



Helium and carbon isotopes in the hot springs of Changbaishan Volcano, northeastern China: A material connection between Changbaishan Volcano and the west Pacific plate?

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ABSTRACT

Changbaishan Volcano is located in northeastern China, approximately 1400 km west of the west Pacific subduction zone. Although the west Pacific plate and Changbaishan Volcano are spatially associated with each other, no previous evidence has demonstrated the existence of a direct material connection between the two. In this study, we utilize helium (³He/⁴He, CO₂/³He) and carbon isotopes (δ¹³C) from the hot springs of Changbaishan Volcano to exclude the possibility of a direct material connection between the volcano and the west Pacific plate at source. A total of 22 gas samples were collected from three hot springs at Changbaishan Volcano in 2002, 2006, 2014 and 2015; isotopic and geochemical analyses were performed on these samples to trace the possible sources of these gases.

Our analysis reveals that values for air-corrected ³He/⁴He ratios range from 3.98 R_A to 6.03 R_A (where R_A represents the atmospheric ³He/⁴He ratio), CO₂/³He ratios vary from 2.20 × 10⁸ to 1.92 × 10¹¹, and δ¹³C values vary from −7.9‰ to −1.6‰. By comparing these measured values to those of typical mantle and crustal sources, we can infer that hot spring gases from Changbaishan Volcano are mostly characterized by inputs from two isotopically distinct sources: deep mantle fluids and shallower, slab-derived fluids. Fluids liberated from the shallower magma chamber are likely to include ancient Izanagi subduction zone fluids, whereas fluids originating from deeper magma chamber likely consist of MORB-like asthenospheric mantle fluids. Based on these results, we suggest that helium and carbon isotopes in hot springs demonstrate the absence of a direct material connection between Changbaishan Volcano and the west Pacific plate.

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1. Introduction

Changbaishan Volcano (also known as Mt. Paektu and Tianchi Volcano) is one of the most active intraplate volcanoes in China (Xu et al., 2012). Unlike volcanoes within Japan island arc, which are located close to the Japan Trench in the west Pacific subduction zone, Changbaishan Volcano is located ~1400 km west of the same subduction zone. Many researchers have attempted to characterize the source of this volcano using multidisciplinary techniques, including seismic tomographic, geochemical and tectonic methods (e.g., Wang et al., 2000; Tang et al., 2006; Zhao and Liu, 2010; Zou and Fan, 2011; Wei et al., 2012). Several studies have demonstrated that the west Pacific plate has subducted beneath northeastern China, forming a wide upper mantle wedge above the west Pacific stagnant slab (WPSS) (e.g., Zhao and Liu, 2010; Wei et al., 2012). According to these studies, the west Pacific plate should exert a strong geodynamic influence on Changbaishan

Volcano. However, the presence or absence of a material connection between the WPSS and Changbaishan Volcano is still a controversial topic (e.g., Wang et al., 2000; Tang et al., 2006; Chen et al., 2007; Hahm et al., 2008; Kuritani et al., 2011; Zou and Fan, 2011).

Gases emitted from hot springs have received a great deal of attention in recent years, as they can be utilized as sensitive tracers of seismic and volcanic activities (Gautheron and Moreira, 2002; Burgisser and Scaillet, 2007; Morikawa et al., 2008; Barry et al., 2013). In the Changbaishan volcanic field, hot springs are widely distributed and continually emitting gases, resulting in the emission of approximately 7.79 × 10⁵ t of CO₂ into the atmosphere every year (Guo et al., 2014). Among the hot spring gases, helium can be used as a sensitive geochemical indicator for volatile provenance due to its inertness and low solubility (Padron et al., 2012). It is well documented that most ⁴He is produced by radioactive decay within the crust and that most ³He is still escaping from the Earth's interior (Morrison and Pine, 1955; Hilton, 1996; Padron et al., 2012; Zelenski et al., 2012). Helium isotopes have been widely used to trace magma genesis and volcanic evolutions in different tectonic settings, as well as to monitor earthquakes and volcanoes (e.g., Kennedy and Van Soest, 2006, 2007; Ohno et al., 2011;

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Ohwada et al., 2012; Padron et al., 2012, 2013). When supplemented with CO₂ chemical and isotopic composition data, helium isotopic data can also be used to compute the percentages of mantle and slab-derived fluids in magmatic mixtures (Sano and Marty, 1995; Sano et al., 2006).

This work presents the study of a total of 22 hot spring gas samples collected in 2002, 2006, 2014 and 2015. Our study aims were the following: 1) to study the sources of hot spring gases from three hot springs at Changbaishan Volcano; 2) to analyze the upwelling processes of hot spring gases from the source to the surface based on the structure of this volcano; 3) to reveal the possible connection between Changbaishan Volcano and the WPSS by comparing their chemical and isotopic compositions with ³He/⁴He, δ¹³C and CO₂/³He values of mantle and crustal sources (Marty and Jambon, 1987; Sano and Marty, 1995; Gautheron and Moreira, 2002; Graham, 2002; Hilton et al., 2002; Shaw et al., 2006).

2. Regional setting

Changbaishan Volcano, on the boundary between China and North Korea (42°00' N, 128°03' E), is located ~1400 km west of the west Pacific subduction zone on 30- to 39-km-thick continental crust (Hetland et al., 2004; Li et al., 2006; Kyong-Song et al., 2016). Global and regional tomographic studies (e.g., Zhao and Liu, 2010; Wei et al., 2012; Tang et al., 2014) have suggested that the west Pacific plate has subducted beneath northeastern China at a dip angle of ~30° and that the subducting Pacific slab has become stagnant in the mantle transition zone; these studies also indicated that a wide mantle wedge may have formed above the WPSS. Because the upwelling of asthenospheric material is known to influence the evolution of back-arc intraplate volcanoes in northeast Asia, including Changbaishan Volcano and Wudalianchi Volcano (Xu et al., 2013b), it is widely accepted that the WPSS exerts a strong geodynamic influence on Changbaishan Volcano (e.g., Zhao et al., 2009; Zhao and Liu, 2010; Wei et al., 2012; Tang et al., 2014).

Changbaishan Volcano has undergone three stages of evolution: the early basalt shield stage, the middle trachyte composite cone stage and the late ignimbrite-forming stage (Wei et al., 2007). The most recent explosive eruption of Changbaishan Volcano was the Millennium eruption, which occurred in 946 CE; this eruption is believed to have been one of the largest eruptions of the past two thousand years in the world (Xu et al., 2013a). Written records also suggest that Changbaishan Volcano experienced at least three small recent eruptions following the Millennium eruption in 1688 CE, 1702 CE and 1903 CE. Currently, the highest point on the volcano is 2749 m above sea level, and the Tianchi caldera lake, which is roughly circular with a maximum diameter of ~5 km, contains approximately 2.04 billion m³ of water (Fan et al., 2007). During a recent period of magmatic unrest from 2002 to 2006, the number of earthquakes at the volcanic center increased by as much as two orders of magnitude, ground inflation was observed, and volcanic gases recorded increases in their CO₂, H₂ and He contents and ³He/⁴He values (Xu et al., 2012). To evaluate the risks and potential hazards posed by this volcano to nearby populations in East Asia, it is necessary to understand the mechanisms responsible for its eruptions and general unrest (Xu et al., 2012; Yu et al., 2013).

Within the volcanic field, large-scale continuous geothermal activities, along with strong gas release, are mainly located in three hot spring areas: Julong hot spring (on the north flank of the region), Jinjiang hot spring (on the southwest flank of the region) and Hubin hot spring (on the shoreline of the caldera lake). Julong hot spring lies to the north of Changbaishan waterfall and has a degassing area of approximately 3300 m². Jinjiang hot spring, which is located beside the Jinjiang River, has a degassing area of approximately 1300 m². Hubin hot spring lies along the lakeshore of Tianchi caldera lake and is approximately 500 m in length.

3. Sampling and analytical methods

Gas samples were collected at the three hot spring areas using water displacement methods to avoid air contamination. To obtain samples for chemical composition analysis, a funnel (with a mouth ~18 cm in diameter) was submerged in spring water, and a hand-pump was used to purge the sampling equipment. The inverted funnel was then placed over the upwelling bubbles to collect gases. After the gases were collected, a syringe was used to inject them into 500-mL pre-vacuumed aluminum bags whose lids were refilled with high-density silicone rubber. Gas samples for isotopic composition analysis were similarly collected in 125-mL inverted glass bottles under spring water.

Helium and CO₂ contents were analyzed with an Agilent 6890 N gas chromatograph (with a routine precision of 5%) using argon as a carrier gas. Analysis of isotopic compositions was conducted at Key Laboratory of Petroleum Resources Research, Institute of Geology and Geophysics, Chinese Academy of Sciences. Isotopic compositions of helium and neon were measured by a VG-5400 mass spectrometric system (in 2002 and 2006) and a Nu Instruments Noblesse SFT noble gas mass spectrometer (in 2014 and 2015). The precision of He and Ne isotopic compositions obtained from repeated measurements of an air standard was <7% and 10%, respectively. The isotopic composition of carbon was analyzed by a Thermo Finnigan MAT252 stable isotope mass spectrometer system (in 2002 and 2006) and a Thermo Finnigan MAT 253 stable isotope mass spectrometer system (in 2014 and 2015). The analytical accuracy of δ¹³C was within 0.6‰ (2σ) with respect to the Pee Dee Belemnite (PDB) standard.

4. Results

In this study, we measured the chemical and isotopic compositions of helium and carbon of 22 samples from Changbaishan Volcano in 2002, 2006, 2014 and 2015. These results are listed in Table 1.

Of the volcanic gases at Changbaishan Volcano, the most abundant content is CO₂, which varies in its abundance between 70.2% and 99.7%, with an average content of 91.8%. CO₂ is also the major volcanic gas component at other volcanoes in China, such as the Wudalianchi intraplate volcano (in Fig. 1) in northeastern China (Xu et al., 2013b) and the Tengchong volcanic field in southwestern China (Zhao et al., 2012). The relative abundance of helium in the volcanic gas samples ranged from 0.9 to 405 ppm, with an average value of 122 ppm.

4.1. ³He/⁴He ratios

The measured ³He/⁴He ratios (R_M/R_A), which are reported relative to atmospheric ³He/⁴He (R_A) (where R_A ≈ 1.4 × 10⁻⁶; Clarke et al., 1976), are corrected for the effects of atmospheric contamination and different solubilities of helium and neon by the ratios of their Bunsen coefficients and He/Ne ratios (Hilton, 1996). The corrected ³He/⁴He ratios (R_C/R_A) are expressed by the following equation:

$$R_C/R_A = [(R_M/R_A \times X) - 1]/(X - 1) \quad (1)$$

Considering that all our samples were collected from hot spring bubbles, the variable X can be calculated by the following equation:

$$X = \left[\left(\frac{{}^4\text{He}/{}^{20}\text{Ne}}{\text{measured}} \right) / \left(\frac{{}^4\text{He}/{}^{20}\text{Ne}}{\text{air}} \right) \right] \times (\beta_{\text{Ne}}/\beta_{\text{He}}) \quad (2)$$

where β_{Ne} and β_{He} are the Bunsen solubility coefficients for neon and helium, respectively, and (⁴He/²⁰Ne)_{measured} and (⁴He/²⁰Ne)_{air} are the

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