



Chlorine isotope composition of volcanic rocks and gases at Stromboli volcano (Aeolian Islands, Italy): Inferences on magmatic degassing prior to 2014 eruption



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ABSTRACT

Among the magmatic volatiles, chlorine (Cl) is degassed at shallow depths offering the opportunity to investigate the behavior of magmatic degassing close to the surface, and the possible occurrence of chemical and isotopic fractionation related to gas/melt partitioning. However, it is still unclear if the isotopic composition of Cl ($\delta^{37}\text{Cl}$) can be used as a proxy of magmatic degassing. In this work, we investigate the concentrations of chlorine and sulfur, and the Cl isotope composition of rocks and plume gases collected at Stromboli volcano, Aeolian Islands, Italy. This volcano was chosen because it is characterized by persistent eruptive activity (i.e., Strombolian explosions) and by the presence of magma at very shallow levels in the conduits. Rocks belonging to the different magmatic series erupted throughout the formation of the volcano have $\delta^{37}\text{Cl}$ values ranging between -1.0 and $+0.7\text{‰}$. The isotopic composition seems independent of the Cl concentration of the rocks, but shows a negative correlation with SiO_2 content. Plume gases have a greater isotopic compositional variability than the rocks ($-2.2\text{‰} \leq \delta^{37}\text{Cl} \leq +1.5\text{‰}$) and the composition seems related to the level of volcanic activity at Stromboli. Gases collected in 2011–2013 during days of ordinary eruptive activity are characterized by $\delta^{37}\text{Cl}$ values ranging from $+0.3$ to $+1.5\text{‰}$ and S/Cl molar ratios between 1.4 and 2.2, similar to previous S/Cl measurements performed at Stromboli with other techniques. Plume gases collected in July 2014, in days of high-level eruptive activity preceding the onset of the 2014 effusive eruption, have negative $\delta^{37}\text{Cl}$ values ($-2.2\text{‰} \leq \delta^{37}\text{Cl} \leq -0.1\text{‰}$) and S/Cl between 0.9 and 1.2, which are among the lowest S/Cl values measured at this volcano. The amplitude of the volcanic tremor and the variation in the inclination of very long period (VLP) seismic signal polarization clearly indicate that in July 2014 the intensity and frequency of Strombolian explosions, as well as the level of magma in the conduits, were higher than normal. This suggests that when magmatic degassing occurs at very shallow depths (magma residing at few tens of meters below the craters), isotopic fractionation between gaseous and dissolved chlorine ($\text{In}\alpha_{\text{gas-melt}} > 0$) can occur, resulting in a depletion of ^{37}Cl in the melt. Finally, we argue that the range of $\delta^{37}\text{Cl}$ values measured in rocks and gases at Stromboli overlaps most of the known mantle reservoirs (i.e., DMM, HIMU and EM-EM1), indicating that the transfer of chlorine from the mantle to the surface occurs without significant isotopic fractionation. This work opens new perspectives for better constraining the isotope signature of chlorine of different magmatic systems and mantle reservoirs on the Earth.

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1. Introduction

At open conduit volcanoes, volatile elements dissolved in magma are discharged into the atmosphere mostly through volcanic plumes and are responsible for the eruption style of volcanoes, as well as for climate

changes after catastrophic events. Therefore, volatile solubility in silicate melts and gas/melt partitioning coefficients provide fundamental information on the magma supply at depth, extent of magmatic degassing, and possible trigger mechanisms of eruptions (Allard et al., 2005; Aiuppa et al., 2010; Oppenheimer et al., 2011 and references therein). Volatile abundance in melt inclusions provides unique opportunities to define the thermodynamic conditions of deep magma bodies and to constrain the models of magmatic degassing for explaining

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eruption dynamics (e.g., Spilliaert et al., 2006a, 2006b; Métrich et al., 2010, 2001).

In particular, sulfur and chlorine provide useful information on magmatic degassing during the last kilometers of ascent of silicate melts due to their high solubility in magmas (e.g., Carroll and Webster, 1994; Alletti et al., 2009; Lesne et al., 2011 and references therein). For example, at Mount Etna volcano (Italy), Spilliaert et al. (2006a, 2006b) inferred that 22–50% of the starting amount of Cl is lost upon the eruption, and the remaining fraction is dissolved in the melt up to the surface; conversely, almost the sulfur is lost at low pressure. At Stromboli volcano (Aeolian Islands, Italy), Métrich et al. (2010) observed that during steady-state activity bulk degassing reaches 99% and $\geq 69\%$ for S and Cl, respectively. They also highlighted that the Cl content of shoshonitic melt inclusions, as well as their S/Cl ratios, are similar to those of the glass matrix. The low bulk degassing of Cl, compared to S, derives from its moderate volatility. In addition, Cl behaves as an incompatible element during magma evolution, thus being concentrated in residual magma (Wade et al., 2006). The solubility of Cl depends on the melt composition, water content and pressure, with opposite effects of pressure and crystallization upon cooling (Webster et al., 1999; Métrich et al., 2010 and reference therein).

Stromboli volcano (Fig. 1) is famous worldwide for the persistence of its eruptive activity (so-called “Strombolian activity”), which began between the third and seventh century AD (e.g., Rosi et al., 2000) and has continued to the present day with no significant interruptions. This eruptive activity consists of explosions usually at intervals of 10–20 min that are associated with continuous degassing. This steady-state explosive activity is driven by the intermittent rise and bursting of gas slugs rising separately from their source melt 2–3 km beneath the summit craters (Ripepe et al., 2001; Chouet et al., 2003; Burton et al., 2007a). Allard et al. (1994) estimated that explosive degassing is a minor contributor to the total gas flux, which is dominated by quiescent degassing. Thus, this volcano represents an ideal natural laboratory to study the behavior of sulfur and chlorine during shallow magmatic

degassing. By using an open-path FTIR scanning system, La Spina et al. (2013) measured homogeneous SO_2/HCl molar ratios ($1.0 < \text{SO}_2/\text{HCl} < 2.7$ from 2009 to 2010) in volcanic plumes from three different craters of Stromboli. They observed that puffing should be strongly linked with the degassing of the most soluble gases such as HCl. Between 2011 and 2013 at Stromboli, Wittmer et al. (2014) performed three field campaigns to directly sample acidic species that are discharged from the magma in the gaseous form, by using active alkaline traps. These authors obtained S/Cl ratios between 1.4 and 2.2, overlapping the range of FTIR measurements performed by La Spina et al. (2013).

In order to better understand the melt/gas partitioning of Cl during magmatic degassing and the genesis of fluids in the hydrothermal systems, in recent years the isotopic composition of chlorine ($^{37}\text{Cl}/^{35}\text{Cl}$) has been studied in gases, rocks and hydrothermal fluids from several volcanic systems (e.g., Bonifacie et al., 2005; Liebscher et al., 2006; Rizzo et al., 2013; Sharp et al., 2010b; Fischer et al., 2015). This isotope ratio is also used to trace shallow magmatic activity (e.g., Li et al., 2015) and to constrain the mantle source (e.g., Barnes et al., 2008, 2009; Bonifacie et al., 2008; Halldórsson et al., 2016; John et al., 2010; Sharp et al., 2013), although it does not show a wide variability in the Earth reservoirs. The mantle-, crust-, and seawater-derived chlorine exhibits variation within a few per mil ($\delta^{37}\text{Cl}$). Such a homogeneity has been ascribed to an excess of H with respect to Cl and to the observation that the dominant volatile chloride species is gaseous HCl with a limited isotopic fractionation between silicate melt and gaseous HCl (Sharp et al., 2010a). On the other hand, significant fractionation processes can occur when gaseous HCl interacts with liquid phases (e.g., in high-temperature fumaroles, Sharp et al., 2010b; Rizzo et al., 2013; Rodríguez et al., 2016). Sharp et al. (2010b) observed that ^{35}Cl is preferentially incorporated into the aqueous chloride species, leaving the HCl(g) with ever-increasing $\delta^{37}\text{Cl}$ values. At Mt. Etna, Rizzo et al. (2013) found that plume gases (representative of gases directly degassed from magma) and rocks exhibited very similar chlorine isotopic compositions. However, some fumarole gases were strongly enriched in ^{37}Cl with respect to the plume gases. The authors ascribed the ^{37}Cl enrichment to the preferential removal of ^{35}Cl when HCl(g) interacts with water. Similarly at Poás volcano (Costa Rica), Rodríguez et al. (2016) observed that the $\delta^{37}\text{Cl}$ values of fumarolic gas that experienced little or no interaction with subsurface water and followed a relatively dry pathway were closest to magmatic values, whereas elevated $\delta^{37}\text{Cl}$ values can be explained by fractionation during subsurface gas–liquid interaction.

In this work, we determined the concentration of chlorine and sulfur and the isotope composition of chlorine of a selected suite of mafic rocks and ultramafic mantle cumulates during the magmatic history of Stromboli volcano. The same measurements were also made on plume gases emitted from the craters and collected during a few campaigns from 2011 to 2014, with the last samples being collected few weeks before the onset of the 2014 effusive eruption. Data from rocks and gases are discussed in terms of magmatic degassing, and put in the context of the increase of eruptive activity in 2014. We finally discuss the isotope composition of chlorine in the mantle source beneath Stromboli.

2. Petrochemical overview of the Stromboli magmatism

Stromboli is part of an active volcanic arc due to ongoing subduction of African plate beneath the European plate (Gasparini et al., 1982). During the last 100 ky, Stromboli has been erupting subaerial products characterized by chemical compositions ranging from calcalkaline (CA) to high-K calcalkaline (HKCA), potassic (KS), and shoshonitic (SHO) series (Fig. 2), with the latter including the magmas of the present activity (e.g., Francalanci et al., 1989).

Mafic products ($\text{SiO}_2 < 55\%$, $\text{MgO} > 4\%$) are present in all of the erupted magmatic series (CA, HKCA, SHO, KS). There is no simple temporal variation in the alkalinity of magmas. In certain cases, products that belong to different series have erupted contemporaneously (Francalanci et al., 1989). The general K increase from the CA to the KS

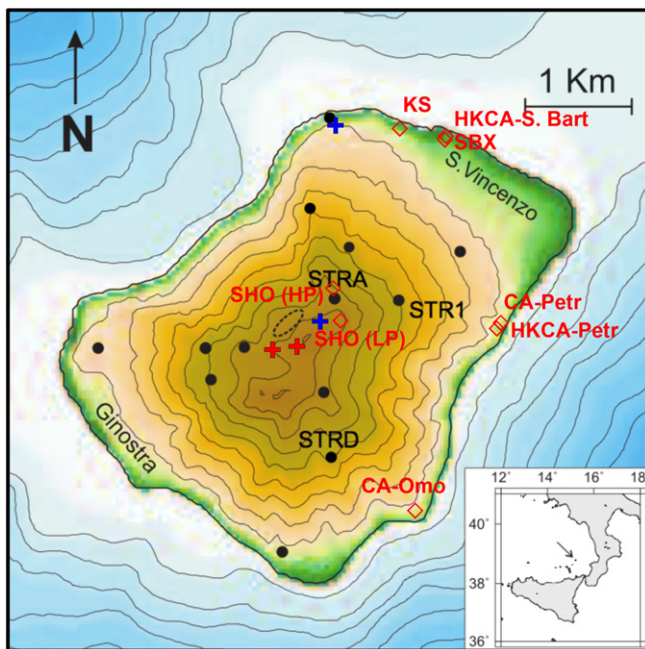


Fig. 1. Map of Stromboli. Crater area (dashed ellipse); Plume sampling sites (red crosses); Background sampling sites (blue crosses); rock sampling sites (red diamonds); INGV broadband seismic network of Stromboli (black dots) (see De Cesare et al., 2009 for details).

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