Contents lists available at ScienceDirect

Chemical Geology

journal homepage: www.elsevier.com/locate/chemgeo

The origin of solutes within the groundwaters of a high Andean aquifer

Clinton Rissmann^a, Matthew Leybourne^{b,*}, Chris Benn^c, Bruce Christenson^d

^a Environment Southland, Invercargill, New Zealand

^b Department of Earth Sciences and Mineral Exploration Research Centre, Laurentian University, 935 Ramsey Lake Rd, Sudbury, Ontario P3E 2C6, Canada

^c Gold Fields Exploration, Vancouver, BC, Canada

^d GNS Science, Lower Hutt, New Zealand

ARTICLE INFO

Article history: Received 17 September 2013 Received in revised form 12 November 2014 Accepted 15 November 2014 Available online 8 January 2015

Editor: Michael E. Böttcher

Keywords: Atacama Desert Hyperarid Groundwater geochemistry Stable isotopes Hydrothermal Brines

ABSTRACT

This paper investigates the origin of solutes within the groundwaters of the Monturaqui-Negrillar-Tilopozo (MNT) aquifer system within the high Andes of the Atacama Desert that discharges into the Salar de Atacama. Key questions include the relative significance of volcanic hydrothermal processes and evaporitic brine recycling over solute supply as well as the pathways of solute ingress to the MNT aquifer system. Groundwaters were analysed for elemental (major, minor and trace) and isotopic ($\delta^{18}O/\delta^2H$; $\delta^{13}C$ -DIC; $\delta^{34}S$ -SO₄; $^{87}Sr/^{86}Sr$) constituents to which various hydrochemical and multivariate statistical methods have been applied. Groundwaters are all classified as thermal and show increasing temperatures (27-35 °C) and concentrations of HCO₃ (4.4-10.4 mmol L^{-1} dissolved inorganic carbon [DIC]) with increasing proximity to Volcano Socompa resulting from an increasing mass flux of steam and magmatic CO_2 ($pCO_2 = 0.016$ to 0.10 atm; $\delta^{13}C-CO_2 = -9.3$ to -3.6% (V-PDB)) boiled off a deep hydrothermal reservoir. Superimposed upon this gradational and relatively smooth spatial increase in heat and mass flow is a sharp, structurally controlled, increase in TDS (826-3632 mg l^{-1}) and a concomitant change in δ^{34} S–SO₄ (+0.79 to 4.9‰ (V-CDT)) and ⁸⁷Sr/⁸⁶Sr values (0.707375-0.706859) associated with the inflow of evaporitic solutes. Evaporitic inputs are chemically and isotopically distinct from localised secondary hydrothermally derived solutes with major, minor and trace element data suggesting an origin within a highly oxidising, alkaline, evaporitic lake receiving dilute inflows enriched in volcanic/fumarolic sulfur mineralisation probably from volcanoes Socompa, Salín or Pular that delimit the eastern topographic extent of the aquifer system. The conceptual model presented in this paper proposes that basal leakage of evaporitic brines from active salar(s), within the high altiplano/volcanic arc, are actively entrained by sub-regional groundwater flow and conveyed to the MNT aquifer system where they mix with solutes derived from localised secondary hydrothermal gas-water-rock interaction. This work provides detail on the origin and processes controlling the solute composition of groundwater inflows to the Salar de Atacama within the volcanically active and hyperarid Atacama Desert and may be of significance to conceptual models of evaporitic brine evolution, recycling of evaporitic brines and hydrothermalism in arid regions.

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1. Introduction

Historically, deciphering the origin and processes controlling the supply of solutes to the meteoric and volcanic-hydrothermal waters of the hyperarid Atacama Desert has been problematic. The main difficult-ly arises from the ubiquity of highly soluble evaporitic salts associated with active salt lakes (Risacher et al., 2003; Boschetti et al., 2007; Risacher and Fritz, 2008; Risacher et al., 2011) and massive volumes (~10,000 km³) of anhydrite buried beneath the current day volcanic arc (Pueyo et al., 2001) both of which may be recycled through the hydrological cycle (Risacher et al., 2003; Banks et al., 2004) or via active volcanism either at the high temperatures of magma emplacement or

* Corresponding author. Tel.: +1 705 675 1151x2263.

E-mail address: mleybourne@laurentian.ca (M. Leybourne).

during convective dissolution or entrainment by primary hydrothermal fluids (Youngman, 1984; Risacher and Alonso, 2001; Tassi et al., 2010). Further ambiguity arises through having to account for solutes derived from low temperature meteoric gas-water-rock processes including leaching of massive sulfide emplacements or volcanic/fumarolic sulfur mineralisation, and mixing with basinal brines or metamorphic fluids (Alpers and Whittemore, 1990; Banks et al., 2004; Cameron and Leybourne, 2005; Leybourne and Cameron, 2006). However, Risacher et al. (2003), Risacher and Fritz (2008) and Risacher et al. (2011) have demonstrated that 75–90% of the dissolved solute load within inflow waters, including thermal springs, supplying evaporitic lakes originate from the recycling of evaporitic brines.

This study seeks to decipher the origin and processes governing the solute composition of groundwaters within a high Andean aquifer that ultimately discharges to the Salar de Atacama. Given the potential for







multiple solute sources, we have used a multi-element, multi-isotope and multivariate statistical approach to assess and constrain the source of solutes within the groundwaters of the MNT aquifer system.

2. Geology and hydrogeology

2.1. Climate

The Atacama Desert is considered the driest place on earth and has been arid to hyperarid since the early Oligocene (~45 Ma; e.g., Alpers and Brimhall, 1988; Arancibia et al., 2006; Clarke, 2006), although the timing of the onset of hyperaridity is debated (Hartley and Chong, 2002; Dunai et al., 2005; Hartley and Rice, 2005; Rech et al., 2006; Reich et al., 2009). Mean annual precipitation (MAP) ranges from <1 mm yr⁻¹ within the central desert (Pampa del Tamarugal) to ~150–200 mm yr⁻¹ along the volcanic front (>4500 m a.s.l. [above sea level]) (Magaritz et al., 1989, 1990; Drees et al., 2006). In all regions, potential evaporation rates greatly exceed MAP, varying from >1500 mm yr⁻¹ in the central desert to 600–1200 mm yr⁻¹ along the volcanic front (Risacher et al., 2003).

For the Monturaqui–Negrillar–Tilopozo area, MAP ranges from ~40 mm yr⁻¹ at 3100 m a.s.l. to ~100 mm yr⁻¹ at 4000 m a.s.l (Quade et al., 2007). Therefore, precipitation events of sufficient magnitude to overcome evaporative thresholds and result in recharge are typically restricted to the current day volcanic arc and eastern altiplano. The majority of this precipitation is derived from intense (\geq 100 mm), long return storm events (~5 year cyclicity; Drees et al., 2006) associated with the South American Summer Monsoon (SASM) that traverse the South Central Andes and bring precipitation to the western altiplano and volcanic arc (Miller, 1976; Aravena et al., 1999). Impounded behind the volcanic arc ephemeral streams recharged during SASM storm events flow into endorheic basins where a number of variably sized (~20 km² to 1600 km²) saline lakes occur.

2.2. Geology

2.2.1. Evolution of MNT trough and aquifer system

The Monturaqui–Negrillar–Tilopozo (MNT) trough is a 60 km long N–S oriented depression that formed contemporaneously with the Miocene–Holocene eruption of the present magmatic arc (or Western Cordillera) (Fig. 1; Bock et al., 2000). The stratigraphy and hydrostratigraphy of the MNT aquifer system is summarised in Table 1. The eastern extent of the MNT trough is defined by young (Pleistocene) Andean type stratovolcanoes of the current day volcanic arc whereas the western extent is controlled by the N–S trending Late Proterozoic Arequipa–Anotofalla basement anticlines of the Sierra Almeida, Sierra Agua Colorada and the Cordon de Lila ranges. The trough is divided into the southern Monturaqui Basin, the central Negrillar and the northern most Tilopozo zones (Wadge et al., 1995; van Wyk de Vries et al., 2001), where the Domeyko Range becomes the western most margin of the Preandean depression.

Emplacement of the current volcanic arc was followed by an ignimbrite flare-up in the Late Miocene–Pliocene, during which the MNT trough was filled with 200–300 m of volcanic ash and ignimbrite, forming the Salín Formation, the principal aquifer of the structural trough. The Salín Formation extends eastward behind the modern day volcanic arc where it outcrops at higher elevations across the altiplano, forming an extensive permeable ignimbrite sheet (Wadge et al., 1995; van Wyk de Vries et al., 2001; Anderson et al., 2002).

2.2.2. Salín Formation

The Salín Formation aquifer is characterised by a poorly consolidated mix of volcaniclastic sediments that are predominantly of dacitic composition, ranging from fine to medium grained sands, significant ash deposits, local discontinuous gravel horizons, ignimbrite sequences, and conglomerates (van Wyk de Vries et al., 2001; Anderson et al., 2002). Gravels and conglomerates are supported within a matrix of volcanic



Fig. 1. Location of Socompa. (a) Location of MNT aquifer system in South America; (b) hydrogeological setting of MNT aquifer system. The extent of the MNT aquifer system is depicted by the dotted line and includes the Monturaqui Valley, the Negrillar Volcanics, and the Tilopozo sections of the aquifer system. The Tilopozo Wetland constitutes the discharge point for groundwater from the MNT aquifer into the Salar de Atacama. Stars identify volcanoes Socompa (6051 m), Salín, Pular and Aracar within the current volcanic arc. Salars are impounded behind and within the volcanic arc and altiplano – small salars not shown. Production wells are shown as coloured circles, with colours denoting water type as per the classification of Section 4.3.

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