



## Chlorine isotopes of thermal springs in arc volcanoes for tracing shallow magmatic activity



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### ABSTRACT

The evaluation of the status of shallow magma body (i.e., from the final intrusion stage, to quiescence, and back to activity), one of the key parameters that trigger and sustain volcanic eruptions, has been challenging in modern volcanology. Among volatile tracers, chlorine (Cl) uniquely exsolves at shallow depths and is highly hydrophilic. Consequently, Cl enrichment in volcanic gases and thermal springs has been proposed as a sign for shallow magmatic activities. However, such enrichment could also result from numerous other processes (e.g., water evaporation, dissolution of old chloride mineral deposits, seawater contamination) that are unrelated to magmatic activity. Here, based on stable isotope compositions of chloride and dissolved inorganic carbon, as well as previous published  $^3\text{He}/^4\text{He}$  data obtained in thermal springs from two recently erupted volcanoes (La Soufrière in Guadeloupe and Montagne Pelée in Martinique) in the Lesser Antilles Arc, we show that the magmatic Cl efficiently trapped in thermal springs displays negative  $\delta^{37}\text{Cl}$  values ( $\leq -0.65\%$ ), consistent with a slab-derived origin but distinct from the isotope compositions of chloride in surface reservoirs (e.g. seawater, local meteoric waters, rivers and cold springs) displaying common  $\delta^{37}\text{Cl}$  values of around 0‰. Using this  $\delta^{37}\text{Cl}$  difference as an index of magmatic Cl, we further examined thermal spring samples including a 30-year archive from two thermal springs in Guadeloupe covering samples from its last eruption in 1976–1977 to 2008 and an island-wide sampling event in Martinique in 2008 to trace the evolution of magmatic Cl in the volcanic hydrothermal systems over time. The results show that magmatic Cl can be rapidly flushed out of the hydrothermal systems within <30 to 80 years after the eruption, much quicker than other volatile tracers such as  $\text{CO}_2$  and noble gases, which can exsolve at greater depths and constantly migrate to the surface. Because arc volcanoes often have well developed hydrothermal systems where magmatic Cl is easily transferred to the surface following its exsolution from shallow magma body, we suggest that  $\delta^{37}\text{Cl}$  has great potential to be a unique proxy to monitor the cessation and revival of infrequent arc volcanoes, particularly at centennial time scales.

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### 1. Introduction

Arc volcanoes produce infrequent but highly harmful explosive eruptions (Linsay et al., 2005; Siebert et al., 2010). Despite more than 50 years of hydrogeological, geophysical (e.g., seismic,

structure, deformation, gravity) and geochemical monitoring in arc volcanoes (e.g., Feuillard et al., 1983; Jackson et al., 1998), it is still difficult to probe activity of shallow magma body, notably the transition from the eruption-driven magmatic degassing stage to dormancy. Compared with the difficulties in interpreting complex secondary hydrogeological and geophysical signals generated in response to the expansion and/or collapse of magma chamber, the geochemical characterization of magmatic volatile tracers allows to retrieve direct information of the parent magma and to evaluate its role at shallow depths in triggering and sustaining

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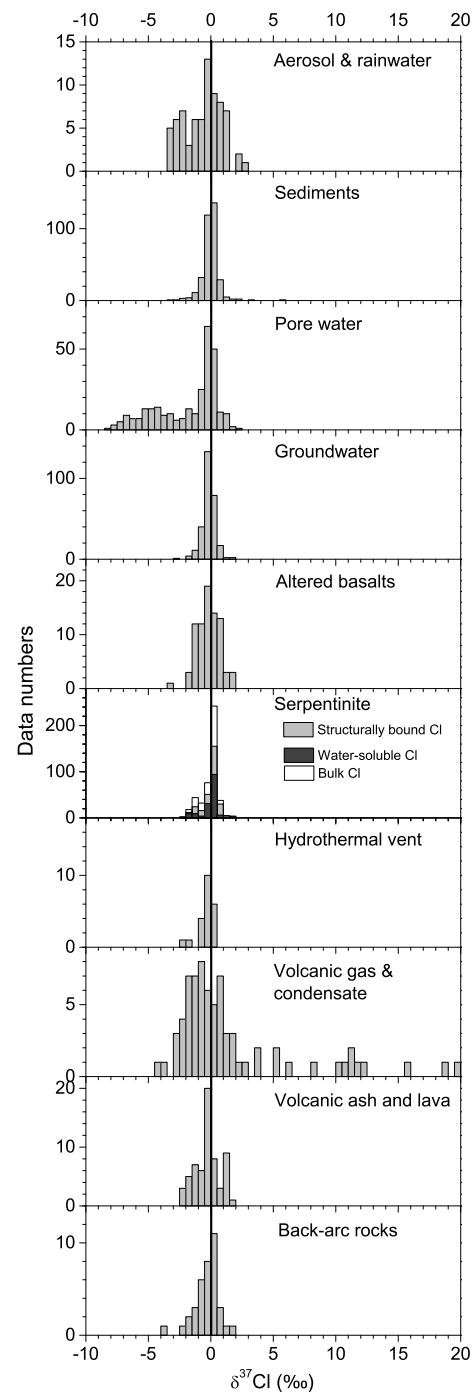
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the volcanic eruption (Edmonds, 2008). For this reason, volatile compounds degassed from arc magmas ( $\text{CO}_2$ , noble gases,  $\text{SO}_2$ ,  $\text{H}_2\text{S}$ , and halogens) have been intensively monitored in fumarole gases and hydrothermal waters in numerous volcanoes (e.g., Symonds et al., 1994; Van Soest et al., 1998; Pedroni et al., 1999; Fischer and Marty, 2005; Ruzié et al., 2012, 2013). However, most of these volatile tracers cannot be unambiguously used to infer shallow magmatic activity but rather be used to infer the degassing style, because either they (e.g.,  $\text{CO}_2$  and noble gases) can exsolve from great depths and migrate up to the surface through fractures, or they (e.g.,  $\text{SO}_2$ ,  $\text{H}_2\text{S}$  and  $\text{H}_2\text{O}$ ) are chemically reactive and easily altered by surface processes (Symonds et al., 1994; Allard et al., 1997; Fischer and Marty, 2005; Gaillard et al., 2011).

In contrast, chlorine (Cl) is unique compared with other volatile tracers because (1) the exsolution/degassing of Cl from magma is the latest, at depths <4 km to 0.4 km (Unni and Schilling, 1978; Webster et al., 1999; Spilliaert et al., 2006) close to the volcanic crisis loci; (2) Cl exists primarily as relatively inactive negatively monovalent state in melt (Evans et al., 2008), minerals (Volfinger et al., 1985), and fluids (Bonifacie et al., 2005); and (3) most importantly, degassed Cl (primarily in form of HCl) is highly hydrophilic and can be efficiently trapped and stored as chloride in the hydrothermal aquifers overlying magma bodies (e.g., thermal springs; Villemant et al., 2005, 2014), unless extreme conditions rarely observed in volcanic aquifers (e.g., sub-boiling temperatures and  $\text{pH} < 1$ ) are reached, which can induce degassing-related Cl loss from thermal springs (Ohba et al., 1994; Sharp et al., 2010). For comparison,  $\text{CO}_2$  and He migrating from the magma chamber are trapped only partially due to their low solubility in water. Based on such unique combination of chemical characteristics of Cl, its concentrations (hereafter  $[\text{Cl}^-]$ ) in thermal springs and fumaroles have been used to monitor shallow magmatic activity (Ohba et al., 1994; Symonds et al., 1994; Aiuppa et al., 2009). However,  $[\text{Cl}^-]$  alone cannot distinguish magmatic input from non-magmatic processes, such as evaporation condensation, dissolution of old chloride mineral deposits and seawater contamination.

Chlorine stable isotope compositions, on the other hand, could unequivocally distinguish magmatic Cl from surface Cl in high-temperature arc systems because of two major reasons. Firstly, Cl in arc magmas is mainly derived from underlying subducting slab (Straub and Layne, 2003; Sun et al., 2007; Pyle and Mather, 2009; John et al., 2010). Chlorine isotopic signatures of various reservoirs across subduction zones are summarized in Fig. 1. Among these reservoirs, seawater chloride is isotopically homogeneous throughout the world oceans with a  $\delta^{37}\text{Cl}$  value<sup>2</sup> of 0‰ (Godon et al., 2004b). Although all the sedimentary Cl (including evaporites) has a relatively large  $\delta^{37}\text{Cl}$  range from  $-3.16\text{‰}$  to  $+5.96\text{‰}$  (Kaufmann et al., 1984; Eggenkamp et al., 1995; Eggenkamp and Schuiling, 1995; Liu et al., 1996; Eastoe et al., 1999, 2001, 2007; Eastoe and Peryt, 1999; Arcuri and Brimhall, 2003; Tan et al., 2006), the seafloor sediments before the trench of Izu–Bonin–Mariana and Central American subduction zones have a relatively narrower  $\delta^{37}\text{Cl}$  range from  $-2.51\text{‰}$  to  $+0.67\text{‰}$  (Barnes et al., 2008, 2009b). The structurally bound Cl in these sediments, which represent the sedimentary Cl portion that can be possibly subducted to arc depth, have an average  $\delta^{37}\text{Cl}$  value of  $-0.81\text{‰}$  ( $n = 24$ ; data from Barnes et al., 2008, 2009b). A few measurements on eclogite-facies mica schists, which are considered to represent the deeply subducted marine sediments, show  $\delta^{37}\text{Cl}$  range from  $-2.2$  to  $+2.2\text{‰}$  with average value of  $-0.04\text{‰}$  (John et al., 2010). Chlorine in altered oceanic crust (AOC) has  $\delta^{37}\text{Cl}$  varying from  $-3.2$  to  $+1.5\text{‰}$



**Fig. 1.**  $\delta^{37}\text{Cl}$  variations of a variety of geological settings, including (1) aerosol and meteoric water (Volpe and Spivack, 1994; Volpe et al., 1998; Koehler and Wassenaar, 2010); (2) sediments, including evaporites (Kaufmann et al., 1984; Eggenkamp et al., 1995; Eggenkamp and Schuiling, 1995; Liu et al., 1997; Eastoe et al., 1999, 2001, 2007; Eastoe and Peryt, 1999; Arcuri and Brimhall, 2003; Tan et al., 2006; Barnes et al., 2008, 2009b); (3) pore water (Desaulniers et al., 1986; Eggenkamp et al., 1994; Ransom et al., 1995; Hendry et al., 2000; Hesse et al., 2000; Coleman et al., 2001; Spivack et al., 2002; Deyhle et al., 2004; Godon et al., 2004a; Lavastre et al., 2005; Bonifacie et al., 2007b; Wei et al., 2008); (4) groundwater (Kaufmann et al., 1984, 1993; Eastoe et al., 1999, 2001; Sie and Frape, 2002; Shouakar-Stash et al., 2007; Zhang et al., 2007; Stotler et al., 2010); (5) altered basalts (Sharp et al., 2007; Bonifacie et al., 2008b; Layne et al., 2009; Barnes and Cisneros, 2012); (6) serpentinite (Barnes et al., 2006, 2008, 2009a; Barnes and Sharp, 2006; Bonifacie et al., 2008a); (7) hydrothermal vent (Bonifacie et al., 2005, 2007b; Sharp et al., 2010); (8) volcanic gas and condensates (Wahrenberger et al., 1997; Barnes et al., 2008; Sharp et al., 2010; Rizzo et al., 2013); (9) volcanic ash and lava (Barnes et al., 2008; Barnes and Straub, 2010; Rizzo et al., 2013); (10) back-arc rock samples (Barnes et al., 2008; Layne et al., 2009; Barnes and Straub, 2010). A solid vertical line of 0‰ is added for reference.

<sup>2</sup> Defined as  $\delta^{37}\text{Cl} = \left(\frac{^{37}\text{Cl}/^{35}\text{Cl}}{^{37}\text{Cl}/^{35}\text{Cl}}\right)_{\text{Sample}} / \left(\frac{^{37}\text{Cl}/^{35}\text{Cl}}{^{37}\text{Cl}/^{35}\text{Cl}}\right)_{\text{Standard}} - 1$ , where standard is Standard Mean Ocean Chloride or SMOC.

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