Journal of Volcanology and Geothermal Research xxx (2018) xxx–xxx

Contents lists available at ScienceDirect

Journal of Volcanology and Geothermal Research

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

Invited research article

Impact of additional dead carbon on the circulation estimation of thermal springs exposed from deep-seated faults in the Dongguan basin, southern China

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ARTICLE INFO

Article history:
Received 28 May 2018
Received in revised form 30 July 2018
Accepted 1 August 2018
Available online xxxx

Keywords:
^{14}C age
$^3He/^4He$
$\delta^{13}C$
Additional dead carbon
Thermal spring
Deep-seated fault

ABSTRACT

In hydrothermal systems, the dissolved carbon of thermal groundwater may contain components derived from mineral alterations under high temperatures (metamorphic source) and/or from magmatic gases, additionally from carbonate dissolution (most common). These components are known as additional dead carbon, which dilutes the modern carbon used for dating, leading to the overestimation of ^{14}C ages. Four typical geothermal springs hosted by deep-seated faults and eighteen non-thermal springs and wells from the Dongguan basin of southern China were investigated to assess the circulation times of the hydrothermal system based on their radiocarbon contents. For the thermal springs, the circulation depth (approximately 3648 m), the apparent ^{14}C ages (20.15 to 12.97 ka) and the $\delta^{13}C$-calibrated ^{14}C ages (16.02 to 8.55 ka) were estimated. The circulation speed of the thermal groundwater was as low as 0.46 m/a, which was too slow for a fracture hydrothermal system hosted by deep faults. The analysis of the helium and carbon isotopes in the thermal groundwater revealed the existence of mantle-derived carbon and thermally altered carbon from limestones. A $\delta^{13}C$-$^3He/^4He$ illustration was proposed to correct for the additional dead carbon content. According to the reassessed ^{14}C ages, the groundwater circulation time was overestimated and the circulation speed of the thermal groundwater was underestimated in hydrothermal system. The reassessed ^{14}C ages can provide a more reasonable result than the initial slow estimate in the context of a fracture hydrothermal system hosted by deep faults. We conclude that measured ^{14}C contents should be reassessed during the use of radiocarbon dating for thermal groundwater if additional dead carbon exists in the water.

1. Introduction

For the purpose of sustainable heat utilization as green energy, the circulation time significantly affects the renewal ability of thermal groundwater in geothermal systems and should be well known. Some traditional groundwater dating tools, such as radiocarbon and accumulated ^4He, have been well used to determine the age of thermal groundwaters (Mazor et al., 1988; Birkle et al., 2001; Mariner et al., 2006). Thermal groundwater is closely associated with various geothermal features and processes, such as active volcanoes (Fischer et al., 1997; Tassi et al., 2003; Ishikawa et al., 2007; Werner et al., 2008; Guido and Campbell, 2012), magma intrusion (Minissale et al., 2007), tectogenesis (Minissale et al., 2007; De Filippis and Billi, 2012) and thermal alteration for the decay of radioactive elements in minerals (Shanker et al., 1987; Hilton et al., 1993; van Middlesworth and Wood, 1998). In some cases, these geothermal processes can generate: (1) geogenic "dead carbon" from mineral alteration at high temperature and/or from magmatic outgassing (Sturchio et al., 1990; Hilton, 1996; Tedesco, 1996; Kavouridis et al., 1999; Yokoyama et al., 1999; Gherardi et al., 2005; Carreira et al., 2010; Thiebaud et al., 2010), which is called additional "dead carbon" and can lead to the overestimation of ^{14}C ages because the "modern carbon" used for dating is diluted; as well as (2) a certain amount of plutonic ^4He from deep faults and/or radiogenic ^4He from mineral radioactive decay (Poreda et al., 1992; Hulston and Lupton, 1996; van Soest et al., 1998; Hoke et al., 2000), which is called "excess gas" and can cause inevitable misinterpretation of accumulated ^4He for groundwater dating. Thus, it is necessary to consider the effects of these components in the process of thermal groundwater dating.

Radiocarbon dating is considered to be the most universal and reliable groundwater dating method (Castro et al., 2000; Kulongoski et al., 2008). Many corrections have been developed due to the diversity of carbon in groundwater and the complexity of water-rock interactions during groundwater transport (Han and Plummer, 2016). Ideally, the maximum reservoir temperatures, helium concentrations, and ^{14}C contents should be positively correlated with the groundwater circulation

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https://doi.org/10.1016/j.jvolgeores.2018.08.002

Please cite this article as: Mao, X., et al., Impact of additional dead carbon on the circulation estimation of thermal springs exposed from deep-seated faults in the Dongguan ..., J. Volcanol. Geotherm. Res. (2018), https://doi.org/10.1016/j.jvolgeores.2018.08.002

time in the hydrothermal system. This close relationship has been examined between geothermometer temperatures estimated with water chemistry, ^{4}He ages and ^{14}C ages in low-to-moderate-temperature geothermal systems in batholith areas (Mariner et al., 2006). Unfortunately, significant correlation among these values have rarely been observed in high temperature geothermal systems or hydrothermal systems hosted by deep-seated faults (Kling et al., 1989; Thompson et al., 1992; Werner et al., 2008), partly because the thermal groundwater is mostly recharged by meteoric water and heated quickly in a fracture system; and partly because the dead carbon or external helium from deep-seated faults is mixed with the groundwater. Various corrected models (e.g., the chemical mass-balance model, δ^{13}C mixing model, and stoichiometric balance of the dissolved carbonate species model) (Pearson and White, 1967; Fontes and Garnier, 1979; Clark and Fritz, 1997) have been successfully employed to discuss groundwater ^{14}C ages based on the consideration of some parameters, such as the percent modern carbon in the input water (Fontes and Garnier, 1979), the carbon isotopic composition of the recharge water (Carmi et al., 2009), and the isotopic composition of the carbonate minerals with which the water reacted (Kroitoru et al., 1989; Coetsiers and Walraevens, 2009). However, the additional dead carbon is rarely discussed and separately corrected when radiocarbon is used to estimate the ages of thermal groundwater.

There are several well-known, high-temperature thermal springs that are rich in inorganic CO_2 in Southwest China, such as the Dagunguo Spring of the Tengchong geothermal field and the Yangbajing Spring of the Tibet geothermal field, which are located in the collision zone of the Pacific and Indian plates and are closely related to crustal magma remelting or the solidification and cooling of intrusive bodies in the crust (Du et al., 2005; Yim et al., 2006; Lau et al., 2008; Guo and Wang, 2012; Peng and Jones, 2012). However, thousands of low-to-moderate-temperature thermal springs are distributed in the provinces of Guangdong, Jiangxi, Hunan, Fujian, Guangxi and Taiwan of southern China, discharging in the vicinity of granites due to a series of magmatic intrusions and deep-seated faults caused by tectonic activity in the subduction zone of the Pacific plate and Eurasian plate (Lin et al., 2010; Zhu et al., 2010; Liu et al., 2011). In addition, inorganic CO_2 representing additional dead carbon can be recognized in most of these low-to-moderate-temperature thermal springs. Their appearance is considered to be partly the result of cooling of the magma intrusions of southern China and partly as the result of mixing with deep geothermal fluids communicated by the deep-seated faults (Zhou et al., 2009).

Guangdong Province, located in southern China, has nearly 300 thermal springs distributed over the fault belts that are mostly associated with granite batholiths (Huang and Goff, 1986; Hochstein et al., 1990; Lin et al., 2007; Guo et al., 2012). The outlet water temperature of these thermal springs can reach up to 98 °C. In addition, the observed ^{3}He/^{4}He ratios in these thermal springs, which range from 1.1 to 4.9 Ra, indicate the occurrence of a mantle component dissolved in the thermal groundwater (Xu et al., 1997). In recent years, these geothermal springs have raised considerable interest in exploiting its potential to produce green energy. The circulation time of thermal groundwater is an important parameter, but it is rarely discussed for the hydrothermal systems hosted by deep-seated faults. The Dongguan basin is a typical sedimentary basin controlled by deep faults in Guangdong Province, for which the sedimentary layer thickness is over 2000 m. Thermal springs are exposed on the marginal fault zone of the basin, and the reservoir temperature estimated by quartz geothermometer after steam loss is 131.0 to 138.9 °C (Mao et al., 2015). The Sanshui Basin is adjacent to the Dongguan Basin and is very similar to the geological structure of the Dongguan Basin. A carbon dioxide gas reservoir is buried in the Sanshui Basin. Its ^{3}He/^{4}He (3.1–3.3 Ra, Ra is atmospheric ^{3}He/^{4}He $= 1.39 \times 10^{-6}$) and δ^{13}C (−4.7–−4.5‰) values suggest that the CO_2 is probably derived from magmatic and biogenic sources, with some contribution from the thermal alteration of crustal rocks (Xu et al., 1991; 1997; Dai et al., 2005). Thus, additional dead carbon from magmatic crystallization and crustal thermal alteration is most likely dissolved in the thermal springs. Our study aims to provide a reliable ^{14}C dating method for thermal springs to estimate the circulation times of hydrothermal systems hosted by deep-seated faults. δ^{13}C and ^{3}He/^{4}He can provide clues about the origin of the dissolved carbon and helium, respectively, which can be used to separate the additional dead carbon and reassess the radiocarbon content in thermal groundwater. We hope to demonstrate that the thermal groundwater circulation times estimated from ^{14}C values using the traditional method must be re-evaluated in hydrothermal systems hosted by deep-seated faults because of the presence of additional dead carbon.

2. Hydrogeologic setting

The Dongguan basin is located in the eastern part of the Pearl River Delta regions, Guangdong Province of southern China (Fig. 1). The terrain is tilted from the northeast to southwest. The landscape is dominated by hilly terraces and alluvial plains. The southwestern part, an estuarine sedimentary plain with relatively flat topography, is approximately 10 m above mean sea level (m.s.l.). The northeastern part, low mountains and hills, is approximately 100 m above m.s.l. According to the nearly 60 years of meteorological records from the Meteorological Station in Dongguan City, it has a subtropical monsoon climate with an average annual temperature of 23.3 °C. The local annual precipitation is about 1336 mm, while the local annual evaporation is as high as 1100 mm.

The basement of the Dongguan basin, in which a series of continuously deposited strata developed, is granite (Fig. 1). The Cambrian to Ordovician strata are mainly limestone. The Devonian-Permian strata are controlled by sandstones with intervening shale layers. The Triassic to Jurassic strata mainly consists of glutenite. The Cretaceous strata are tuffaceous conglomerates, sandstone and shale. The Paleogene and Neogene strata are tuffaceous sandstone interbedded with shale. The Quaternary strata are estuarine sediments, mainly composed of sand, gravel, clay sand and sandy clay. The Yanshanian intrusive rocks are widely distributed, and their main lithology is granodiorite, granite porphyry and granite.

The series of complicated tectonic activities that formed the main deep-seated faults, which are oriented in the northeast direction, was accompanied by continuous stratigraphic depositions that underwent frequent metamorphism and intense magmatism during three regional geological stages (Sinian-Silurian, Devonian-Mid Triassic, and Later Triassic-Present) (GBGMI, 1988). The two deep-seated faults in the studied area, F1 and F2, are the Heyuan Fault and Zijin-Boluo Fault, respectively, as shown in Fig. 1. Granite batholiths are the most widespread type of batholith outcrops, accounting for >40% of the bedrocks at Guangdong Province (Song et al., 2011). The Sanshui basin is adjacent to the southwest area of the Dongguan basin, and it is regarded as the reservoir of inorganic CO_2 associated with the deep faults and magmatic activity of southern China (Xu et al., 1997; Dai et al., 2005).

Four hydro-stratigraphic groups were recognized according to the different types of lithology and pore structures in the studied area. The coarse-medium sands and gravel of the Quaternary strata are rich in pore water and represent the first hydro-stratigraphic group, which are distributed in the delta plain, mountain basins and coastal plains (Xu et al., 1997). The local main hydro-stratigraphic group, which represents the second hydro-stratigraphic group, comprises red sandstones and siltstones of Devonian to Neogene strata with abundant fissure water. This group is widely distributed across the studied area, down to the 1500 m sediment thickness level in the Dongguan basin (Zhou et al., 2011). The limestone fissure water, which mainly exists in Cambrian to Ordovician strata and represents the third hydro-stratigraphic group, is distributed at the edge of the Dongguan basin (Yu et al., 2015). The last hydro-stratigraphic group is rich in fissure water and distributed at the fault zone of granite and tuff, which contain abundant groundwater with a non-uniform spatial distribution pattern (Chen