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Fluid Inclusion, H-O, S, Pb and noble gas isotope studies of the Aerhada Pb-Zn-Ag deposit, Inner Mongolia, NE China



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ABSTRACT

The Aerhada Pb-Zn-Ag deposit is located in the western segment of the Great Hinggan Range Ag-Pb-Zn-Cu-Mo-Au-Fe metallogenic belt in NE China. Orebodies occur mainly as vein type and are hosted by sandstone and siliceous slate. Three stages of primary mineralization, including an early arsenopyrite-pyritequartz, a middle polymetallic and silver sulfides-quartz and a late sphalerite-pyrite-calcite-fluorite are recognized. Four types of fluid inclusions have been identified in the ore-bearing quartz and fluorite veins, i.e., liquid-rich, gas-rich, three-phase CO2 aqueous inclusions, and pure gas or liquid aqueous inclusions. Microthermometric studies on fluid inclusions reveal that homogenization temperatures from early to late stages range from 253° to 430 °C, 195° to 394 °C and 133° to 207 °C, respectively. Fluid salinities range from 2.9 to 14.0 wt.% NaCl equiv. The vapor composition of the ore fluid is dominated by H₂O, CO₂ and CH₄, with minor proportions of N₂. The fluid $\delta^{18}O_{H2O}$ and δD_{H2O} values vary from +1.6 to +9.3% and -122 to -56%, respectively, and reflect a magmatic fluid and a meteoric fluid dominant hydrothermal system for the early and late stages of mineralization, respectively. The calculated $\delta^{34}S_{H2S}$ values of hydrothermal fluids in equilibrium with sulfides range from +5.2 to +7.1%, suggesting a mixed source for sulfur, i.e., the local magmatic and sedimentary rocks. The Pb isotope compositions of sulfides are similar to those of the local magmatic and sedimentary rocks, implying that lead and possibly silver relate to these sources. The noble gas isotope compositions of fluid inclusions hosted in ore minerals suggest that the ore-forming fluids were dominantly derived from a deep mantle source. Fluid mixing and dilution are inferred as the dominant mechanisms for ore deposition. The Aerhada Pb-Zn-Ag deposit can be classified as a medium to low temperature hydrothermal vein type deposit.

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1. Introduction

The Great Hinggan Range (GHR) in the easternmost part of the Central Asian Orogenic Belt is an important Ag-Pb-Zn-Mo-Cu-Au-Fe metallogenic belt in NE China (Zeng et al., 2010, 2011, 2012a, b; Zhang and Li, 2014, 2017; Ouyang et al., 2014; Zhai and Liu 2014; Zhai et al., 2014a,b,c, 2015; Leng et al., 2015; Shu et al., 2016; Chen et al., 2012, 2017). In recent years, a number of Pb-Zn-Ag polymetallic deposits have been discovered in the GHR, namely the Erentaolegai Ag deposit (Chen and Zhang, 2004), the Jiawula Pb-Zn-Ag deposit (Zhai et al., 2013), the Meng'entaolegai Ag-Pb-Zn deposit (Zhu et al., 2006), the Bianjiadayuan Ag-Pb-Zn deposit (Wang et al., 2013b), the Bianbianshan Au-Ag-Cu-Pb-Zn

* Corresponding author. E-mail address: zhanghongyu@cugb.edu.cn (H. Zhang). deposit (Zeng et al., 2012b), the Shuangjianzishan Ag-Pb-Zn deposit (Liu et al., 2016) and the Caijiaying Cu-Pb-Zn deposit (Wang et al., 2004). Furthermore, the hydrothermal vein type Pb-Zn-Ag polymetallic ore deposits are common and prominent in the region (Zhu et al., 2006; Zeng et al., 2012b; Ouyang et al., 2014; Wang et al., 2014). It is proposed that the magmatic-hydrothermal deposits in the GHR formed during two different metallogenic episodes (Li et al., 2012), an early episode in the Late Permian (i.e., 256 ± 7 to 272 ± 3 Ma) and a late episode in the Jurassic and Cretaceous (i.e., 129 ± 3 to 167 ± 2 Ma). Most of the porphyry Cu-Mo deposits formed during the second metallogenic episode, which was genetically related to the widespread intrusion of A-type granites (Mao et al., 2005), whereas timing for the vein type Pb-Zn-Ag deposits remain poorly constrained.

The Aerhada Pb-Zn-Ag deposit (1.0 Mt Pb + Zn@5.18% and 1000 t Ag@57.66 g/t) is located in the west segment of the GHR in NE



China (Zhang et al., 2015). Previous studies on the Aerhada deposit, mostly published in Chinese, addressed the ore deposit geology (Gao and Qian, 2005; Tao, 2006), mineralogy (Wang et al., 2013a), whole rock geochemistry (Yang et al., 2014), granite geochronology (Ke et al., 2016), sulfur and lead isotope compositions and ore deposit genesis (Qian and Gao, 2006; Zhang et al., 2007a, 2015). However, issues related to fluid evolution, metal source, and precipitation mechanisms remain poorly constrained.

Our study utilizes detailed fieldwork and numerous new fluid inclusion and multiple isotope (H-O, S, Pb, He-Ar) data coupled with previous published data. We aim to address the issues related to the source, nature and evolution of the ore-forming fluids, sources of metals, and mechanisms of ore deposition of the Aerhada Pb-Zn-Ag deposit.

2. Regional geology

The western part of the GHR is located in the Paleozoic orogenic belt on the south side of the South Mongolian tectonic belt and is placed between the North China plate and the Siberian plate, which is an important part of the Central Asian Orogenic Belt and metallogenic belt (Fig. 1a; Nie et al., 2007a). The Aerhada Pb-Zn-Ag deposit is located eastward of the late Paleozoic to Mesozoic Chaganobo-Aoute-Chaobuleng magmatic belt at the southeastern margin of the Siberia plate, which was involved in northeastern East Ujimqin-Erenhot synclinorium (Fig. 1b; Zhang et al., 2007a; Huang et al., 2013). The NE-trending Erlianhot-Hegenshan and Chagan Obo-East Ujimqin faults control the spatial distribution of ore deposits in the district. Regional tectonics comprise NE-, NNE-, NW- and EW-trending faults (Huang et al., 2013). Folding is also apparent with a series of NE-trending composite anticlines and synclines, and their axes follow the trends of the regional faults (Zhang et al., 2015). The main ore-bearing folds in the area are dominated by the Erengaobi synclinorium (Wang, 2003).

The upper Paleozoic rocks are widely exposed in the region, and include the Middle Ordovician Hanural Formation, the Silurian Wodouhe Formation, and the Devonian to lower Permian Baolige Formation. The Hanural Formation which mainly appears in northeast of the Erengaobi area, consists of micaceous schists, sandy slates intercalated with marbles, tuffaceous sandstones and siltstones. The Devonian rocks consist of the lower Devonian Nigiuhe Formation, and the middle to upper Taerbageta Formation, the upper Devonian Angeryinwula Formation, which are mostly fine grained clastic rocks, while the lower Permian Baoligaomiao Formation mainly comprises sandstones, siltstones, conglomerates and volcano-clastic rocks, and locally are the host of the ore deposits (Gao and Qian, 2005; Tao, 2006; Ma et al., 2014; Zhang et al., 2015). The Jurassic Baiyanhua, Manite, Baiyin Gaolao, Manitu and Manketou Obo Formations are composed of lavas, volcanoclastic and clastic sedimentary rocks (GMBIMAR, 1991).

The regional Paleozoic Central Asian–Mongolian orogenic deformation, together with the Mesozoic volcanic and igneous intrusions are a favorable setting for the local mineralization (Fig. 1b; Jin et al., 2005; Chang et al., 2014; Zhang et al., 2015). Three distinct periods of magmatic activity, namely Hercynian, Indosinian and Yanshanian located in the west of the GHR district, are recorded that are mainly composed of granites and diorites. The Hercynian alkali granites are widely distributed, occur as batholiths and stocks and follow a general strike to the northeast which is consistent with the regional faults (Nie et al., 2007a). Quartz diorites and syenites of Indosinian ages appear only northeast of the Aerhada deposit. The Yanshanian calc-alkaline granites are widespread in north and northeast of the Aerhada area, and are emplaced along fold structures. All the later plutonic rocks have high alkalinity, lower initial (87 Sr/ 86 Sr)i and positive. ϵ Nd(t) values

with younger model ages (Hong et al., 2000). The volcanic rocks in the area are coeval or slightly earlier than the Early Permian alkaline granites and together they constitute the Early Permian riftrelated alkaline magmatic belt (Jahn et al., 2009).

3. Ore deposit geology

In the ore district volcano-sedimentary rocks of the Upper Devonian Angervin Wula Formation and the Upper Jurassic Baivin Gaolao Formation overlain by Tertiary and Quaternary rocks are exposed (Fig. 2a). The Devonian Angeryin Wula strata, which are the main host rocks of the ores, are common in the mining area and comprise andesitic tuff, sandstone, siltstone, argillaceous and siliceous slate. The upper Jurassic Baiyin Gaolao Formation, consists of alternations of conglomerate and rhyolitic pebbly tuff and rhvolite (Chen et al., 2009). It is located in northwest of the ore district with an unconformity contact to the underlying Angeryin Wula Formation (Fig. 2a). The Indosinian biotite-granite batholith and the early Yanshanian Anerji Wula biotite-granite stock located 2-3 km to northeast and northwest of the ore deposit, respectively (Zhang et al., 2015). ENE-, NW- and NS-trending faults and fractures are the main tectonic elements in the ore district. Both intrusive rocks and dikes are rare in the actual ore district.

Three distinct ore zones (I, II and III) have been identified in the Aerhada deposit (Fig. 2a), in which the Pb-Zn-Ag mineralization occurs as veins. Zone I, which is located in the northern part of the deposit, has a length of 3200 m and a width of 350 m to >600 m with a general strike of 305° (Fig. 2a, b). The ore-bearing veins strike southwest, with a dip ranging from 25° to 55° (Fig. 2c). This ore zone hosts the majority of the ores. A total of nearly 50 mineralized veins have been identified between exploration lines of No.7 and 39 in ore zone I, of which 47 orebodies are estimated to contain a reserve of ~0.6 Mt of Pb+Zn and 630 t of Ag. Ore zone II is placed in the central part of the deposit and has a length of 1600 m and a width of 100 m (Fig. 2a). Six major orebearing veins in this belt are inclined to southwest with a dip of 67° and strike northwest with angles between 280° and 300°. Ore zone III, which is located in southwest of the district, is 3500 m long and 100 to 400 m wide, with a general strike of 300°. The orebodies except from the base metals and silver also contain trace amounts of Sn, Ga, Ge, In and Au.

The main ore minerals are galena, sphalerite, and pyrite (Fig. 3a-c), with minor arsenopyrite (Fig. 3a), chalcopyrite, chalcocite and argentite. The alteration minerals comprise quartz, chlorite, epidote, kaolinite, sericite, calcite, green fluorite and clay minerals (Fig. 3d). In general, mineral assemblages of quartzepidote, quartz-chlorite-epidote-sericite, and calcite-fluoritekaolinite-clay minerals formed from early to late stages. Hydrothermal alteration is intense only around the quartz veins, including silicification and carbonatization. Other alteration types consist of kaolinization, sericitization, chloritization and epidotization. The alteration zonation is not clearly developed as in most places the different alteration types overprint one another. However, in some parts alteration occasionally exhibits a zonation pattern of siliceous, carbonate, sericite, epidote, chlorite and argillic (kaolinite) alteration from the mineralized vein towards the host rock.

Based on field cross-cutting relationship of veins, ore and alteration mineral assemblages and their textural relationships in the deposit, we have distinguished three paragenetic stages, i.e., early arsenopyrite-pyrite-quartz (stage I) (Fig. 3a), followed by galena-sphalerite-pyrite-silver mineral-quartz (stage II) (Fig. 3b, c) and sphalerite-pyrite-calcite-fluorite (stage III) (Fig. 3c, d).



Fig. 1. (a) Schematic tectonic map of China (based on Meng et al., 2013); (b) Geological map showing the major ore deposits in the western segment of the southern Great Hinggan Range (based on GMBIMAR, 1991).

4. Sampling and analytical methods

Over fifty samples were collected from different mining levels in the Aerhada deposit, including sulfides, e.g., galena, sphalerite and pyrite that intergrown with vein quartz and fluorite. Ten samples were selected for fluid inclusion analyses, including three quartz samples representing stage I, six quartz samples for stage II, and one fluorite sample for late stage III. Microthermometric measurements were performed using a Linkam THMSG 600 heating-freezing stage coupled to a ZEISS microscope at China University of Geosciences, Beijing. The temperature range for the analysis was from -196° to $+600 \ ^{\circ}$ C. The estimated accuracy of the freezing and heating measurements were $\pm 0.1 \ ^{\circ}$ C from -100to 25 $\ ^{\circ}$ C, $\pm 1 \ ^{\circ}$ C from 25 to 450 $\ ^{\circ}$ C, and $\pm 2 \ ^{\circ}$ C above 400 $\ ^{\circ}$ C, respectively. Fluid salinities were calculated utilizing the equations from Hall et al. (1988). Fluid inclusion laser Raman spectroscopy analyses were carried out in the Beijing Research Institute of Uranium Geology, Beijing, using a Renishaw RM-2000 Raman microscope. This instrument records peaks in the range of $100-4000 \text{ cm}^{-1}$ full-band with a resolution of $1-2 \text{ cm}^{-1}$ and the laser beam spot size was about 1 µm. The inclusions were analyzed for the most common gases, monoatomic and polyatomic ions and molecules.

All mineral separates selected were handpicked and checked under a binocular microscope to ensure a purity of >98%. The oxygen, hydrogen, sulfur and lead isotope analyses were performed at the Beijing Research Institute of Uranium Geology, Beijing. Six different quartz and calcite samples from the stages I, II and III were analyzed for their oxygen (quartz and calcite) and hydrogen (fluid



Fig. 2. (a) Geological map of the Aerhada Pb–Zn–Ag deposit (modified from Zhang et al., 2007a); (b) Geological section of the 808 m mining level of ore belt I (after Xu and Cui, 2005) and (c) Geological cross section of exploration line 31 of ore belt I (after Xu and Cui, 2005).

inclusions) isotopic compositions using a MAT-253 stable isotope ratio mass spectrometer. Oxygen was liberated from quartz by reaction with BrF₅ and converted to CO₂ on a platinum-coated carbon rod, following the extraction technique of Clayton and Mayeda (1963). The analysis of the oxygen isotopes for calcite was measured following the detailed procedures outlined by Cao et al. (2015). The hydrogen isotopic compositions of the fluid inclusions were analyzed for the same samples. Samples were first degassed by being heated under vacuum for 3 h at 150 °C. Water was released by heating the samples to approximately 500 °C by means of an induction furnace. Water was converted to hydrogen by passing over heated zinc powder at 410 °C. The analytical precision was better than ±0.2‰ for δ^{18} O and ±2‰ for δ D. The isotopic ratios are reported in standard δ notation (‰) relative to SMOW. Sulfur isotopic ratios were measured on a Delta V plus mass spectrometer, using the VCDT standard. The analytical precision and accuracy are better than ±0.2‰. The lead isotope analyses were performed by using a thermal ionization mass spectrometry (ISOPROBE-T). The measured Pb isotopic ratios of the international standard NBS981 is around 0.1‰.

Samples collected for noble gas isotope analyses belong to stage II. Helium and argon isotope compositions were measured on a MI-1201IG inert gas mass spectrometer at the Isotope Laboratory, Institute of Mineral Resources, Chinese Academy of Geological Sciences. ³He was measured by using an electronic multiplier and ⁴He by a Faraday cup. Sulfides were crushed under vacuum, and the released noble gases were purified four times by titanium sponge getter, zircon-aluminum getter and active carbon cooling trap with liquid nitrogen. The released helium and argon were admitted to the spectrometer and then analyzed, following the procedure of Wang et al. (2008).

5. Analytical results

5.1. Fluid inclusions

At room temperature, four types of fluid inclusions were identified on the basis of the phase relationships:

(1) Aqueous liquid and vapor inclusions (type I): The majority of these inclusions are colorless and transparent, followed by yellow, brown, gray or black. Their length typically ranges from 5 to 10 μ m, even some are up to ~20 μ m. They have negative-crystal, oval, fusiform, elongated, and irregular forms. They contain ~60 to 80 vol.% liquid and homogenize to liquid upon heating. Type I inclusions account for approximately 80% of the total population and their hosts are quartz and fluorite (Fig. 4a, b).



Fig. 3. Representative Pb-Zn-Ag ores and vein cutting relationships from the Aerhada deposit. (a) Arsenopyrite-pyrite vein in early stage I; (b) Pyrite-chalcopyrite-sphaleritegalena ore in stage II vein; (c) Late pyrite vein (stage III) crosscutting galena and sphalerite vein (stage II); (d) Calcite and fluorite veins (stage III) crosscutting pyrite-galena vein (stage II). Abbreviations: Apy-arsenopyrite; Cp-chalcopyrite; Gn-galena; Py-pyrite; Sp-sphalerite; Cal-calcite; Fl-fluorite.



Fig. 4. Photomicrographs of fluid inclusions in the Aerhada Pb–Zn–Ag deposit. (a)-(b) Liquid-rich aqueous inclusions in quartz and fluorite, respectively; (c)-(d) Vapor-rich aqueous inclusions in quartz; (e) CO₂ three phase aqueous inclusions; (f) Pure gas and liquid aqueous inclusions.

- (2) Vapor-rich aqueous inclusions (type II): The majority of type II inclusions also appear colorless and transparent, with some in brown or black, and with the same dimensions as type I. Their shapes include negative-crystal, oval and fusiform forms. They typically contain >60 vol. % vapor and homogenize to vapor upon heating. They comprise approximately 15% of the total population (Fig. 4c, d).
- (3) CO₂ three-phase aqueous inclusions (type III): These inclusions are gray or brown, and they consist of liquid and vapor CO₂ and aqueous liquid. These inclusions are 5–15 μ m and appear with negative-crystal and oval forms. They have V/L + V ratios ranging from 30 to 50% (Fig. 4e).
- (4) Monophase vapor or liquid aqueous inclusions (type IV): The majority of this type inclusions are colorless, transparent or black. These inclusions are 5–15 μ m and have negative-crystal, oval and fusiform or even irregular forms (Fig. 4f). Both type III and IV inclusions account only for ~5% of the total population.

Microthermometric results and calculated salinities are listed in Table 1 and displayed in Fig. 5. Homogenization temperatures of fluid inclusions from early to late stages varied from ~253° to 430°, 195° to 394°, and 133° to 207 °C, respectively. The obtained final ice melting temperatures for type I fluid inclusions range from -10.1° to -2.8 °C for stage I, -9.3° to -1.7 °C for stage II and -6.8° to -2.2 °C for stage III, which correspond to salinities of 4.7 to 14.0, 2.9 to 13.2, and 3.7 to 10.2 wt.% NaCl equiv, respectively, according to the relationship between final ice melting temperatures and salinities (Hall et al., 1988).

Laser Raman analyses of individual fluid inclusions indicate that the vapor phases present at type I and II inclusions in stage I are H₂O and CO₂ (Fig. 6a, b) with minor amounts of CH₄ (Fig. 6a). The vapor phases contained in type I inclusions that relate to stages II and III are H₂O, CH₄ and CO₂ with minor amounts of N₂ (Fig. 6c–f). Furthermore, Type III inclusions comprise liquid and vapor CO₂ (Fig. 6b). The fluid inclusion can be described in the H₂O-NaCl-CO₂ ± CH₄ ± N₂ system.

5.2. Hydrogen-oxygen isotopes

The measured δ^{18} O values for quartz in stages I and II display a relatively narrow range, i.e., from 13.3 to 15.9‰ (Table 2). The calculated $\delta^{18}O_{H2O}$ values of the ore-forming fluids in equilibrium with quartz range from 8.0 to 9.3‰, using the equation of Clayton et al. (1972). The δD_{H2O} values range from -122 to -113‰. The δ^{18} O values obtained for calcite in stage III range from 13.4 to 13.7‰ (Table 2), corresponding to $\delta^{18}O_{H2O}$ values that

 Table 1

 Microthermometric results of fluid inclusions from the Aerhada Pb–Zn–Ag deposit.

range from 1.6 to 1.9%, calculated based on the equation of O'Neil et al. (1969).

5.3. Sulfur-lead isotopes

Our new sulfur isotope data for the Aerhada deposit combined with previous published data from Zhang et al. (2007a,b) are displayed in Table 3 and Fig. 8. In detail, the δ^{34} S values range from 1.2 to 5.3% for galena, 4.6 to 7.5% for sphalerite, 4.4 to 8.6% for pyrite and 6.4 to 6.8% for arsenopyrite, respectively. Based on the obtained sphalerite-galena and pyrite-galena sulfur isotope data (Table 3), the calculated S isotope equilibrium temperatures on the basis of equations from Ohmoto and Rve (1979) show a wide range from 195° to 529 °C for the Aerhada mineralization. The wide range of the obtained temperatures from sulfur isotopic pairs and fluid inclusions can be attributed to isotopic disequilibrium of the examined sulfides. Alternatively, the attained fluid inclusion temperatures could only represent the minimum fluid trapping temperatures which are lower than the crystallization temperatures of the contained sulfides. Lastly, the calculated $\delta^{34}S_{H2S}$ values of the ore-forming fluids in equilibrium with sulfides ranged from 5.2 to 7.1%, utilizing the equations of Seal (2006).

The lead isotope compositions of sulfides from the Aerhada deposit were compared with the previous published data (Table 4, Zhang et al., 2007a,b, 2012; Nie et al., 2007a,b). In general, galena is characterized by 206 Pb/ 204 Pb and 208 Pb/ 204 Pb ratios ranging from 18.187 to 18.685 and 37.693 to 39.301, respectively, which are higher than the ratios obtained from pyrite, i.e., 206 Pb/ 204 Pb and 208 Pb/ 204 Pb ratios from 18.153 to 18.293 and 37.653 to 37.909. The 206 Pb/ 204 Pb and 208 Pb/ 204 Pb ratios for sphalerite range from 18.174 to 18.431 and 37.672 to 38.213 and are similar to the ratios obtained from arsenopyrite, e.g., 18.258 to 18.429 and 37.842 to 38.421.

5.4. He-Ar isotopes

The noble gas (He and Ar) isotope compositions measured for sulfides (galena and pyrite) in this study are listed in Table 5. The obtained ³He/⁴He ratios of fluid inclusions from the analyzed sulfides show a range from 2.50 to 5.41 Ra, where Ra represents the ³He/⁴He ratios of the atmosphere with a value of 1.4×10^{-6} (Mamyrin and Tolstikhin, 1984). ⁴⁰Ar/³⁶Ar ratios of fluid inclusions in all analyzed samples are relatively constant and display a narrow range of 291 to 301 (average of 296). The ⁴He concentrations vary from 1.35 to 4.03×10^{-8} cm³ STP g⁻¹, and the ⁴⁰Ar concentrations range between 0.37 and 11.73×10^{-7} cm³ STP g⁻¹.

Sample	Mineralization stage	Host mineral	N	Vapor (vol.%)	Th (°C)		Tm (°C)		Salinity (wt.% NaCl _{eqv.})	
					Range	Mean value	Range	Mean value	Range	Mean value
14AEHD-2	Early stage	Q	20	10-40	$280.4\sim430.1$	354.1	$-8.7\sim-2.8$	-5.9	$4.7 \sim 12.5$	9.0
15AEHD-8		Q	20	5-40	$252.9\sim 397.2$	349.7	$-8.4\sim-4.7$	-6.3	$7.5 \sim 12.2$	9.6
15AEHD-4		Q	16	5-30	$296.8\sim389.1$	332.3	$-10.1\sim-3.1$	-6.3	$5.1 \sim 14.0$	9.5
14AEHD-10	Middle stage	Q	19	5-40	$259.8\sim 393.8$	315.6	$-9.3\sim-2.4$	-5.6	$4.0 \sim 13.2$	8.5
14AEHD-23		Q	23	5-30	$245.3\sim347.6$	311.8	$-8.7\sim-2.1$	-5.2	$3.5 \sim 12.5$	8.1
15AEHD-9		Q	43	5-45	$213.9\sim 392.5$	307.8	$-9.1\sim-2.4$	-5.6	$4.0 \sim 13.0$	8.5
15AEHD-24		Q	39	5-50	$238.4\sim346.6$	307.6	$-8.5\sim-2.8$	-4.6	$4.7 \sim 12.3$	7.3
14AEHD-26		Q	19	5-30	$238.6\sim351.1$	303.0	$-8.8\sim-4.1$	-6.5	$6.6 \sim 12.6$	9.8
15AEHD-11		Q	20	5-20	$194.8\sim358.2$	288.1	$-5.1\sim-1.7$	-4.0	$2.9 \sim 8.0$	6.4
15AEHD-12	Late stage	Fl	13	5-20	$133.1\sim207.3$	158.3	$-6.8\sim-2.2$	-4.1	$3.7 \sim 10.2$	6.6

Abbreviations: N: number of measured fluid inclusions; Th: Homogenization temperature; Tm: Final ice melting temperature; Q: quartz, Fl: fluorite.



Fig. 5. Histogram of homogenization temperature and salinity of the ore fluid for the Arehada Pb-Zn-Ag deposit.

6. Discussion

6.1. Fluid evolution

Microthermometric studies on fluid inclusions hosted in quartz and fluorite in textural equilibrium with sulfides from stages I to III display a gradual decrease of temperature, i.e., the homogenization temperatures range from 430° to 253 °C, 394° to 195 °C and 207° to 133 °C, respectively (Table 1, Fig. 5, 11). Furthermore, fluid salinity shows a similar decreasing trend, i.e., from 9.6 to 6.6 wt.% NaCl equiv. Therefore, the Aerhada hydrothermal system is characterized by both temperature and salinity decrease during the evolution of the ore fluid (Figs. 5 and 11). Such a decrease in salinity with drop of temperature is indicative for fluid mixing and dilution (Fig. 11). Laser Raman analyses on fluid inclusions also suggest that the vapor phase in stage I was dominated by H_2O and CO_2 (Fig. 6a, b) with minor CH_4 whereas in stages II and III vapor mainly composed of H₂O, CH₄ and CO₂ (Fig. 6c–f) with minor N₂. From stages I to III the CO₂ contents tend to decrease gradually whereas the CH₄ contents to increase. This trend suggests that the ore fluid evolved from an early NaCl-H₂O-CO₂ \pm CH₄ through a middle NaCl-H₂O-CH₄ \pm CO₂ \pm N₂ into a late NaCl-H₂O-CH₄ \pm CO₂ system, i.e., that the mineralizing fluid was diluted and reduced. We conclude that the ore-forming fluid responsible for the deposition of the Aerhada ores evolved due to fluid mixing, associated with the incursion of a relative cooler, reducing and dilute fluid in the hydrothermal system, which triggered the precipitation of the Pb-Zn-Ag sulfides.

6.2. Source of ore-forming fluids, sulfur and metals

The $\delta^{18}O_{H2O}$ values for stages I and II are similar to those of magmatic waters, and those for late stage III are even lower. The δD_{H2O} values for stages I and II ore fluids plot below the field of



Fig. 6. Representative Raman spectra of fluid inclusions in the Aerhada deposit. (a) and (b) CO₂-bearing fluid inclusions from early stage quartz; (c) and (d) CH₄, CO₂ and N₂-bearing fluid inclusions from middle stage quartz; (e) and (f) CH₄ and CO₂-bearing fluid inclusions from late stage fluorite.

Table 2

Hydrogen and oxygen isotopic data of the Aerhada Pb-Zn-Ag deposit.

Sample	Mineralization stage	Minerals	δD_{V-SMOW} (‰)	$\delta^{18}O_{V-SMOW}$ (‰)	Th (°C)	$\delta^{18} O_{\rm H2O} (\%)$
15AEHD-4	Early stage	Quartz	-113	13.8	332.3	8.0
15AEHD-8	Early stage	Quartz	-122	13.3	349.7	8.0
15AEHD-9	Middle stage	Quartz	-119	15.7	307.8	9.1
15AEHD-24	Middle stage	Quartz	-112	15.9	307.6	9.3
15AEHD-12	Late stage	Calcite	-58	13.4	154.7	1.6
15AEHD-20	Late stage	Calcite	-56	13.7	154.7	1.9

Table 3

Sulfur isotope compositions of sulfides from the Aerhada Pb-Zn-Ag deposit.

No.	Sample	Sulfide	δ^{34} S (‰)	No.	Sample	Sulfide	δ^{34} S (‰)
1	14AEHD-17	Galena	4.5	16	TW11	Galena	4.7
2	15AEHD-23	Galena	4.6	17	TW7	Sphalerite	6.9
3	15AEHD-25	Galena	4.3	18	TW8	Sphalerite	5.5
4	14AEHD-21	Sphalerite	4.6	19	TW9	Sphalerite	7.5
5	15AEHD-5	Sphalerite	6.6	20	TW10	Sphalerite	7.4
6	14AEHD-17	Pyrite	6.1	21	TW11	Sphalerite	7.0
7	14AEHD-21	Pyrite	4.4	22	TW2	Pyrite	8.6
8	15AEHD-5	Pyrite	5.9	23	TW3	Pyrite	7.0
9	15AEHD-23	Pyrite	6.7	24	TW5	Pyrite	7.2
10	15AEHD-25	Pyrite	6.2	25	TW8	Pyrite	6.1
11	TW1	Galena	1.2	26	TW9	Pyrite	6.6
12	TW4	Galena	4.7	27	TW10	Pyrite	7.0
13	TW7	Galena	5.3	28	TW4	Arsenopyrite	6.8
14	TW8	Galena	3.9	29	TW7	Arsenopyrite	6.6
15	TW9	Galena	4.2	30	TW11	Arsenopyrite	6.4

Note: Number 1–10 data are from this study, and 11–30 data are collected from Zhang et al. (2007a,b).



Fig. 7. $\delta^{18}O_{H20}$ - δD isotope plots of ore fluids for the Aerhada Pb-Zn-Ag deposit. The green rectangle area represents the D-O isotope compositions for numerous hydrothermal Pb-Zn-Ag deposits in the southern part of the Great Hinggan Range (Zhu et al., 2006; Zeng et al., 2012b; Han et al., 2013; Ouyang et al., 2014; Wang et al., 2014 and Sun et al., 2014). Data for the Mesozoic meteoric water are from Zhang (1985). Fields of magmatic water are based on Hedenquist and Lowenstern (1994); the metamorphic water box and the meteoric water line are adopted from Giggenbach (1992) and Hoefs (2009).



Fig. 8. Histogram of sulfur isotope compositions for sulfides from the Aerhada deposit.

the typical magmatic waters and those for stage III are similar to the magmatic water (Fig. 7). This drop of the $\delta^{18}O_{H2O}$ values can be explained due to an addition of meteoric waters in the hydrothermal system, resulting in a migration of the $\delta^{18}O_{H2O}$ and δD_{H2O} values towards the meteoric water line. The relatively low δD_{H2O} ratios in the early and middle stages and subsequent increase of δD_{H2O} values in late stage fluids can be attributed to the effects of the secondary fluid inclusions to the hydrogen isotope composition, as these failed to decrepitate and contributed isotopically light meteoric water thereby lowering the δD_{H2O} values (Goldfarb and Groves, 2015). The obtained D-O isotope data from the Aerhada deposit are very similar to those of numerous hydrothermal vein type Pb-Zn-Ag deposits in the southern GHR ore belt (i.e., those deposits showed δD values from -130 to -50% and $\delta^{18}O_{H2O}$ values from -5.5 to 8%, Fig. 7), which were mostly related to magmatic and meteoric fluids (Zhu et al., 2006; Zeng et al., 2012b; Han et al., 2013; Ouyang et al., 2014; Wang et al., 2014). This conclusion is consistent with our fluid inclusion studies, showing a fluid mixing during its evolution.

The present δ^{34} S values of sulfides are combined with previous data from the Aerhada Pb-Zn-Ag deposit. They display a range from +1.2 to +8.6‰ (mean = +5.8‰, *n* = 30), which is similar to but more positive than the δ^{34} S values of sulfides from the majority of magmatic hydrothermal deposits (-3 to +1‰; Hoefs, 2009). The calculated δ^{34} S_{H2S} values of ore-forming fluids in equilibrium with the sulfides range from 5.2 to 7.1‰ (average of 6.1‰), which lay between the values of magmatic sulfur and sedimentary sulfur (Ohmoto and Rye, 1979). This suggests that sulfur in the Aerhada deposit was derived from local magmatic and sedimentary sources.

Except from our lead isotopic data, we have used previously published Pb isotopes from the Aerhada Pb-Zn-Ag deposit, as well as the available ones from the local tuffs, sandstones, Triassic diorites, and Jurassic to Cretaceous granites (Zhang et al., 2007a,b, 2012; Nie et al., 2007a,b) (Table 4, Fig. 9). From the ²⁰⁶Pb/²⁰⁴Pb versus ²⁰⁷Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb binary diagrams (Fig. 9, after Zartman and Doe, 1981) the majority of Pb isotopic compositions of the Aerhada sulfides plot in the field between the upper crust and orogen. Notably, the Pb isotope compositions of the sulfides plot between the local Mesozoic magmatic (i.e., granite) and the sedimentary rocks (i.e., sandstone) (Fig. 9). This suggests that lead and possibly silver, due to their close geochemical association, associate with these sources.

The ³He/⁴He ratios (2.5 to 5.4 Ra) of the ore-forming fluids which relate to the formation of Pb-Zn-Ag ores in stage II are much higher than those attributed to the crust (0.01-0.05 Ra; Stuart et al., 1995) and very close to the ones of the mantle (6-7 Ra; Burnard et al., 1999). This suggests that the ore-forming fluids mostly contain mantle helium. In the ³He versus ⁴He (Fig. 10a) and ³He/⁴He versus ⁴⁰Ar/³⁶Ar (Fig. 10b) diagrams, all the obtained data fall close or in the mantle He field, indicating a significant contribution of mantle helium for the Aerhada ore system. Furthermore, the noble gas isotopic data from the Aerhada deposit are consistent with numerous Pb-Zn-Ag ore deposits from the central and southern segment of the GHR (Wang et al., 2008), reflecting that those hydrothermal deposits might share some similar processes for their genesis. Considering their tectonic setting, a large number of the hydrothermal deposits formed in NE China are related to the subduction of Paleo-Pacific plate and some with the Paleo-Asian plate subduction (i.e., the Duobaoshan Cu deposit, Zeng et al., 2014). In such a tectonic setting, the possible addition of radiogenic helium from the subducted crustal materials into the hydrothermal system would lead helium isotopic ratios for the mantle lower than typical MORB, e.g., 8 Ra (Ozima and Podosek, 2002). In addition, we have calculated the contribution of the mantle helium for the Aerhada ore system utilizing the equation of Kendrick et al. (2001). Our results suggest that the mantle-derived helium contributed in the Aerhada mineralizing fluid from 32 to 69% (average of 53%).

6.3. Implications for Pb-Zn-Ag mineralization

During the evolution of the Aerhada hydrothermal system, both temperature and salinity decrease, which is consistent with a fluidmixing and dilution scenario (Zhai et al., 2013). This also suggests that the coupled drop of temperature and salinity of the ore fluids provoked the deposition of minerals from stages I to III, i.e., arsenopyrite, pyrite, Pb-Zn-Ag minerals, quartz, fluorite and calcite. The ore-forming fluids ascended along the NE- and NW-trending extensional faults and fractures and then reacted with host rocks, i.e., the Angeryin Wula sandstones. Wall-rock interaction changed their physicochemical parameters and isotopic

 Table 4

 Lead isotope compositions of sulfides and local rocks from the Aerhada Pb–Zn–Ag deposit.

No.	Sample No.	Sulfide/Rock	²⁰⁸ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁶ Pb/ ²⁰⁴ Pb	No.	Sample No.	Sulfide/Rock	²⁰⁸ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁶ Pb/ ²⁰⁴ Pb
1	14AEHD-17	Galena	38.458	15.65	18.439	24	TW10	Pyrite	37.653	15.379	18.174
2	14AEHD-18	Galena	38.4	15.641	18.427	25	TW2	Pyrite	37.874	15.47	18.281
3	14AEHD-23	Galena	39.298	15.75	18.684	26	TW3	Pyrite	37.909	15.484	18.293
4	14AEHD-30	Galena	39.301	15.75	18.685	27	TW5	Pyrite	37.896	15.478	18.291
5	15AEHD-18	Galena	38.565	15.683	18.459	28	AR3	Tuff	38.092	15.535	18.366
6	15AEHD-23	Galena	38.172	15.547	18.398	29	AR5	Tuff	38.095	15.541	18.348
7	14AEHD-34	Arsenopyrite	38.421	15.64	18.429	30	AR6	Tuff	38.035	15.516	18.349
8	TW1	Galena	37.954	15.499	18.316	31	AR7	Tuff	37.997	15.51	18.325
9	TW4	Galena	37.899	15.479	18.295	32	AR28	Tuff	37.983	15.505	18.32
10	TW7	Galena	37.693	15.391	18.19	33	AR33	Tuff	38.01	15.515	18.333
11	TW8	Galena	38.161	15.585	18.415	34	AR48	Tuff	38.217	15.578	18.37
12	TW9	Galena	37.705	15.393	18.187	35	CBL Q09	Sandstone	38.103	15.703	18.495
13	TW11	Galena	38.211	15.593	18.415	36	CBL Q10	Sandstone	38.122	15.716	18.501
14	TW7	Sphalerite	37.672	15.385	18.187	37	CBL Q13	Granite	37.502	15.301	18.124
15	TW8	Sphalerite	37.673	15.384	18.174	38	CBL Q12	Granite	37.5	15.298	18.12
16	TW9	Sphalerite	37.755	15.409	18.198	39	CBL Q11	Granite	37.498	15.278	17.996
17	TW10	Sphalerite	38.213	15.602	18.431	40	CG2	Diorite	38.12	15.512	18.335
18	TW11	Sphalerite	37.962	15.5	18.317	41	CG5	Diorite	37.991	15.5	18.246
19	TW4	Arsenopyrite	37.842	15.454	18.258	42	CG6	Diorite	38.108	15.523	18.381
20	TW7	Arsenopyrite	37.898	15.48	18.297	43	CG8	Diorite	38.085	15.529	18.529
21	TW11	Arsenopyrite	38.191	15.594	18.427	44	CG3	Diorite	37.831	15.465	18.172
22	TW8	Pyrite	37.673	15.37	18.153	45	CG7	Diorite	37.961	15.488	18.22
23	TW9	Pyrite	37.689	15.387	18.181						

Note: Number 1-7 data are from this study, 8-34 data are from Zhang et al. (2007a,b), 35-39 data are from Nie et al. (2007a, b) and 40-45 data are from Zhang et al. (2012).

Table 5He and Ar isotopic compositions $(10^{-7} \text{ cm}^3 \text{ STP g}^{-1})$ of fluid inclusions hosted in sulfides from the Aerhada Pb–Zn–Ag deposit.

Sample No.	Mineral	⁴ He	³ He/ ⁴ He	⁴⁰ Ar	⁴⁰ Ar/ ³⁶ Ar	³ He/ ⁴ He (Ra)	⁴ He _{mantle} (wt.%)
14AEHD-17	Pyrite	0.403	69.38	1.173	301.2	4.96	63.00
14AEHD-23-2	Galena	0.135	75.70	0.372	290.6	5.41	68.76
14AEHD-29	Galena	0.151	35.04	0.538	297.0	2.50	31.73
14AEHD-30	Galena	0.318	54.88	0.605	295.3	3.92	49.80

Note: Ra is the ${}^{3}\text{He}/{}^{4}\text{He}$ ratios of the atmosphere (1.4 \times 10⁻⁶, Mamyrin and Tolstikhin, 1984); ${}^{4}\text{He}_{mantle}$ values represent weight percent of mantle helium relative to crustal helium.



Fig. 9. ²⁰⁶Pb/²⁰⁴Pb versus ²⁰⁷Pb/²⁰⁴Pb (a) and ²⁰⁶Pb/²⁰⁴Pb versus ²⁰⁸Pb/²⁰⁴Pb (b) plots of sulfides and various rocks from the Aerhada deposit. Data are collected from Zhang et al. (2007a,b, 2012) and Nie et al. (2007a, b). The Pb isotope curves for crust, mantle and orogenic belt are based on Zartman and Doe (1981).

compositions and facilitated also the ore fluid to leach out variable amounts of metals and sulfur from the host sedimentary rocks. Influx of the meteoric waters in the conduits led to fluid mixing and dilution which destabilized the sulfide ligands and resulted in their precipitation.

The obtained oxygen, hydrogen, sulfur, and lead isotopic compositions from the Aerhada deposit suggest that the ore fluids are most likely derived from a deep magma and modified due to mixing and dilution with meteoric waters. The sulfur and metals, i.e., lead and probably silver originate from both local magmatic and sedimentary sources. Furthermore, the noble gas isotopes indicate that the ore fluids have a deep mantle origin, which is consistent with the regional tectonic setting. The GHR geodynamic evolution involved large-scale oceanic plate subduction and subsequent lithospheric delamination and extension. In such an extensional setting, the regional NE- and NW-trending faults and fractures are used as channels which transported the ore fluids (Gao and Qian, 2005; Tao, 2006).



Fig. 10. (a) ³He versus ⁴He (modified from Mamyrin and Tolstikhin, 1984) and (b) ³He/⁴He versus ⁴⁰Ar/³⁶Ar plots (modified from Wang et al., 2015) of fluid inclusions hosted in sulfides from the Aerhada deposit. The dashed green regions with data from Wang et al. (2008) show the He-Ar isotope compositions for numerous hydrothermal Pb-Zn-Ag deposits from the central and southern segment of the Great Hinggan Range.



Fig. 11. Homogenization temperature (Th) versus salinity plots of ore fluids for the Aerhada deposit. Inset illustration for different fluid evolution paths is based on Shepherd et al. (1985).

As discussed above, our field, fluid inclusion and isotope data suggest that the Aerhada deposit shares numerous geological and geochemical characteristics with the majority magmatic-hydrothermal ore deposits in the southern segment of the GHR (Chu et al., 2001; Wang et al., 2004; Zhu et al., 2006; Han et al., 2013; Cong et al., 2014; Ouyang et al., 2014; Zhang et al., 2015). Therefore, based on ore deposit geology, fluid inclusion and isotope data, this deposit could be classified as a typical medium-low temperature hydrothermal vein type Pb-Zn-Ag deposit.

7. Conclusions

The conclusions from this study are summarized below:

- Fluid inclusion studies indicate that fluid mixing and dilution are inferred as the main mechanisms for ore deposition;
- (2) Multiple (D-O, S, Pb, He-Ar) isotopic compositions obtained from the deposit suggest that the ore fluids are most likely derived from deep-seated magma and modified by the addition of meteoric waters, and that the metals relate to both local magmatic and sedimentary sources;

(3) The Aerhada deposit can be classified as a typical medium to low temperature hydrothermal vein type Pb-Zn-Ag deposit.

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