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Tracing chlorine sources of thermal and mineral springs along and across the Cascade Range using halogen concentrations and chlorine isotope compositions



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ABSTRACT

In order to provide constraints on the sources of chlorine in spring waters associated with arc volcanism, the major/minor element concentrations and stable isotope compositions of chlorine, oxygen, and hydrogen were measured in 28 thermal and mineral springs along the Cascade Range in northwestern USA. Chloride concentrations in the springs range from 64 to 19,000 mg/L and δ^{37} Cl values range from +0.2% to +1.9% (average $= +1.0 \pm 0.4\%$), with no systematic variation along or across the arc, nor correlations with their presumed underlying basement lithologies. Additionally, nine geochemically wellcharacterized lavas from across the Mt. St. Helens/Mt. Adams region of the Cascade Range (Leeman et al., 2004, 2005) were analyzed for their halogen concentrations and Cl isotope compositions. In the arc lavas, Cl and Br concentrations from the volcanic front are higher than in lavas from the forearc and backarc. F and I concentrations progressively decrease from forearc to backarc, similar to the trend documented for B in most arcs. δ^{37} Cl values of the lavas range from -0.1 to +0.8% (average $= +0.4\pm0.3\%$). Our results suggest that the predominantly positive δ^{37} Cl values observed in the springs are consistent with water interaction with underlying ³⁷Cl-enriched basalt and/or altered oceanic crust, thereby making thermal spring waters a reasonable proxy for the Cl isotope compositions of associated volcanic rocks in the Cascades. However, waters with δ^{37} Cl values > +1.0% also suggest additional contributions of chlorine degassed from cooling magmas due to subsurface vapor-liquid HCl fractionation in which Cl is lost to the aqueous fluid phase and ³⁷Cl is concentrated in the ascending magmatic HCl vapor. Future work is necessary to better constrain Cl isotope behavior during volcanic degassing and fluid-rock interaction in order to improve volatile flux estimates through subduction zones.

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1. Introduction

Volatiles and fluid mobile elements (e.g., Cl, Br, B, F, S, Pb, As) are liberated from subducted oceanic slabs as a consequence of metamorphic dehydration reactions and returned to the Earth's surface (e.g., Bebout, 2007; Manning, 2004). Understanding the behavior of volatiles in subduction zones is important for quantifying global element fluxes. Cascadia is regarded as the warmest subduction zone (Syracuse et al., 2010) and thermal and petrologic models predict that the slab is strongly dehydrated before it reaches depths of magma genesis (e.g., van Keken et al., 2011).

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Seismic images suggest extensive serpentinization of the mantle wedge beneath the Cascadia forearc due to upward migration of slab-derived fluids (Bostock et al., 2002; Brocher et al., 2003).

Quantifying the concentrations of magmatic volatiles dissolved in groundwater and discharged through thermal springs allows for a more comprehensive estimate of the overall volatile fluxes through subduction zones (which rarely include volatile inventories in thermal springs), assessment of processes that control the abundance of volatiles liberated from the slab, and monitoring of volcanic activity due to fluctuations in shallow magmatic inputs. However, a major challenge with this approach is distinguishing magmatic versus non-magmatic volatile components in the springs. In the central Oregon Cascade Range, halogen concentrations and ratios of the radioisotopes ³⁶Cl and ¹²⁹I (Hurwitz et al., 2005), carbon isotopes (Evans et al., 2004; James et al., 1999),

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and helium isotopes (Evans et al., 2004) of thermal spring waters suggest that most of the halogens are magmatically derived. In contrast, Mariner et al. (2003) argue that Cl (and excess nitrogen) in central Cascade thermal springs are mainly sourced from underlying sedimentary units. It remains uncertain what relative chloride contributions are derived from [1] seawater/seaspray, [2] underlying sedimentary units (including connate water), [3] magma, and/or [4] Cl extracted from subsurface volcanic units via water-rock interactions.

Chloride, and to a lesser extent, Br, F, and I are often used as fluid tracers in subduction zones and for interpreting the extent and style of magma degassing (e.g., Fehn, 2012; Kendrick et al., 2012; Straub and Layne, 2003; Villemant and Boudon, 1999; Villemant et al., 2005). Stable Cl isotope ratios also have been used to trace sources through subduction zones (e.g., Barnes et al., 2008; Barnes et al., 2009; Chiaradia et al., 2014; Li et al., 2015). However, apart from the study of Li et al. (2015), all of these isotopic studies have focused on volcanic outputs in the form of gas and lava, and not addressed sources of Cl in thermal waters.

Reported δ^{37} Cl values of thermal waters are few and the available data cluster near 0%: Indonesian volcanic springs (-0.3 to +0.4%; Eggenkamp, 1994), Icelandic thermal waters (-0.3 to +0.3%; Barnes and Stefansson, 2012), Yellowstone springs (-0.1% to +0.4%; Zhang et al., 2004), Taupo Volcanic Zone (-0.8% to +0.7%; Bernal et al., 2014), and Soufrière in Guade-loupe and Montagne Pelée in Martinique in the Lesser Antilles (-0.65% to +0.12%; Li et al., 2015). None of these studies report halogen concentrations or Cl isotope compositions for the associated volcanic rocks, thus making it difficult to ascertain the extent to which the host rocks may control the chemistries of the thermal springs.

Here we report chemical and stable chlorine isotope compositions of waters from 28 thermal (\geq 8.3 °C above the mean ambient air temperature) and/or mineral springs (containing \geq 500 mg/L total dissolved solids (TDS)) in the Cascade Range (Fig. 1). Water sample sites are distributed over 900 km along strike, and cover the forearc, volcanic front, and backarc regions. We also report halogen concentrations and the Cl isotope composition of nine well-characterized lavas (8 basalts and 1 dacite) from the Columbia Transect in southwest Washington (Fig. 1) (Leeman et al., 2004, 2005). The new data are used to assess the sources, spatial variability, and isotopic composition of chloride in the springs along and across the arc.

2. Geologic overview the of Cascade Range and associated mineral and thermal springs

The Cascade Range stretches approximately 1300 km from northern California to southern British Columbia, and is associated with the eastward subduction of the Juan de Fuca, Gorda, and Explorer plates beneath North America (Fig. 1). Near the active trench, the subducting plates are relatively young (2–28 my) (Wilson, 1988), resulting in a warm subduction zone compared to most other volcanic arcs (Leeman et al., 2005; Syracuse et al., 2010). The Cascade Range comprises volcanic deposits, mainly basalt to dacite in composition, of Eocene–Quaternary age with cumulate thickness up to several kilometers across the older western and more recent High Cascades sectors (Hildreth, 2007). The underlying basement includes marine shales, limestones and sandstones, Paleozoic accreted oceanic terranes of OIB- and MORB-like basalt, and tectonic slivers of serpentinized peridotites (Wells et al., 2014).

Many previous studies of thermal springs in the Cascade Range were largely motivated by the region's geothermal potential (e.g., Ingebritsen et al., 1989, 1991; Mariner et al., 1990). Recent work has focused on Cl flux, and its source(s), because Cl discharge is



Fig. 1. Map of isotopically distinguishable terranes along the Cascade volcanic arc as outlined by Schmidt et al. (2008). Dashed contours show approximate depths to the slab in km (McCrory et al., 2004). Dashed lines on oceanic plates are magnetic anomalies (Wilson, 1988). Major volcanoes are illustrated as red triangles: (LA) Lassen, (MS) Mt. Shasta, (ML) Medicine Lake, (MMc) Mt. McLaughlin, (CL) Crater Lake, (NV) Newberry Volcano, (TS) Three Sisters, (MJ) Mt. Jefferson, (MH) Mt. Hood, (SH) Mt. St. Helens, (MA) Mt. Adams, (SM) Simcoe, (MR) Mt. Rainer, (GP) Glacier Peak, (MB) Mt. Baker, (MG), Mt. Garibaldi, (MC). Mt. Cayley, (MM) Mt. Meager. Springs sampled as part of this study are labeled and indicated by blue circles. Columbia Transect lavas are indicted by yellow stars (Group 2 basalts), black stars (Group 1 basalts) and green star (high crustal melt dacite) in SW Washington (Leeman et al., 2004, 2005). Modified from Schmidt et al. (2008). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

used as a proxy for heat, and thus magmatic activity (e.g., Evans et al., 2004; Hurwitz et al., 2005). Cascade Range thermal waters are predominantly meteoric in origin, largely Na–Cl/Ca–Na–Cl type, and have a large range in temperature, discharge rate, and composition irrespective of their regional geographic locations (e.g., Ingebritsen et al., 1991).

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