



$^{40}\text{Ar}/^{39}\text{Ar}$ geochronology of supergene K-bearing sulfate minerals: Cenozoic continental weathering and its paleoclimatic significance in the Tu–Ha Basin, northwestern China

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ARTICLE INFO

Article history:

Received 19 March 2015

Received in revised form 29 December 2015

Accepted 3 January 2016

Available online 8 January 2016

Keywords:

Weathering

$^{40}\text{Ar}/^{39}\text{Ar}$ geochronology

Supergene K-bearing sulfates

Tulufan–Hami Basin

Aridification

ABSTRACT

$^{40}\text{Ar}/^{39}\text{Ar}$ incremental-heating analyses of supergene jarosite and yavapaiite were conducted for three weathering profiles at the Hongshan, Liuhuangshan, and Caihuagou deposits in the Tu–Ha Basin. The analyses provide numerical constraints on the timing and duration of weathering and supergene enrichment. Well-constrained plateau ages and best-fitting inverse isochrons yield $^{40}\text{Ar}/^{39}\text{Ar}$ ages ranging from 33.3 ± 0.5 Ma to 3.3 ± 0.4 Ma (1σ). Our $^{40}\text{Ar}/^{39}\text{Ar}$ ages, combined with the published ages, indicate that a protracted history of weathering and supergene enrichment and, by inference, arid–semiarid climate (with at least a moderate amount of precipitation (>10 cm/y)) favorable to intense chemical weathering emerged at 33.3 Ma, 27.7–23.3 Ma, and 16.4–14.7 Ma, and prevailed from 11 to 7.8 Ma. Then, a progressive change from arid–semiarid toward hyperarid conditions and predominantly hyperarid conditions may have persisted since at least ca. 3.3 Ma. The climatic implications inferred from the weathering geochronology are in agreement with the chemical parameters and isotopic compositions of the Cenozoic sedimentary sequence from the Lianmuqin section in the Tu–Ha Basin, attesting to the reliability of weathering geochronology by the $^{40}\text{Ar}/^{39}\text{Ar}$ method as an indicator of paleoclimate in arid areas. Our results suggest that the retreat of the Paratethys Sea, which would have reduced eastward water vapor transport by the westerlies to the Tu–Ha Basin, led to its aridification in the Oligocene and that increased rain shadow effects, resulting from uplift of the Tibetan Plateau and Tian Shan Mountains, played important roles in the aridification history of the Tu–Ha Basin since the late Miocene.

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1. Introduction

Weathering profiles record a balance between weathering and erosion processes in the geomorphologic and geochemical evolution of the earth's surface (Tardy and Roquin, 1992; Thomas, 1994; Vasconcelos, 1999a). Supergene minerals are formed via weathering processes as thick covers on continents when chemical–physical weathering overwhelms chemical–physical erosion, and thick, chemical and mineralogical weathering profiles develop (Thomas, 1994; Vasconcelos, 1999a). The depth and complexity of weathering profiles, combined with the preservation of supergene minerals

throughout the profile, could constitute a long-term record of past climate evolution (Thomas, 1994; Vasconcelos, 1999a). Precise dating and geochemical investigation of supergene minerals may provide important information on the paleoclimatic conditions prevailing during weathering (Alpers, 1988; Vasconcelos, 1999a).

The potassium-bearing alunite-group sulfates (generalized composition $\text{AB}_3(\text{SO}_4)_2(\text{OH})_6$, jarosite- $\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$ and alunite- $\text{KAl}_3(\text{SO}_4)_2(\text{OH})_6$) are ubiquitous alteration products in supergene profiles developed over sulfide ore deposits and porphyry Cu deposits (Anderson, 1982; Vasconcelos, 1994, 1999b). Vasconcelos et al. (1994) applied laser step heating $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology to study weathering profiles from the western US and West Africa, demonstrating that supergene jarosite and alunite are suitable minerals for direct dating. The formation of alunite group minerals in weathering profiles requires a balance between (1) depression of the water table by tectonically induced uplift or denudation, and the resultant exposure of underlying sulfides to the effects of oxidative weathering, and (2) an arid–semiarid climate (at least moderate precipitation >10 cm/y) (Clark et al., 1990).

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However, because jarosite and alunite may be unstable when exposed to saline groundwater; the preservation of jarosite-alunite in weathering profiles is interpreted to record a transition toward arid conditions (Vasconcelos, 1999a). Therefore, $^{40}\text{Ar}/^{39}\text{Ar}$ dating of supergene alunite-group minerals represent a good paleoclimatic indicator and have been used as such in several studies, mainly from the Atacama Desert of Northern Chile and Peru (Alpers et al., 1988; Clark et al., 1990; Sillitoe et al., 1996; Mote et al., 2001; Bouzari et al., 2002; Quang et al., 2003, 2005; Araibia et al., 2006), North America (Arehart et al., 1992; Rye et al., 1993; Marsh et al., 1997; Vasconcelos et al., 1994), West Africa (Vasconcelos et al., 1994), and Australia (Bird et al., 1990; Vasconcelos et al., 2003). However, direct dating of weathering and supergene mineralization in Central Asia by K–Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ of the supergene K-bearing sulfates is rare. Only Xu et al. (2008), He et al. (2009), and Chen et al. (2014) reported the K–Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ results for jarosite or other supergene K-bearing minerals (ferricopiapite, voltaite, yavapaiite) from the oxidation zone of the Hongshan (K–Ar ages) and Baiyin deposits ($^{40}\text{Ar}/^{39}\text{Ar}$ ages, Gansu Province).

The Tulufan–Hami (Tu–Ha) Basin is one of the major hyperarid regions in northwestern China. This basin is surrounded by two large Asian mountain ranges: the Tibetan Plateau to the south and the Tianshan range to the north and northwest. Compared with extensive studies on the other arid basins (e.g., Tarim, Junggar) in Northwest China (Sun et al., 2006, 2008, 2009, 2013, 2015; Tang et al., 2011; Zhang and Sun, 2011), less is known about the history of the aridity and the associated forcing mechanisms of aridification in the Tu–Ha Basin. Only one study of multiple climatic proxy parameters from the discontinuous sequence of the Lianmuqin section in the Tu–Ha Basin indicates a record of Cenozoic climatic change (Cheng, 2005). Deep weathering profiles and associated supergene oxidation zones are widely distributed throughout the Tu–Ha Basin (Tu et al., 1963; Fang et al., 2002; Xu et al., 2006, 2008; Qin et al., 2001, 2008; He et al., 2009), in which alunite group minerals, especially jarosite, are commonly found. Their abundance in weathering profiles and their ease of access for complete sampling at deposit sites provide an excellent opportunity to date weathering processes and the formation of the oxidation zone of sulfide deposits by the $^{40}\text{Ar}/^{39}\text{Ar}$ method. In this paper, we address the reliable and promising dating method— $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology—to study three weathering profiles (the Hongshan deposit, the Liuhuangshan deposit, and the Caihuagou deposit) to place precise constraints on the timing and duration of weathering and supergene enrichment in the Tu–Ha Basin. Based on these results and previously published data (Xu et al., 2008; He et al., 2009; Yang, 2013), we infer the paleoclimatic conditions that led to the formation of the oxidation zone of these sulfide deposits and discuss the forcing mechanisms controlling the regional aridification of the Tu–Ha Basin.

2. Geological setting

The Tu–Ha Basin, located in the north of the Tibetan Plateau, is surrounded by the Tian Shan Mountains to the north and northwest (Fig. 1). The basin is currently characterized by a hyperarid climate, with a mean annual temperature of 14 °C, a mean annual rainfall of approximately 28 mm (Tulufan ~ 17 mm/yr; Hami ~ 34 mm/yr), and mean annual evaporation over 3000 mm, which makes it one of the driest deserts in the Asian interior (Fig. 2a, b) (Li et al. 1993; Miao et al., 2012). Large areas of the Tu–Ha Basin are mantled by thick supergene oxidation zones, which developed over sulfide ore deposits and host the most common supergene sulfate minerals: jarosite, alunite, and yavapaiite (Fig. 2) (Tu et al., 1963; Fang et al., 2002; Xu et al., 2006, 2008; Qin et al., 2001, 2008; He et al., 2009). In this paper, we focus on studying three sulfide deposits: the Hongshan deposit, the Liuhuangshan deposit, and the Caihuagou deposit (Fig. 1).

The Hongshan deposit (E91°28′–92°00′, N42°34′–42°40′) is a large high-sulfur hydrothermal Cu–Au deposit, which is situated in the center

of the Tu–Ha Basin (Qin et al., 2001; Fang et al., 2002) (Fig. 1). The elevation of this deposit is 450–500 m above sea level, and the landscape is dominated by very gentle topographic relief (Fig. 2a). The southern part of this deposit is composed of quartz diorite-monzonitic granite-biotite granite, whereas the north is composed of Silurian–Devonian basic volcanic rocks. The central portion of this deposit, related to copper and gold mineralization, comprises Mesozoic intermediate-acidic volcanic rocks that are mainly composed of quartz porphyry, albite porphyry, dacite, dacite porphyry, andesitic porphyry, and dioritic porphyry (Qin et al., 2001; Fang et al., 2002; Xu et al., 2008). Ore minerals include pyrite, chalcocite, bornite, chalcocite, and natural gold (Fang et al., 2002). The supergene enrichment forms a 50–60 m deep weathering profile, which provides suitable phases for weathering geochronology (Qin et al., 2001, 2008; Xu et al., 2006, 2008). Supergene jarosite mainly occurs at the bottom of the iron cap (0.5 m to 2 m below the surface) as a yellow subhorizon (Fig. 2c). A new supergene K-bearing sulfate mineral (yavapaiite) occurs at 14–15 m below the surface and is also found in the oxidation zone of this sulfide deposit.

The Liuhuangshan deposit (E88°28′20″, N41°58′00″) is located along the southwestern margin of the Tu–Ha Basin (Fig. 1). The elevation of this deposit is approximately 1050–1200 m above sea level. The ore deposit was discovered in the Hercynian quartz porphyry rock body, which is intruded into the Upper Ordovician Liuhuangshan sedimentary stratum (Li, et al., 2003). The wall rock alteration near the ore body is very strong, and it mainly presents as silicification, pyritization, sericitization, and baratization (Li, et al., 2003). The primary ores are pyrite, chalcocite, cupriferous pyrite, sphalerite, and chalcocite (Li, et al., 2003). In this deposit, intense weathering has produced thick weathered profiles, up to 30 m thick (Fig. 2d). Supergene jarosite mainly occurs at the bottom of the iron cap as a yellow sub-horizon whose thickness can be up to 8 m.

The Caihuagou deposit (E88°53′00″–88°56′40″, N42°08′20″–42°09′56″) is located approximately 20 km northeast of the Liuhuangshan deposit (Fig. 1). The elevation of this deposit is 1100–1280 m above sea level. The ore deposit is hosted by the lower Devonian Arpishmebulaq Formation of the central Asia group, in which rocks have different degrees of deformation and metamorphism because of regional metamorphism (Si et al., 2011). The main rocks here are chlorite quartz schist, sericite quartz schist sandwiched marble, and biotite quartz schist. Primary ores are mainly pyrite, chalcocite, and sphalerite, as well as small amounts of molybdenite, and tetrahedrite (Si et al., 2011). An oxidation zone has formed at the top of the deposit, and it mainly includes jarosite, limonite, covellite, and malachite. Supergene jarosite mainly occurs at the bottom of the iron cap and is distributed at 0.5 m to 3 m below the surface (Fig. 2e).

3. Sampling and analytical procedures

Supergene K-bearing sulfates suitable for $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology were collected from the three weathering profiles. During the course of sampling, we collected jarosite samples exhibiting cavity infillings (Fig. 2f, g), which are generally devoid of contaminants and thus ideal for $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology. At Hongshan, samples were collected from discrete mining pits that span approximately 20 m depth: supergene jarosite samples collected from 0.5 m to 2 m below the surface and yavapaiite samples collected from 15 m below the surface (pale pink mineral, Fig. 2h). At the Liuhuangshan deposit, the jarosite samples were obtained from 4 m to 5 m below the surface (although systematic sampling from the top to the bottom was permitted, the remaining collected samples had not been dated). At the Caihuagou deposit, jarosite samples were obtained only at the bottom of the iron cap on the hill-tops, and collection of additional samples at depth was not possible due to inaccessibility. We investigated eleven supergene K-bearing mineral samples in the three weathering deposits.

Polished thin sections of each sample were studied by transmitted- and reflected-light microscopy to determine the mineralogy and

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