



Assessment of land subsidence mechanisms triggered by dolomitic marble dissolution from hydrogeochemistry and stable isotopes of spring waters



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ARTICLE INFO

Article history:

Available online 6 April 2015

Editorial handling by M. Kersten

ABSTRACT

This study presents geochemical results of springs draining a small humid tropical watershed composed of geologically dolomitic marbles in Central Sri Lanka. Water samples were investigated for their major ion chemistry, water stable isotope composition ($\delta^2\text{H}_{\text{H}_2\text{O}}$ and $\delta^{18}\text{O}_{\text{H}_2\text{O}}$) and isotope composition of dissolved inorganic carbon ($\delta^{13}\text{C}_{\text{DIC}}$). From major ion chemistry Ca^{2+} and Mg^{2+} were the dominant cations and were balanced mostly by HCO_3^- . All collected spring water samples scattered around the local meteoric water line with values from -31.9‰ to -46.8‰ for $\delta^2\text{H}_{\text{H}_2\text{O}}$ and from -5.5‰ to -7.4‰ for $\delta^{18}\text{O}_{\text{H}_2\text{O}}$ against VSMOW. This indicates local groundwater recharge pathways by regional precipitation rather than water from deeper aquifer systems. Concentrations of dissolved inorganic carbon (DIC) ranged from 0.91 to 9.38 mM L^{-1} and $\delta^{13}\text{C}_{\text{DIC}}$ ranged from -22‰ to -14‰ against VPDB with an average of -16‰ . Samples of spring water from carbonate rocks had increased DIC and $\delta^{13}\text{C}_{\text{DIC}}$ together with elevated pH values. Combined $\delta^{13}\text{C}_{\text{DIC}}$ and Ca^{2+} and Mg^{2+} contents suggest that groundwater evolution was dominated by dissolution of dolomitic marble. This unexpected weathering process was favored by intense rain and high ambient temperatures and excessive CO_2 production in tropical soils. The intense weathering resulted in karst structures with high hydraulic conductivities that rendered the terrain tectonically less stable.

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1. Introduction

Environmental isotopes of hydrogen, carbon and oxygen are important geochemical tools and were extensively used to investigate carbon sources and its biogeochemical turnover in hydrogeological systems (Cartwright et al., 2012; Clark and Fritz, 1997; Kanduč et al., 2012; van Geldern et al., 2015). For instance, hydrogeochemistry and isotope compositions of water ($\delta^{18}\text{O}_{\text{H}_2\text{O}}$ and $\delta^2\text{H}_{\text{H}_2\text{O}}$) and carbon ($\delta^{13}\text{C}_{\text{DIC}}$) were applied to understand flow patterns of groundwater (Cartwright et al., 2012; Mayo and Loucks, 1995), water rock interactions (Kanduč et al., 2014, 2012), moisture distribution in the vadose zones (Aggarwal et al., 2004; Araguás-Araguás et al., 2000; Murad and Krishnamurthy, 2008; Yamanaka, 2012), processes in hot springs (Chandrajith et al., 2013) and carbonate weathering (Assayag et al., 2009; Atekwana and Krishnamurthy, 1998; Ayalon et al., 1998; Barth et al., 2003; Cerling et al., 1991; Falcone et al., 2008; Yamanaka, 2012; Zaihua et al., 1997).

One of the key parameters of these studies is dissolved inorganic carbon (DIC) that consists of H_2CO_3^* (i.e. the sum of H_2CO_3 and CO_2), HCO_3^- and CO_3^{2-} (Drever, 1997). The distribution of these species depends largely on pH and results from various processes including biogeochemical reactions as respiration, diffusion and gas exchange, as well as the precipitation or dissolution of carbonate minerals (Assayag et al., 2009). Both, contents of DIC and its isotope composition can help to outline carbon sources from open and closed systems. They can also differentiate between kinetic mechanisms of calcite dissolution and deposition (Falcone et al., 2008; Murad and Krishnamurthy, 2008; Yoshimura et al., 2001; Zaihua et al., 1997). Moreover, $\delta^{13}\text{C}_{\text{DIC}}$ can also serve as a tracer to investigate underground water movements in karst systems (Emblanch et al., 2003; Gonfiantini and Zuppi, 2003; Kanduč et al., 2012). In all studies a general trend is that solubility of carbonates and dolomites increase with the availability of CO_2 . Concentrations of CO_2 in soil air are significantly higher than in the atmosphere and typically range between ~ 3000 and $30,000 \mu\text{atm}$ (Sposito, 1989; van Geen et al., 1994). Thus, the $p\text{CO}_2$ of groundwater is also often enriched with respect to atmospheric levels. For temperate karst waters at an Irish cave Tooth and Fairchild (2003) reported a median

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value of 8000 μatm while van Geldern et al. (2015) calculated values of up to 21,400 μatm for a karst spring in southern Germany.

In addition, isotope exchanges and fractionation kinetics in carbonate systems depend on available rock surfaces that are exposed to the groundwater (Confiantini and Zuppi, 2003). Usually, carbonate weathering is more intense when compared to that of silicate weathering because of faster dissolution kinetics (Liu and Dreybrod, 1997; Liu et al., 2011; Plummer et al., 1978; Quade et al., 2003; Roy et al., 1999; Ryu et al., 2008). Details on mineral dissolution and related kinetics are given in Brantley et al. (2008). Most of these previous studies were carried out in sedimentary limestone terrains. Such limestones often hold numerous cracks, joints, cleavages, and bedding planes for easy water infiltration and thus offer pronounced karstic erosion pathways and dissolution features. In contrast to sedimentary limestones, metamorphic limestones or marble are more difficult to weather due to their crystalline nature and limited or often non-existent fracture spaces and their resulting overall lower permeability. Therefore, weathering in metamorphic carbonates mostly occurs along structurally weak zones (Winkler, 1987).

During the last few years, the Matale region in Sri Lanka became a terrain with pronounced needs for hydrogeological studies because it showed cracking of buildings that were presumably associated with land subsidence. In addition, changes of spring water levels as well as complete drying of wells together with spontaneous formation of new springs along fissures were observed (Naalir, 2013). Unexpectedly, most of these incidences were particularly pronounced along bands of crystalline carbonate rocks that consist to a large part of dolomitic marbles (Nikagolla et al., 2012). These incidences may link to yet poorly characterized underground erosion and subsequent collapses of subsurface cavities that were also shown by geophysical investigations of the area (Nawagamuwa et al., 2010). In this investigation we aimed at understanding geochemical processes of a metamorphosed karst aquifer in the Matale region in Sri Lanka in order to find potential reasons for the observed incidents. The study involved geochemical and environmental isotope compositions of springs in a small headwater catchment that is underlain by dolomitic marble and also investigated tropical weathering scenarios of metamorphosed dolomitic rocks.

1.1. Description of the study area

Crystalline rocks without any primary porosity cover over 90% of the landmass of Sri Lanka. The occurrence of groundwater in the hard rock terrain is mainly associated with openings formed as a result of secondary porosity developed along structurally weak zones such as foliations, fractures and joint systems. Such features can provide reasonable aquifer characteristics in the local rock formations (Jayasena et al., 2008). The study area (Fig. 1) belongs to the intermediate climate zone of Sri Lanka and is characterized by a wet-humid climate with average annual precipitation of nearly 1500 mm. The region is known for the occurrence of sparsely distributed natural perennial springs. They mostly occur in foot slopes of ridges that are underlain by dolomitic marble. Monthly average air temperatures of the area vary from 27 °C to 30 °C while the annual evapotranspiration varies from 750 mm to 1200 mm. This often causes perennial stream flow in the study area with significant discharges from springs that also trigger elevated inter- and base flow. The topographic elevation in the region varies from 200 m to 1300 m above mean sea level and the area thus exposes deep valleys with steep to moderate hill slopes. The lithology of the region predominantly comprises high-grade Precambrian metamorphic rocks that belong to the geological unit of the Highland Complex (Fig. 1). The rocks in the study area consist mainly of dolomitic marbles that are inter-bedded with various

forms of gneiss and quartzite. Three major marble bands form a valley in north–south direction.

The main surface drainage in the study area is controlled by the river ‘Suduganga’ that traverses the study area from South to North. The aquifers of the region are mainly linked to deep fracture zones in the metamorphic basement. The NW–SE trending fracture patterns are dominant in the southern part, while E–W trending fractures are more common in the northern part of the study region. These fractures offer the major directions of groundwater flow because these bed-rocks do not possess any primary porosity. Therefore, local groundwater flows follow tectonic elements including joints, fractures, faults or fissures and solution cavities formed due to dissolution of carbonate rocks. Resulting karst features appear mostly as surface “pot holes” close to the locations of springs and might represent interconnected but yet poorly characterized cave systems that may result from extensive dissolution of marble.

Metamorphosed marbles are widespread in the Highland Complex of Sri Lanka and mostly formed under temperatures between 850 and 900 °C and pressures between 8 and 10 kbar (Kriegsman and Schumacher, 1999; Osanai et al., 2006). As a result, these marbles are crystalline. Locally, these rocks are also marked by the presence of other mineral phases such as dolomite, olivine, diopside and phlogopite (Osanai et al., 2000). With this the metamorphic limestone in the study region of Matale are dolomitic and their chemical analysis showed 35% of CaO, 32% of MgO and 25% of SiO₂ (Nawaratne and Liyanage, 2014). Such rocks would usually undergo little chemical weathering when compared to weathering rates of the sedimentary limestone due to their massive nature that does not permit primary porosity. Springs and seeps in the study region appear exclusively in the marble band in the N–S trending valley. The discharges of these springs vary, but are usually over 100 L min⁻¹.

2. Materials and methods

A total of 24 samples were collected with 21 of them from natural springs, two from deep wells (>30 m) and one from the Suduganga River. Deep well water samples were collected using a hand pump that was operated in laminar flow for 10 min before sampling. All samples were measured for the parameters listed in Table 1. Field parameters including pH, electrical conductivity (EC) and temperature were also recorded at the time of sampling using portable probes (Hach SensION MM150, Hach Company, Loveland, CO, U.S.A.). Water samples were filtered on site into two HDPE bottles with disposable syringes through nylon membrane filters with a pore size of 0.45 μm . One HDPE bottles for cation analyses was acidified with 1% v/v conc. HNO₃ and another was kept non-acidified for anion analyses. Cations (Na⁺, K⁺, Ca²⁺, Mg²⁺, Fe²⁺) and anions (HCO₃⁻, Cl⁻, F⁻, NO₂⁻, NO₃⁻ and SO₄²⁻) analyses were performed with an Ion Chromatograph Dionex ICS-2000 (Thermo Dionex, Sunnyvale, CA, U.S.A.). Analytical reproducibility was within 5% ($\pm 1\sigma$) for Na⁺, K⁺, Ca²⁺, Fe²⁺, F⁻, within 10% ($\pm 1\sigma$) for Mg²⁺, Ca²⁺, NO₂⁻, NO₃⁻, SO₄²⁻ and within 16% ($\pm 1\sigma$) for Cl⁻ and PO₄³⁻. However NO₂⁻ and PO₄³⁻ contents were less than 0.1 and 0.02 mg L⁻¹, respectively for all samples and hence are not discussed further. All other cations reported in Table 1 were determined by ICP-MS (Thermo X-Series 2, Thermo Scientific, Bremen, Germany).

Measurements of $\delta^{18}\text{O}_{\text{H}_2\text{O}}$ and $\delta^2\text{H}_{\text{H}_2\text{O}}$ in water samples were carried out by wavelength scanned cavity ring-down infrared spectroscopy on a Picarro L1102-i (Picarro Inc., Santa Clara, CA, USA) instrument that was coupled with a vaporization module. Isotope results are reported in the standard delta (δ) notation versus the Vienna Standard Mean Ocean Water (V-SMOW) standard and expressed in per mil (‰) according to Eq. (1).

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