

Origin of calcium sulfate-type water in the Triassic carbonate thermal water system in Chongqing, China: A chemical and isotopic reconnaissance

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ABSTRACT

Chongqing, located in the southwestern China, was named as “A spa city of the world”. Its most geothermal reservoirs are developed in carbonate rock aquifers, and those thermal waters are characterized by high concentrations of sulfate and low HCO_3^- . However, little was known about the origin and genesis of those thermal waters, which is necessary for their protection and determination of regime of exploitation. Therefore, thermal and surface waters were sampled seasonally and analyzed for hydrogeochemical and isotopic compositions ($\delta^{18}\text{O}/\delta^2\text{H}$; $\delta^{34}\text{S}\text{-SO}_4$; $\delta^{18}\text{O}\text{-SO}_4$) to decipher the origin of the thermal waters in the Triassic carbonate aquifers in Chongqing area. Key questions include the relative significance of hydrothermal processes and solute supply. The results showed that: (1) the major chemical composition of analyzed the thermal waters from Chongqing area was characterized by Ca-SO_4 ; (2) the $\delta^2\text{H}$ and $\delta^{18}\text{O}$ values of the thermal waters ranged from -48.6‰ to -63.1‰ with an average value of -54.2‰ , and from -6.5‰ to -9.2‰ with an average value of -8.0‰ , respectively, indicating that the thermal waters originated from the local rain water with an elevation of 415 m to 1453 m above mean sea level; (3) the estimated geothermal temperatures varied from 63.8 °C to 78.3 °C (Quartz), indicating that the depth of the geothermal reservoir varied from 411 m to 1728 m, which is located in the Lower Triassic Jialingjiang formation; (4) the $\delta^{34}\text{S}\text{-SO}_4^{2-}$ and $\delta^{18}\text{O}\text{-SO}_4^{2-}$ in the geothermal waters ranged from 29.7‰ to 34.1‰ with a mean value of 32.1‰ , and from 12.5‰ to 16.5‰ with a mean value of 15.2‰ , respectively, suggesting that the high SO_4^{2-} concentrations resulted mainly from the dissolution of gypsum in the second part of the Lower Triassic Jialingjiang Formation; (5) three processes of water-rock interactions (gypsum dissolution, carbonate dissolution by carbonic acid and carbonate dissolution by sulfuric acid) in the thermal system were unveiled by the hydrogeochemical and isotopic models. Among these three processes, gypsum dissolution seems to be primarily responsible for solute composition of the thermal water. This study also indicates the integration of hydrogeochemical and isotopic data is very useful tool to decipher the origin and genesis of the carbonate thermal waters.

1. Introduction

The carbonate thermal waters, which are characterized by Ca-sulfate, have been well known because they are used for curative purpose since the Middle Ages in the world (Boschetti et al., 2005; Capecchiacci et al., 2015). Meanwhile, the large changes of $\delta^{34}\text{S}$ in carbonate-associated sulfate (CAS) in limestones over geological time have been found (Claypool et al., 1980; Kah et al., 2001, 2004; Bottrell and Newton, 2006), especially at the Permian–Triassic boundary. The extreme changes of $\delta^{34}\text{S}$ occurring over very short geological time scales have long attracted special interest in the global sulfur cycle.

Chongqing City, the only municipality in southwestern China, holds rich shallow geothermal resources, which are mostly distributed in the core area of a series of eastern Sichuan parallel anticline developed in

the Triassic–Jurassic strata. Because the exploitation of the shallow geothermal resources is mainly for the spa leisure tourism, Chongqing was named as “A spa city of China” in 2011 and “A spa city of the world” in 2012. Some studies have been focused on the geochemical characteristics of the thermal waters (Cao, 2007; Yang et al., 2017), and results indicated that most thermal waters are found from the carbonate aquifers and show very high concentrations of SO_4^{2-} (more than 1000 mg/l) and low HCO_3^- values (less than 200 mg/l) in Chongqing area, which are very similar to other Triassic carbonate thermal waters in the world (Boschetti et al., 2005; Capecchiacci et al., 2015). Nevertheless, little was known about the origin and genesis of thermal waters, especially on the dominant sources of sulfur of thermal waters in Chongqing area, which is necessary for their protection and determination of regime of exploitation.

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A wide range of approaches are used to study the origin and genesis of thermal waters, including the applications of structural, hydraulic, hydrogeochemical and isotopic methods, and the investigation of fluid inclusions (González et al., 2000; Rissmann et al., 2015). Sulfate is a ubiquitous component in thermal waters, and is derived from a variety of sources such as precipitation and oxidation of sulfide minerals in the strata and dissolution of sulfate evaporates. Because of large S isotope variation exists in Earth's rocks (~80–100‰) with small S isotope fractionations (~1–2‰) during the oxidation/dissolution of S-bearing minerals (Clark and Fritz, 1997; Szykiewicz et al., 2015), the $\delta^{34}\text{S}\text{-SO}_4$ can precisely pinpoint major sources of S participating in water-rock interaction. Thus, in the past decades, $\delta^{34}\text{S}\text{-SO}_4^{2-}$ has been widely used to trace the sources of sulfate in geothermal waters (Ohmoto, 1986; Martinez, 1993; Jennifer et al., 2000; Kusakabe et al., 2000; Çelmen and Çelik, 2009; Fatma et al., 2011; Peters et al., 2011; Risacher et al., 2011; Dupalová et al., 2012; Loges et al., 2012; McDermott et al., 2015; Rissmann et al., 2015), and formation characteristics of gypsiferous units (Orti et al., 2002; Palmer et al., 2004; Çelmen, 2008). Meanwhile, some geothermal waters originated from the carbonates have been studied such as the hot springs in Hungary (Dublyansky, 1995) and in Italy (Boschetti et al., 2005; Capecchiacci et al., 2015) and the thermal power stations in Kazakhstan and Germany (Keller, 1991; Berge and Veal, 2005), and results indicated that the origin and genesis of the

thermal waters from carbonates are more complex than those from other thermal reservoirs.

Thus, seasonal variations in the concentrations of dissolved SO_4^{2-} and their isotopic compositions coupled with hydrochemical parameters, and $\delta^2\text{H}$ and $\delta^{18}\text{O}$ of the thermal and surface waters were used to determine the origin of the thermal waters, and discuss the potential SO_4^{2-} sources of the thermal waters derived from the carbonates in Chongqing area. The purpose of this study is that hydrogeochemical and isotopic data were integrated to decipher the origin of the carbonate thermal waters and trace quantitatively the sources of the solute compositions in these thermal waters. Three processes of water-rock interactions (gypsum dissolution, carbonate dissolution by carbonic acid and carbonate dissolution by sulfuric acid) in the carbonate thermal system were quantitatively unveiled by the hydrogeochemical and isotopic models, respectively. The results indicate that the gypsum dissolution rather than the carbonate dissolution by carbonic acid or sulfuric acid is the dominant control process for the solute compositions of the thermal waters in the study area.

2. Outline of study area

Chongqing municipality is located in the eastern Sichuan Basin, southwest China (Fig. 1), the transitional area between the Qinghai-Tibet

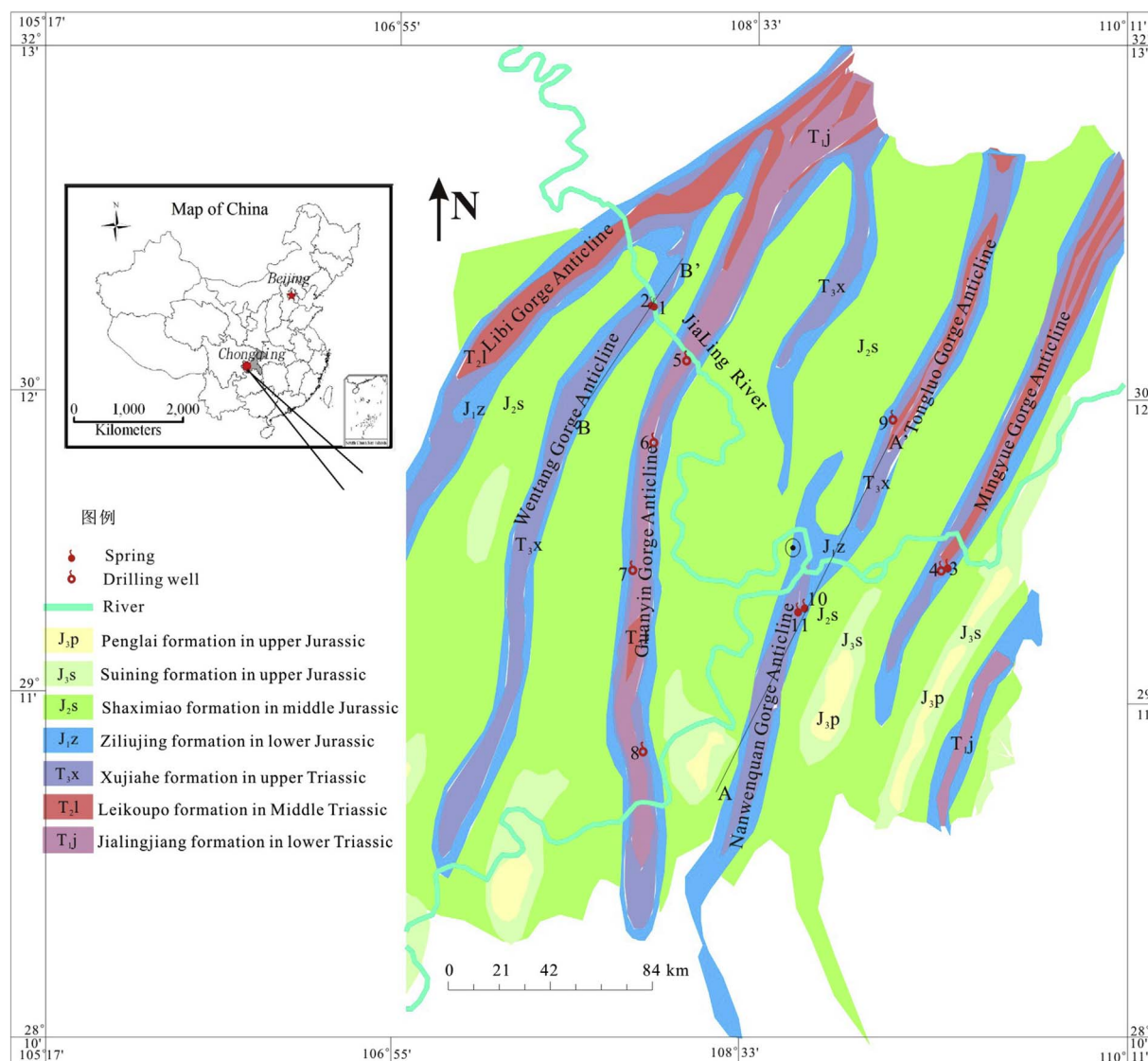


Fig. 1. Location, hydrogeological map and distribution of thermal water samples in Chongqing.

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