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Exponential decay of concentration variance during magma mixing: Robustness of a volcanic chronometer and implications for the homogenization of chemical heterogeneities in magmatic systems

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ABSTRACT

The mixing of magmas is a fundamental process in the Earth system causing extreme compositional variations in igneous rocks. This process can develop with different intensities both in space and time, making the interpretation of compositional patterns in igneous rocks a petrological challenge. As a time-dependent process, magma mixing has been suggested to preserve information about the time elapsed between the injection of a new magma into sub-volcanic magma chambers and eruptions. This allowed the use of magma mixing as an additional volcanological tool to infer the mixing-to-eruption timescales. In spite of the potential of magma mixing processes to provide information about the timing of volcanic eruptions its statistical robustness is not yet established. This represents a prerequisite to apply reliably this conceptual model. Here, new chaotic magma mixing experiments were performed at different times using natural melts. The degree of reproducibility of experimental results was tested repeating one experiment at the same starting conditions and comparing the compositional variability. We further tested the robustness of the statistical analysis by randomly removing from the analysed dataset a progressively increasing number of samples. Results highlight the robustness of the method to derive empirical relationships linking the efficiency of chemical exchanges and mixing time. These empirical relationships remain valid by removing up to 80% of the analytical determinations. Experimental results were applied to constrain the homogenization time of chemical heterogeneities in natural magmatic system during mixing. The calculations show that, when the mixing dynamics generate millimetre thick filaments, homogenization timescales of the order of a few minutes are to be expected.

1. Introduction

Magma mixing is a major petrogenetic process concurring in the generation of the wide compositional diversity of igneous rocks on Earth (Bacon, 1986; Bateman, 1995; Eichelberger, 1978; Sparks et al., 1977) in both the plutonic and volcanic environment (Albert et al., 2015; Anderson, 1976, 1982; De Rosa et al., 1996; Kratzmann et al., 2009; Perugini and Poli, 2005; Wiebe, 1994). In this work, according to Perugini and Poli (2012) we refer to magma mixing as the process combining the physical dispersion of the two magmas and the development of the chemical exchanges between them.

The evidence of magma mixing processes remained recorded by a range of structural and textural features in igneous rocks, including the occurrence of mineral phases showing thermo-chemical disequilibria (Anderson, 1984; Didier and Barbarin, 1991; Hibbard, 1981; Wada, 1995; Wallace and Bergantz, 2002), magmatic enclaves dispersed into compositionally different host rocks (Bacon, 1986; Vetere et al., 2015), and bandings of different magma compositions (Flinders and Clemens, 1996; Morgavi et al., 2016).

Geochemically, magma mixing is witnessed by extreme compositional variations of major and trace elements and isotopes, which can occur in the rocks even at very short length scales, of the order of millimetres or micrometres (Laeger et al., 2017; Montagna et al., 2015; Wiesmaier et al., 2015). The presence of compositionally different domains at such short length scale can cause diffusive fractionation processes of chemical elements that, due to their different mobility in magmas (Perugini et al., 2006), generate volumes of melts whose compositional variation is difficult to reconcile with classical geochemical models (e.g. Fourcade and Allegre, 1981).

The use of numerical models and experimental petrology provided a powerful tool in the study of magma mixing, and several attempts have been made to capture the most relevant parameters involved during the interaction between magmas (Bergantz et al., 2015; Kouchi and Sunagawa, 1985; Laumonier et al., 2014a, 2014b, 2015; Schleicher et al., 2016). These studies also highlighted an extreme complexity of the

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mixing process in space and time due to the interplay of the fluid dynamic regime and chemical exchanges between the interacting magmas.

The complexity of magma mixing processes has been shown to follow chaotic dynamics characterized by scale-invariant (fractal) compositional patterns (Flinders and Clemens, 1996; Perugini and Poli, 2005; Perugini et al., 2003a, 2003b, 2003c; Petrelli et al., 2011, 2016). The chaotic nature of magma mixing processes depends on the kinematics governing the flow fields of the magmatic mass. In particular, the fundamental process producing physical mixing is the stretching and folding of two fluids. The stretching and folding process also represents the basic dynamics leading to chaotic behaviour. As stretching and folding dynamics develop in time, an intricate lamellar pattern of flow structures is generated in which the interface area between magmas grows exponentially (e.g. Ottino et al., 1988). This is a pre-requisite for efficient mixing because chemical exchanges are enhanced through diffusion. Several physical processes can cause stretching and folding dynamics in magmas. The most important are: convective motions and plume-like dynamics of a light magma ascending into a denser one (e.g. Bateman, 1995; Cardoso and Woods, 1999; Cruden et al., 1995; Snyder and Tait, 1996), forced and fountain-like dispersion (e.g. Campbell and Turner, 1986), and ascent of magmas towards the Earth surface in dikes and channels (e.g. Blake and Campbell, 1986; Koyaguchi, 1985; Koyaguchi and Blake, 1989). These processes have been studied using both experimental and numerical approaches highlighting that they are capable of generating stretching and folding dynamics between fluids (e.g. Hydon, 1995; Meleshko and Van Heijst, 1995; Metcalfe et al., 1995).

An important point emerging from magma mixing studies is that it is a time dependent process, i.e. the longer the mixing time, the more homogeneous the mixture. For this reason, it has recently been suggested, on the basis of high-temperature experiments performed with natural melts, that the compositional variability due to magma mixing might be utilized to define new geo-chronometers to estimate the mixing-to-eruption timescales (Perugini et al., 2010, 2015b). However, in order to derive empirical relationships to constrain eruption timescales, a thorough assessment of the reliability of magma mixing experiments in terms of robustness and reproducibility of the results is needed. This is mostly due to the large experimental difficulties arising when dealing with such a complex process at high-temperature and with high-viscosity natural melts. These difficulties are further amplified by the chaotic nature of the mixing process that, by definition, might impede the reliable study of the long-term evolution of the process (e.g. Strogatz, 2001).

Motivated by these considerations, we performed a new set of chaotic magma mixing experiments using natural compositions with the aim to develop a robust statistical framework for the construction of mixing-to-eruption geo-chronometers. We evaluate the degree of reproducibility of mixing experiments, their statistical robustness and reliability. In particular, experiments at different mixing times and at condition relevant for natural magmatic systems were performed. The degree of reproducibility of experimental results was 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 further tested the robustness of our statistical analysis by randomly removing from the analysed experimental samples a progressively increasing number of samples and verifying the quality of the relationships obtained empirically. We then applied the obtained results to constrain the homogenization time of chemical heterogeneities in natural systems during the magma mixing process.

2. Materials and methods

2.1. Starting materials

Two natural lava samples from the island of Vulcano (Aeolian archipelago, Italy; (e.g. Gioncada et al., 2003; Keller, 1980) were used as end-members in the experiments. The least evolved (more mafic) end-member is a shoshonite sampled at the Vulcanello lava platform (hereafter shoshonitic composition SC; Davì et al., 2009). The most evolved is a high-K rhyolitic obsidian from the Pietre Cotte lava flow, belonging to La Fossa cone (hereafter rhyolitic composition RC; Clocchiatti et al., 1994; De Astis et al., 1997; Piochi et al., 2009). The chemical compositions of the shoshonitic and rhyolitic end-members are listed in Table 1.

Alteration-free rock samples were cleaned with distilled water and then crushed in an agate mortar to obtain a fine-grained powder. End-member powders were subsequently homogenized through two cycles of melting at 1600 °C for 4 h in a high temperature furnace (Nabertherm® HT 04/17) at ambient pressure using a $Pt_{80}Rh_{20}$ crucible followed by crushing and fine powdering in an agate mortar (e.g. Morgavi et al., 2015). End-member melts were characterized for their rheology using an Anton Paar RheolabQC viscometer installed at the Department of Physics and Geology of the University of Perugia (Italy). Viscosity of end-member melts at experimental temperature (1200 °C) are given in Table 1.

2.2. Experimental apparatus

The experimental device used for chaotic mixing experiment, named Chaotic Magma Mixing Apparatus (COMMA; Morgavi et al., 2015) and hosted at the Department of Physics and Geology of the University of Perugia (Italy), was specifically designed to work with viscous (up to 10^8 Pa s) natural and synthetic melts up to a temperature of 1500 °C. The COMMA produces chaotic dynamics in the mixing system using a stirring protocol known in the literature as Journal Bearing System (JBS; e.g. Swanson and Ottino, 1990). Our high-temperature system consists of a $Pt_{80}Rh_{20}$ crucible (i.e. the outer cylinder), filled with the two end-member compositions and an inner, off-centred spindle (i.e. the inner cylinder; Fig. 1). Two parameters define the geometry of the COMMA assembly: (a) the ratio of the radii between the two cylinders, $r = R_{in}/R_{out} = 1/3$, and (b) the eccentricity ratio to the outer cylinder $\varepsilon = \delta/R_{out} = 0.3$, where δ is the distance between the centres of the inner and outer cylinders (R_{in} and R_{out}) (Fig. 1). The alternate rotation of the crucible and the spindle in opposite directions triggers chaotic mixing within the system (e.g. Galaktionov et al., 2002; Swanson and Ottino, 1990). The time-periodic motion of the crucible and the spindle introduces two additional parameters: the rotation angle θ of the outer cylinder (i.e. the crucible) and the ratio Ω between the rotation angles of the outer and the inner cylinders (i.e. the crucible and the spindle). For the present work, θ and Ω were fixed to 2π and 3 respectively. The main fluid-dynamical parameters characterizing COMMA experiments are summarized in Table 2. The reader is redirected to the work of Morgavi et al. (2015) for a thorough description of the experimental apparatus.

Experiments were performed using volume percentages of RC and SC end-members of 88% and 12%, respectively. At completion of each

Table 1
Concentrations (in weight %) of major elements in the rhyolitic and shoshonitic end-member glasses used in the chaotic mixing experiments. The viscosity of both end-members is also reported.

	Shoshonitic end-member (SC) wt%	Rhyolitic end-member (RC) wt%
SiO_2	52.22	74.32
Al_2O_3	16.38	13.21
K_2O	2.94	5.81
TiO_2	0.78	0.10
FeO_t	8.84	1.60
CaO	8.84	0.96
Na_2O	5.39	3.75
MgO	4.61	0.25
Total	100.00	100.00
Viscosity (Pa s) @ 1200 °C	1.8×10^2	1.6×10^5