highlighted that the erupted pyroclastic products were derived from mixing between a rhyolitic end-member and a latite of similar composition to the enclaves detected in the Pietre Cotte lava flow. They suggest the unlocking of the rhyolite by re-heating due to the injection of the hotter latite, emphasizing the direct link between the mixing process, occurring in the shallow reservoir, and the subsequent eruption.

Timescales estimates for the AD 1739 eruption

The presented magma mixing experiments aimed to define the mixing-to-eruption timescales, i.e. the time elapsed between the beginning of the mixing in the magmatic reservoir and the subsequent eruption, for the AD

1739 eruption at Vulcano Island. BSE images of some representative experiments (Fig. 3) show the presence of stretched and folded alternating filaments of the two end-members produced by the complex dynamics acting during the mixing process (*i.e.*, mixing protocol). Chemical investigations of major elements (*i.e.*, Si, Mg, Ca, Ti, Fe, Na, Al, and K) have been made for experimental products in order to evaluate the evolution of the normalized concentration variance, $\sigma^2(C^i)$, as a function of time. As for the experimental products, the normalized variance was also estimated for the natural sample.

Mixing-to-eruption timescales were then estimated using Eq. (3). The obtained values show a variability ranging from ca. 10 to 39 h (*i.e.*, the minimum and the maximum value, respectively), with an average of 29□9 (s.d.) h. The obtained timescales, of the order of days, are very short if compared to the few other estimates for volcanic systems of the Aeolian archipelago ranging from years to decades (*e.g.*, Gioncada *et al*., 2005 and Petrone *et al*., 2018); however, they agree with the lower bound of the estimates reported by Petrone *et al.* (2018), of the order of days to months, for clinopyroxene portions capturing the final stages (*i.e.*, final injections) of the mixing process at Stromboli.

The obtained timescales were then used to infer the temporal evolution of the shallow magmatic system beneath the La Fossa cone before the AD 1739 eruption. In detail, at the initial time t=0, a rhyolitic reservoir was at ca. 3–5 km. The ascent of the latitic magma from a deeper reservoir (20–25 km; De Astis *et al*., 2013) produced a re-heating of the rhyolite, triggering the unlocking of the system and the mixing process, lasting for a maximum of ca. 39 h, and finally leading to the AD 1739 eruptive event.

The role of crystals on mixing dynamics: an introductory overview

Despite of the great occurrence of magma mixing in nature, the physicochemical processes involved in the process are still poorly understood (*e.g.*, Perugini & Poli, 2012). When two or more magmas mix a variety of hybrids can develop. The composition of the end-member magmas has a strong influence on the outcome of mixing. Because magmas usually carry early crystals, these are mechanically transferred to the hybrid magmas during mixing. The transferred crystals typically develop disequilibrium features owing to reaction with the new host magma composition (*e.g.*, Ubide *et al*., 2014).

To deeper investigate the influence of crystals on mixing dynamics, we performed a mixing experiment using a crystal-bearing mafic end-member. The experimental and analytical procedures were analogue to the ones adopted for the crystal-free mixing experiments. Detailed information on experimental conditions are reported in Table 2.

We calculated the normalized concentration variance, $\sigma^2(C^i)$, for the analyzed major elements in the experimental product (*i.e.*, Si, Mg, Ca, Ti, Fe, Na, Al, and K) and then compared the results to the values obtained for the first series of experiments performed at identical run duration conditions (refer to Table 2 and to the work of Rossi *et al.*, 2017 for further information). Results pointed to a considerable role played by the crystals in significantly enhancing mixing process due to their mechanical transfer from the parental magma to the other (Fig. 3G), also promoting the development of diffusion dynamics due to the increasing contact areas between the interacting magmas (Ubide *et al*., 2014).

Further investigations are worthy of interest, since the understanding of the role of perturbing factors on modulating the mixing dynamics may have also potential implications on a better comprehension of the pre-eruptive conditions of magmatic systems.

CONCLUDING REMARKS

Igneous and volcanic rocks subjected to magmatic interaction preserve a “frozen image” of a precise moment during the evolution of the process (*e.g.*, Perugini & Poli, 2012) whose spatial and temporal development can be tracked by the investigation of the mixing-related features observed in the rocks.

Performing mixing experiments under controlled experimental conditions, we could attest that mixing-related morphologies observed in natural rocks are faithfully reproducible in the experimental products and that there is a match in the trends of chemical elements between experimental and natural samples. Also, we used

mixing as a volcanic geo-chronometer to make inferences on the time elapsing between the occurrence of mixing in the pre-eruptive dynamics of active volcanoes and the eruptions. This may have significant implications in the context of volcanic hazard mitigation and planning of emergency activities since short mixing-to-eruption times (and, as a consequence, high velocities of ascending magmas) may imply little warning in volcanic crises.

Although our experimental approach presents some limitations (Rossi *et al.*, 2017), whose overcoming should be the subject of future in-depth studies, it could represent a good starting point in the development of a unifying model able to explain the complex magma mixing dynamics and their role in the pre-eruptive dynamics of active volcanoes.

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