



Reaction path models of magmatic gas scrubbing



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ABSTRACT

Gas–water–rock reactions taking place within volcano-hosted hydrothermal systems scrub reactive, water-soluble species (sulfur, halogens) from the magmatic gas phase, and as such play a major control on the composition of surface gas manifestations. A number of quantitative models of magmatic gas scrubbing have been proposed in the past, but no systematic comparison of model results with observations from natural systems has been carried out, to date. Here, we present the results of novel numerical simulations, in which we initialized models of hydrothermal gas–water–rock at conditions relevant to Icelandic volcanism. We focus on Iceland as an example of a “wet” volcanic region where scrubbing is widespread. Our simulations were performed (using the EQ3/6 software package) at shallow (temperature < 106 °C; low-T model runs) and deep hydrothermal reservoir (200–250 °C; high-T model runs) conditions. During the simulations, a high-temperature magmatic gas phase was added stepwise to an initial meteoric water, in the presence of a dissolving aquifer rock. At each step, the chemical compositions of coexisting aqueous solution and gas phase were returned by the model. The model-derived aqueous solutions have compositions that describe the maturation path of hydrothermal fluids, from immature, acidic Mg-rich waters, toward Na–Cl-rich mature hydrothermal brines. The modeled compositions are in fair agreement with measured compositions of natural thermal waters and reservoir fluids from Iceland. We additionally show that the composition of the model-generated gases is strongly temperature-dependent, and ranges from CO_{2(g)}-dominated (for temperatures ≤ 80 °C) to H₂O_(g)-dominated (and more H₂S_(g) rich) for temperatures > 100 °C. We find that this range of model gas compositions reproduces well the (H₂O–CO₂–S_{TOT}) compositional range of reservoir waters and surface gas emissions in Iceland. From this validation of the model in an extreme end-member environment of high scrubbing, we conclude that EQ3/6-based reaction path simulations offer a realistic representation of gas–water–rock interaction processes occurring underneath active magmatic–hydrothermal systems.

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1. Introduction

A recurrent, but sometimes overlooked, control factor on the chemistry of volcanic gas emissions is the interaction between magmatic gases and groundwater/hydrothermal systems. This process, commonly referred as *magmatic scrubbing* (Symonds et al., 2001), takes place as rising magmatic gases encounter any aquifer interposed between the source magma stored at depth and the surface. The resulting gas–water–rock interactions cause partitioning of water-soluble species (S, halogens) into aqueous solutions, and irreversibly modify the composition of the primary magmatic gas phase. Quantitative assessment of scrubbing is, therefore, essential for interpreting mechanisms and evolution of volcanic–hydrothermal unrests (Doukas and Gerlach, 1995; Gerlach et al., 2008;

Ilyinskaya et al., 2015; Symonds et al., 2001, 2003; Werner et al., 2008, 2012; Shinohara et al., 2015). The mechanism of magmatic gas scrubbing by hydrothermal systems was introduced in the fifties (White, 1957), but it was only in the 1990s that scrubbing was invoked as a most important process to explain the anomalous low fluxes of magmatic SO₂ and HCl observed at many volcanoes worldwide, both before and after eruptions (Doukas and Gerlach, 1995; Reed, 1997). Quantitative modeling of magmatic gas scrubbing started with Symonds et al. in 2001. By using the reaction path modeling approach and the CHILLER (Reed, 1982, 1998; Spycher and Reed, 1988) and GASWORKS (Symonds and Reed, 1993) programs, the Authors numerically investigated scrubbing at shallow- and deep-water conditions, by simulating the injection of increasing amounts of a magmatic gas (T = 915 °C) to aqueous solutions (ranging from diluted meteoric water and to hydrothermal solutions), in the 0.1–5 MPa pressure range. Results of these simulations highlighted the large impact of scrubbing on the chemistry (abundance of SO_{2(g)}, HCl_(g), HF_(g)) and, to a minor extent, CO_{2(g)} and H₂S_(g)) of volcanic gases. Symonds

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et al. (2001) also discussed the implications of scrubbing for volcano monitoring, and concluded that volcanic emissions of $\text{SO}_{2(g)}$ and $\text{HCl}_{(g)}$ are essentially reduced when scrubbing occurs. In such conditions, $\text{CO}_{2(g)}$ remains the most useful magmatic gas species to be monitored, until interactions between magmatic gas and hydrothermal aquifers are eluded by opening of a dry degassing pathway to the surface, shortly prior to or during a volcanic eruption. More recently, Marini and Gambardella (2005) first tested the ability of the EQ3/6 software package to model the irreversible gas mass exchanges occurring during addition of magmatic gas to pure water, at near-surface conditions (0.1 MPa). The obtained results were qualitatively similar to those of Symonds et al. (2001), confirming utility and flexibility of EQ3/6 for scrubbing calculations. More recent applications of EQ3/6 were presented in Di Napoli et al. (2013) and Ilyinskaya et al. (2015).

In spite of the major advance reached in past studies, application of thermodynamics models to scrubbing have not yet been validated using measured (natural) gas compositions. In this paper, we apply the reaction path modeling approach (Helgeson, 1968) with the specific objective of quantitatively investigating magmatic gas scrubbing at two volcanoes exhibiting extensive scrubbing: Hekla volcano and the Krýsuvík volcanic system, both in Iceland (Hk and Kr in Fig. 1). These case studies are here taken as archetypes of systems in which gas–water–rock interactions proceed in shallow and deep-reservoir hydrothermal environments, respectively. So doing, we provide more theoretical and observational confirmation to our initial attempts to model magmatic gas scrubbing at Icelandic volcanoes (Ilyinskaya et al., 2015) and, even more importantly, extend these to higher-temperature hydrothermal interactions. The large mass of previous work on hydrothermal systems has clearly demonstrated that compositions of surface hydrothermal manifestations are controlled by a variety of processes,

occurring at both deep reservoir conditions (e.g., fluid–mineral reactions; Arnórsson et al., 1983; Giggenbach, 1981, 1988; Reed and Spycher, 1984; Stefánsson and Arnórsson, 2000, 2002) and upon ascent of fluids from the reservoir to surface (e.g., boiling, degassing, mixing, oxidation and further water–rock interactions; Arnórsson, 1985; Arnórsson et al., 2007; Fournier, 1989; Kaasalainen and Stefánsson, 2012; Markússon and Stefánsson, 2011; Nordstrom et al., 2009). In comparison to these well characterised processes, the interaction mechanisms (scrubbing) of magmatic volatiles inside hydrothermal reservoirs have received less attention so far, and motivate the present study. We ultimately demonstrate that EQ3/6 scrubbing models satisfactorily reproduce the chemical compositions of cold (Hekla-type) natural gas emissions and, when combined with other processes such as boiling, of the near-boiling hydrothermal steam vents in Iceland. We also find good match between our model fluids and compositions of reservoir fluids in boreholes. Our developed model approach, after the validation study described here, can now generically be used to investigate scrubbing process at any volcano worldwide.

2. Materials and methods

2.1. EQ3/6 code

Reaction path models of magmatic gas scrubbing are here carried out by means of EQ3/6 (version 7.0; Daveler and Wolery, 1992; Wolery, 1979, 1992a,b; Wolery and Daveler, 1992), a software package combining together the EQ3NR and EQ6 codes (see Appendices A and B). By using thermodynamic and kinetic constraints, this software performs

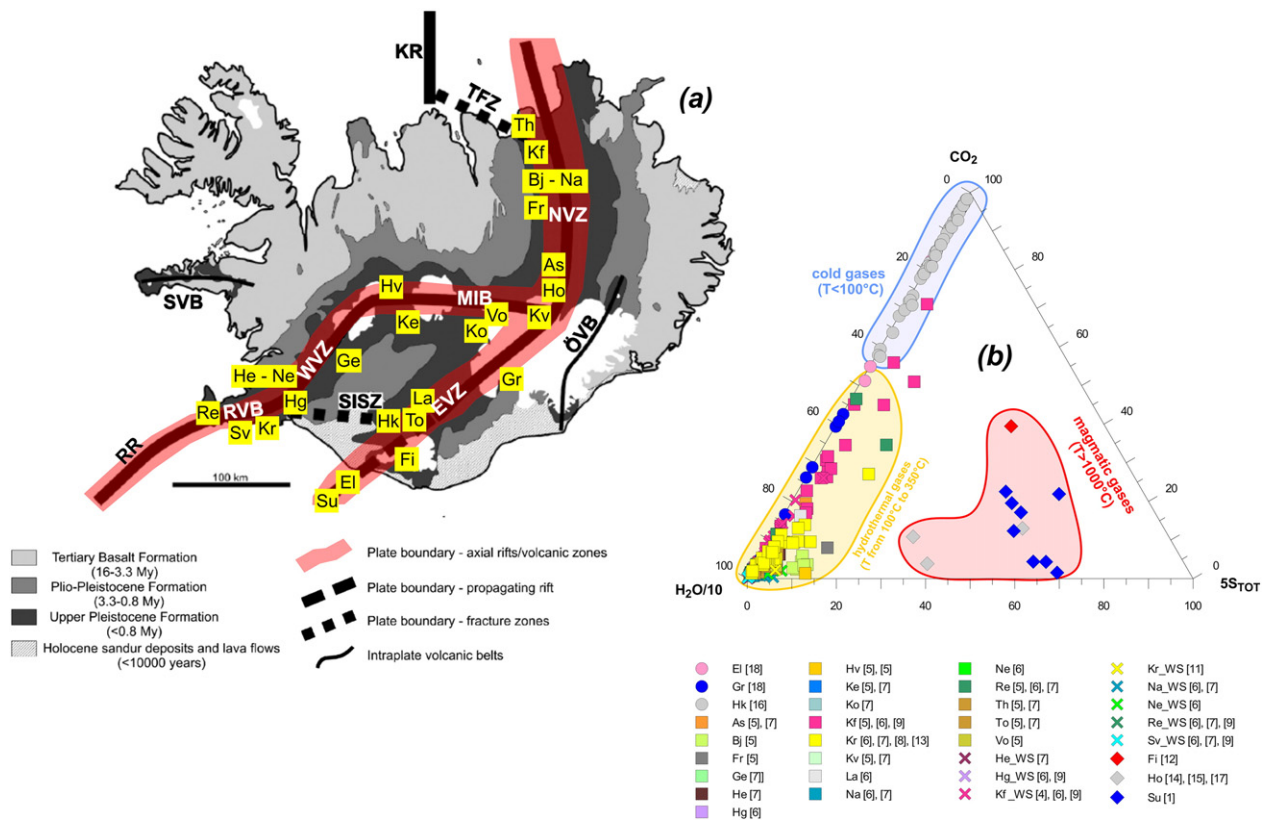


Fig. 1. (a) Simplified geological map of Iceland (modified from Thordarson and Larsen, 2007), showing the location of the main volcanic and geothermal systems on which the gas dataset (Table 3) is based upon. RR, Reykjanes Ridge; RVB, Reykjanes Volcanic Belt; SISZ, South Iceland Seismic Zone; Wvz, West Volcanic Zone; MIB, Mid-Iceland Belt; EVZ, East Volcanic Zone; NVZ, North Volcanic Zone; TFZ, Tjörnes Fracture Zone; KR, Kolbeinsey Ridge; ÖVB, Örafi Volcanic Belt; SVB, Snæfellsnes Volcanic Belt; (b) Triangular plot of $\text{H}_2\text{O}/10$ - CO_2 - 5S_{TOT} abundances in hydrothermal and magmatic gas samples from Iceland (for data source, see Table 3). S_{TOT} is total sulfur (e.g., $\text{H}_2\text{S} + \text{SO}_2$). Magmatic, hydrothermal and cold gas samples plot into compositionally distinct domains (see text for discussion).

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