



Chlorine isotope and Cl–Br fractionation in fluids of Poás volcano (Costa Rica): Insight into an active volcanic–hydrothermal system



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ARTICLE INFO

Article history:

Received 23 November 2015

18 April 2016

Accepted 31 May 2016

Available online 02 June 2016

Keywords:

Poás volcano

Chlorine

Bromine

Chlorine isotope

ABSTRACT

Halogen-rich volcanic fluids issued at the surface carry information on properties and processes operating in shallow hydrothermal systems. This paper reports a long-term record of Cl–Br concentrations and $\delta^{37}\text{Cl}$ signatures of lake water and fumaroles from the active crater of Poás volcano (Costa Rica), where surface expressions of magmatic-hydrothermal activity have shown substantial periodic changes over the last decades. Both the hyperacid water of its crater lake (Laguna Caliente) and subaerial fumaroles show significant temporal variability in Cl–Br concentrations, Br/Cl ratios and $\delta^{37}\text{Cl}$, reflecting variations in the mode and magnitude of volatile transfer. The $\delta^{37}\text{Cl}$ signatures of the lake, covering the period 1985–2012, show fluctuations between $+0.02 \pm 0.06\%$ and $+1.15 \pm 0.09\%$. Condensate samples from adjacent fumaroles on the southern shore, collected during the interval (2010–2012) with strong changes in gas temperature (107–763°C), display a much larger range from $-0.43 \pm 0.09\%$ to $+14.09 \pm 0.08\%$. Most of the variations in Cl isotope, Br/Cl and concentration signals can be attributed to interaction between magma-derived gas and liquid water in the volcanic–hydrothermal system below the crater. The $\delta^{37}\text{Cl}$ were lowest and closest to magmatic values in (1) fumarolic gas that experienced little or no interaction with subsurface water and followed a relatively dry pathway, and (2) water that captured the bulk of magmatic halogen output so that no phase separation could induce fractionation. In contrast, elevated $\delta^{37}\text{Cl}$ can be explained by partial scavenging and fractionation during subsurface gas–liquid interaction. Hence, strong Cl isotope fractionation leading to very high $\delta^{37}\text{Cl}$ in Poás' fumaroles indicates that they followed a wet pathway. Highest $\delta^{37}\text{Cl}$ values in the lake water were found mostly in periods when it received a significant input from subaqueous fumaroles or when high temperatures and low pH caused HCl evaporation. It is concluded that combined monitoring of $\delta^{37}\text{Cl}$ and Br/Cl in Laguna Caliente and adjacent fumaroles provides valuable information on activity in the subsurface hydrothermal system with significant relevance for volcanic surveillance of Poás.

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1. Introduction

Chlorine and bromine are generally extracted from degassing magma in the form of $\text{HCl}_{(\text{g})}$ and $\text{HBr}_{(\text{g})}$ (Gerlach, 2004; Aiuppa et al., 2005; Martin et al., 2006). While chlorine is generally the most abundant halogen in volcanic gases (Taran and Zelenski, 2015), bromine is present in amounts that are orders of magnitude lower. Gerlach (2004) reported an average molar Br/Cl ratio of 0.0022 ± 0.0020 for high-temperature gas condensates from arc volcanoes. Upon shallow emplacement of magma, both halogens are expelled during cooling and crystallizing, driven by their relatively high H_2O vapour/melt partition coefficients (Villemant and Boudon, 1999; Balcone-Boissard et al., 2010). In subsurface volcanic hydrothermal systems associated with intrusions, they tend to be

concentrated in fluid phases, showing a strongly hydrophilic behaviour. Since shallow hydrothermal processes can fractionate Cl and Br (Berndt and Seyfried, 1990, 1997; Lüders et al., 2002; Liebscher et al., 2006; Foustoukos and Seyfried, 2007), the proportions of these halogens in volcanic fluids are potential signals of interaction and separation processes in the trajectory between magma and atmosphere, such as boiling, condensation, evaporation, mixing and mineral precipitation (Villemant et al., 2003, 2005, 2014; Wu et al., 2012; Fischer et al., 2015). Likewise, the chlorine isotope compositions of fluids from shallow magma degassing are sensitive to fractionation during interaction between deep gases and shallow water bodies within a volcanic edifice (Sharp et al., 2010; Li et al., 2015). Volcanic fumaroles confirm this since the available data show large $\delta^{37}\text{Cl}$ variations compared to other terrestrial materials (Eggenkamp, 1994; Wahrenberger et al., 1997; Musashi and Eggenkamp, 2000; Barnes et al., 2008; Sharp et al., 2010; Rizzo et al., 2013).

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This paper explores the extent and causes of Cl–Br and chlorine isotope fractionation using time series data for fluids from the active crater of Poás volcano (Costa Rica). Samples were taken from its hyperacid lake (Laguna Caliente) and from a persistently active fumarole field on the southern shore. The lake data cover a period of almost three decades (1985–2012) when marked changes in activity and physico-chemical properties were recorded (Rowe et al., 1992a, 1992b; Martínez et al., 2000; Martínez, 2008; Rouwet et al., 2016; and references therein). The fumarole data represent a two-year interval (2010–2012) in which the temperatures ranged between 763 and 107 °C, and showed a declining trend. For a preceding period (2001–2007), Sharp et al. (2010) documented a strong $\delta^{37}\text{Cl}$ fractionation in other (low-temperature) fumaroles that were temporarily active within the crater, and attributed this to a distillation–recondensation process within the plumbing system. Fischer et al. (2015) presented data sets on the chemical and isotopic compositions of fumarolic discharges that cover both periods. Our results demonstrate that halogen signatures of Laguna Caliente and associated fumaroles signal active magma–gas–liquid interaction processes in the subsurface volcanic–hydrothermal system of Poás. The observed time series trends, in conjunction with other chemical parameters recorded at this volcano, provide insights into the potential of chlorine and bromine for monitoring purposes as an aid to mitigate hazards from eruptive activity.

2. Geological setting

Quaternary volcanism in Central America is associated with subduction of the Cocos plate under the Caribbean plate. The convergence offshore from Nicoya Peninsula in Costa Rica occurs at a rate of

74–85 mm/yr at 20–22° azimuth (DeMets et al., 2010). Poás, in the Cordillera Central of Costa Rica (Fig. 1), is a complex basaltic andesitic stratovolcano with a maximum elevation of 2708 m.a.s.l. Erupted products mainly consist of calc-alkaline basaltic and andesitic lavas and pyroclastics (Prosser and Carr, 1987; Cigolini et al., 1991; Malavassi, 1991). Its 1.3 km wide active crater hosts an acidic lake known as Laguna Caliente in a 300 m-diameter pit, and a ~30 m high pyroclastic cone, which is a site of persistent fumarolic activity (Fig. 2). The structure is often referred as “the dome” but since it is composed of tephra from the 1953–55 eruption (Casertano et al., 1987), we will use the term “composite pyroclastic cone” or “CPC”, following Martínez et al. (2000) and Martínez (2008). This 1953–55 eruption was the latest with juvenile materials. Such events are relatively rare at Poás, in contrast to the frequent occurrences of phreatic explosions.

Over the last decades, the water of Laguna Caliente has shown pH values ranging between –0.87 and 1.75, and temperatures from 22 to 94 °C (Martínez, 2008). Concentrations of sulphate (3300–285,000 mg/kg or 34–297 mmol/kg) and chloride (2500–15,000 mg/kg or 70–3244 mmol/kg) fluctuate but have been always extremely high, which also holds for dissolved rock-forming elements (Rowe and Brantley, 1993; Martínez, 2008). The activity of the crater area is characterized by a long history of intermittent phreatic eruptions and persistent fumarolic emissions. Martínez (2008) subdivided the activity of Poás since the early 1970's into five stages. During Stage I (1972–August 1980), fumarolic discharges were strong within the lake and were accompanied with occasional phreatic explosions. Stage II (September 1980–April 1986) was characterized by a relative quiescence in the lake and absence of phreatic activity despite a strong discharge of high-temperature fumaroles through the CPC. In 1981,



Fig. 1. Location map of Poás volcano.

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