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GR Focus Review

Metalliferous coal deposits in East Asia (Primorye of Russia and South China): A review of geodynamic controls and styles of mineralization



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ABSTRACT

Metalliferous coal deposits, mainly hosting Zr(Hf)–Nb(Ta)–REE and U(Mo,Se)–REE ores, in East Asia (Primorye of Russia and South China) primarily result from the evolution of plumes ascending from deep mantle and/or asthenospheric flows, both of which incorporate some reworking of the continental crust. This mantle–crust interaction not only led to coal-basin formation but also played a significant role in extensive volcanism and ore-generating hydrothermal activity. Three mineralization styles are identified in these deposits: tuffaceous, hydrothermal-fluid, and mixed tuffaceous–hydrothermal types. The tuffaceous Zr(Hf)–Nb(Ta)–REE deposits have source magmas with an alkali basalt composition, although felsic, mafic, and intermediate types of tuffaceous horizons have been identified in the study area. The mineralization occurred not only in the coal but also in the host rocks, and not only during peat accumulation but also during the later stages of coal development (including coal-flication and late epigenetic processes). Rare metals in the metalliferous coal deposits are generally either associated with clay and organic matter or occur as secondary minerals derived from decomposition of the primary magmatic rare-metal bearing minerals (e.g. Nb-bearing rutile) under the influence of organic acids and hydrothermal fluids. © 2015 International Association for Gondwana Research. Published by Elsevier B.V. All rights reserved.

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1. Introduction

The term "metalliferous coal" has been widely used to describe coals with practical or potential economic significance for valuable metal production (Hower et al., 2000; Meitov, 2001; Seredin and Finkelman, 2008; Dai et al., 2012; Mastalerz and Drobniak, 2012; Sia and Abdullah, 2012), as well as for the traditional use as an energy source. The term is typically applied to coals with unusually high concentrations of potentially valuable trace elements, typically at least 10-times higher than the respective averages for the same elements in world coals generally (Seredin and Finkelman, 2008). Recent studies have shown that the metals are enriched not only in the coal themselves but also in their host rocks (roof and floor strata) and in non-coal bands or partings within the coal seams (Seredin and Finkelman, 2008; Dai et al., 2010; Zhao et al., 2013; Dai et al., 2014a).

The concentrations of rare metals in the ashes of some metalliferouscoals are equal to or higher than those found in conventional types of rare metal ores (Seredin and Finkelman, 2008; Seredin and Dai, 2012) and much higher than those in common coal ashes and coalassociated sedimentary rocks (Ketris and Yudovich, 2009). For example, weathered-crust elution-deposited ores with ion-exchangeable rare earth elements generally have total rare earth element contents of 0.03-0.25% (Bao and Zhao, 2008; Chi and Tian, 2008), but the concentrations of rare earth elements in common coal ashes average around 0.035% (Ketris and Yudovich, 2009) and 0.1-1.5% in the ashes of metalliferous coals (Seredin and Dai, 2012). Uranium concentrations are around 0.0016% in the ashes of common coals (Ketris and Yudovich, 2009), but 0.1-0.2% in conventional sand-hosted roll-type deposits and up to 0.4% in the ashes from metalliferous-coal deposits (Dai et al., 2015a,b). Concentrations of Zr and Nb are 210 and 20 µg/g respectively in average coal ashes (Ketris and Yudovich, 2009), but reach over 2000 and 200 μ g/g respectively in ashes from coal-hosted Zr(Hf)– Nb(Ta)–REE ore deposits (this paper).

Metalliferous coals have been used as sources for metal extraction for over 100 years, since the coals of Wyoming and Utah were used for Au and Ag recovery in the late 19th and early 20th centuries (Jenney, 1903; Stone, 1912). Metalliferous coal deposits containing high concentrations of Ge, Ga, rare earth elements and Y (REY, or REE if Y is not included), Li, Sc, Zr, Hf, Nb, and Ta have recently attracted significant attention as a new source for these rare metals, which are becoming increasingly important in various high-technology applications (Seredin, 2012a; Seredin et al., 2013; Arbuzov et al., 2014). For example, germanium is currently being extracted as a raw material from three Ge-bearing coal deposits: Lincang (Yunnan province of China), Wulantuga (Inner Mongolia of China), and Spetzugli (Primorye, Russian Far East). Together, the latter deposits account for more than 50% of the total annual industrial Ge production in the world (Seredin et al., 2013; Dai et al., 2014b). The Ge in the three coal-hosted Ge ore deposits was derived from the granite basement by hydrothermal solutions during syngenetic and diagenetic stages, and is organically bound (Zhuang et al., 2006; Seredin and Finkelman, 2008; Qi et al., 2011; Dai et al., 2014b,c). A pilot plant for extraction of Ga and Al from coal combustion products (fly ashes) was installed at the Jungar deposit, Inner Mongolia, China (Dai et al., 2012; Seredin, 2012b) at the beginning of 2011. The annual processing capacity of this plant is 800,000-t Al₂O₃ and approximately 150-t Ga (Dai et al., 2012; Seredin, 2012b). Intensive research on resource evaluation and on the origin of the REY, Li, Sc, Zr, Hf, Nb, Ta, and U enrichment in these metalliferous coal deposits (both coals and host rocks), as well as methods for their extraction from coal ashes, have been and are currently being conducted in the USA (Finkelman and Brown, 1991; Hower et al., 1999; Mardon and Hower, 2004; Mastalerz and Drobniak, 2012), Europe (Eskenazy, 1987a,b; Blissett et al., 2014; Yossifova, 2014), Australia (Jaireth et al., 2014), India (Prachiti et al., 2011; Saikia et al., 2015), and, in particular, China (Dai et al., 2010; Wang et al., 2011; Dai et al., 2012; Seredin and Dai, 2012; Zhuang et al., 2012; Sun et al., 2013; Zhao et al., 2013; Dai et al., 2014a,b,c) and Russia (Seredin, 1991, 1996; Arbuzov et al., 2000; Kryukova et al., 2001; Arbuzov et al., 2003; Seredin, 2004; Seredin et al., 2006; Seredin and Tomson, 2008; Arbuzov et al., 2011; Seredin et al., 2013; Arbuzov et al., 2014).

On the other hand, coal-hosted rare-metal ore deposits generally contain high concentrations of toxic trace elements, which could have adverse effects on human health and environments. For example, fly ashes derived from the three giant coal-hosted Ge deposits, Lincang (Yunnan, China), Wulantuga (Inner Mongolia, China), and Spetzugli (Primorye, Russia), are highly enriched in toxic trace elements, including up to (on an organic-free basis) 2.12% As, 1.56% F, 1.22% Sb, 0.55% Pb, 0.04% Be, 0.017% Tl, and 0.016% Hg (Dai et al., 2014b). These high elemental concentrations in the fly ashes are due to their high levels in the raw coals (Zhuang et al., 2006; Qi et al., 2011; Dai et al., 2014c). The coal-hosted U-Se-Mo-Re-V ore deposits preserved within marine carbonate successions in South China are highly enriched in organic sulfur (e.g., 8.77%–10.30% in Yanshan coals, Yunnan, South China; Dai et al., 2008), F (up to 3362 µg/g in Heshan coals, Guangxi, South China; Dai et al., 2013b), Hg (654 ng/g in the Fusui coals, Guangxi, South China; Dai et al., 2013a), as well as Cr, Ni, and Cd (Shao et al., 2003; Zeng et al., 2005; Dai et al., 2015b; Liu et al., 2015). The Late Permian coal in the Huayingshan Coalfield in southwestern China is considered to be a coal-hosted Zr-Nb-REE ore deposit (Zhuang et al., 2012; Dai et al., 2014a) and is enriched in Se (6.99 μ g/g; Dai et al., 2014a). Additionally, high concentrations of V, Cr, Co, and Ni in the Late Permian coals of southwestern China were derived from the Emeishan basalts, which served as a sediment source region during peat accumulation (Zhou et al., 2000; Dai et al., 2012; Wang et al., 2012; Zhuang et al., 2012). The enrichment of toxic trace elements in the coals is usually attributed to a combination of geological processes and tectonic controls during peat accumulation, as well as during subsequent diagenetic and epigenetic activities (Ding et al., 2001; Sia and Abdullah, 2012; Yossifova, 2014; Dai et al., 2015b). Thus, investigations of the relationships between tectonic background (e.g., mantle plume formation) and toxic trace elements in coal and in coal-hosted ore deposits might provide a better understanding of the environmental influences of toxic elements during rare metal recovery from coal ash and during coal combustion.

From genetic and practical points of view, the relation between tectonic setting (e.g., mantle plume formation or subduction/collision-related processes) and the development of coal-bearing strata (including coal-hosted ore deposits) may not only provide geologic information about the formation of coal-bearing sequences and regional tectonic history to assist exploration and exploitation of coal resources, but may also assist in the exploration for rare metal ore deposits. This is because the distribution of both coal beds and coal-hosted ore deposits resulted from the same processes of peat/sediment accumulation and rank advance, and were further influenced by interaction between the organic matter and basinal fluids, sediment diagenesis, and synsedimentary volcanic inputs (Ren, 1996; Ruppert et al., 1996; Ward, 2002).

As an example of these interactions, thin beds of kaolinite-rich sediment (tonsteins) of pyroclastic origin, deposited in the original peat-forming environment, have been found in many coal seams in southwestern China (Zhou et al., 1982, 2000; Dai et al., 2011, 2014a) and Russia (Admakin and Portnov, 1987; Admakin, 1991). The tonsteins in the Late Permian coals of that region probably resulted from waning activity of a mantle plume and may serve to indicate the periphery of the Emeishan Large Igneous Province (Dai et al., 2011, 2014d). Some tonsteins, typically of alkali origin, may contain valuable trace elements that could be potentially utilized, or may serve as indicators in exploration for alkali ore deposits (Zhou et al., 2000; Spears, 2012; Dai et al., 2014a). In many cases, the tonsteins contain primary sanidine and/or zircon of magmatic origin that could be used for radiometric age determination, providing absolute ages for chrono-stratigraphic correlation within the globally accepted geologic timescale (Bohor and

Triplehorn, 1993; Burger and Damberger, 1985; Hill, 1988; Knight et al., 2000; Kunk and Rice, 1994; Hess et al., 1999; Lyons et al., 1992, 2006; Guerra-Sommer et al., 2008; Maksimov and Sakhno, 2011); tonsteins may be incorporated with mined coal products and, if not removed when the mined coal is beneficiated (e.g., mineral-rich particles in the crushed coal are removed by density separation in an associated preparation plant), may also become part of the coal product when it is used (Ward, 2002).

Although there have been many studies on metalliferous coal deposits, these are mainly focused on the origin and mode of occurrence of the rare metals involved. However, some critical questions, such as the tectonic controls on the deposits, have rarely been studied, and the forms of rare-metal mineralization have never been summarized. A better understanding of these critical questions is nevertheless of great importance for the exploration and economic evaluation of metalliferous coal deposits, and for controlling environmental pollution from release of toxic trace elements during rare metal recovery. In this paper, we discuss the tectonic controls on the localization of the metalliferous coal deposits and the mechanisms of rare-metal mineralization in the world's largest coal-hosted ore districts (South China and South Primorye of Russia; Fig. 1). It should be mentioned that the two regions have not been studied in equivalent depth; the South China deposits are much better characterized by geochemical data than the Russian deposits, although some gaps in the data still exist. Additionally, isotopic analyses of the tuffaceous ores are absent in both regions.

2. Regional tectonic and magmatic control of ore deposits

East Asia has more coal-hosted rare-metal ore deposits than anywhere else in the world (Seredin and Finkelman, 2008; Arbuzov et al., 2014). For example, in addition to Nb(Ta)–Zr(Hf)–REE and U(Mo,Se)– REE ore deposits, several large coal-hosted Ge ore deposits, each of which contain Ge reserves of more than 1000 t, occur in the Primorye and Sakhalin areas of Russia, as well as the Inner Mongolia and Yunnan provinces of China. The two regions of coal-hosted ore deposits, South Primorye and South China, are characterized by their circular, curved and tapering shapes, respectively (Fig. 1). Coal-hosted rare-metal



Fig. 1. Location of the coal-hosted rare-metal ore deposits in East Asia: (1) South Primorye, (2) South China. The red dotted lines are the boundaries of the Ussuri and Emeishan concentric structures. Data complied from Xu et al. (2001), He et al. (2010), and Seredin and Tomson (2008).

deposits are widely distributed in these two regions (Seredin and Finkelman, 2008).

The two rare-metal regions have different sizes (Fig. 1), ages of formation (mainly Late Permian age for south China and Cenozoic age for South Primorye), and different sedimentary environments for the coal-bearing strata (continental, continental-marine transitional, and marine facies for south China; continental environment for South Primorye). However, their distributions are at least partly controlled by local concentric and radial faults, represented by the Ussuri and Emeishan structures, respectively. The Emeishan structure is associated with the well-known Emeishan large igneous province (ELIP), which is considered to be mantle plume-related (Xu et al., 2008; Xu et al., 2010; Y. Sun et al., 2010 and references therein). The Ussuri structure is also associated with flood basalts, forming the Shufan, Shkotovo, and Sandugan plateaus in addition to numerous smaller basaltic fields that may also be plume-related (Seredin and Tomson, 2008). A number of coal-bearing basins were developed during the formation and evolution of these structures; and were accompanied by extensive volcanism (Zhou et al., 1982; Dai et al., 2011) and hydrothermal activity (Zhou and Ren, 1992; Seredin and Mokhov, 2007; Seredin, 2012a; Dai et al., 2013a,b), which in turn were associated with the formation of raremetal ore deposits. The rare-metal ore deposits are mainly of volcanicash, hydrothermal-fluid, and mixed volcanic-ash and hydrothermalfluid origin (Seredin and Dai, 2012; Dai et al., 2013a,b, 2014a,c).

However, the plume-like processes did not only influence the formation of the metalliferous coal deposits. Their host basins were likely initiated by widespread within-plate extension and subsidence of the South China region in the Permian-Triassic (He et al., 2003; Peate and Bryan, 2008; Y. Sun et al., 2010), and within-plate extension associated with strike-slip displacements and rift-related volcanism in the Cenozoic for East Asia (Ren et al., 2002; Yin, 2010). In this scenario, localized plume-like processes created the dome-like radial and concentric fault systems, providing channels for voluminous magmatic products and making small depressions at the Earth's surface. These depressions were sites for subsequent accumulation of terrigenous and volcanoclastic sediments at certain stages of regional evolution (Seredin and Tomson, 2008). Perhaps both the regional extension and the more local plume-like mantle upwellings had a common deepseated cause, which was also responsible for the rare-metal mineralization. This review suggests that asthenospheric flows reworking continental crust rich in rare metals could be the cause, although deep mantle plumes should not be completely excluded from consideration at the current stage of investigations.

2.1. South Primorye

2.1.1. General characteristics of the Ussuri concentric structure and its regional position

This coal-bearing district is located within the Ussuri structure, which is approximately 150 km in diameter (Seredin and Tomson, 2008). Faults within it form a concentric geometry that includes numerous Cenozoic coal-bearing depressions, small extrusions of alkaline volcanics, and large-volume basaltic flows (Fig. 2). The latter, including the Shufan, Shkotovo, and Sandugan Plateaus, made-up of Neogene intraplate basalts, has a total volume at least 1700 km³ (Rasskazov et al., 2003) that may indicate a plume association for the Ussuri structure (Seredin and Tomson, 2008).

On the other hand, Paleogene–Neogene coal basins, associated basaltic fields, and small extrusions of alkaline rocks are widespread in East and Central Asia (Fig. 3). Their distribution as a whole is almost linear, controlled by regional strike-slip faults, with concentrations of the basins and volcanics at fault intersections and bends. Cenozoic volcanic fields, including Neogene–Quaternary basaltic plateaus, occur in many places throughout the Far East, including South Primorye (Rasskazov et al., 2003; Okamura et al., 2005; Chashchin et al., 2007, 2011).

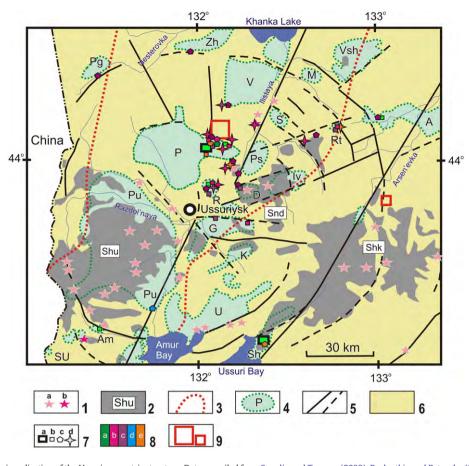


Fig. 2. Cenozoic geology and mineralization of the Ussuri concentric structure. Data compiled from Seredin and Tomson (2008), Pavlyutkin and Petrenko (2010), Maleev (1958), Sakhno et al. (2004), and Maksimov and Sakhno (2008, 2011). (1) small extrusions of the Neogene alkaline and subalkaline (a), and Eocene adakitic (b) rocks; (2) Miocene–Pliocene flood basalts with the plateau names: Shu – Shufan, Shk – Shkotovo, Snd – Sandugan; (3) main distribution of the Oligocene–Miocene felsic volcanoclastics; (4) Paleogene–Neogene coal-bearing basins partly covered by younger sedimentary and volcanic rocks and the basin names: (A) Arsen'evka, (Am) Amba, (D) Danilovka, (G) Glukhovka, (IV) Ivanovka, (K) Komarovka, (M) Merkushovka, (P) Pavlovka, (Pg) Pogranichnaya, (Ps) Poiskovaya, (Pu) Pushkinsk, (Sh) Shkotovo, (S) Sibirtsevo, (SU) Siny Utes, (R) Rakovka, (Rt) Rettikhovka, (U) Uglovka, (V) Vadimovka, (Vsh) Vishnyakovka, (Zh) Zharikovo; (5) faults figured based on geological mapping, satellite images, and relief; (6) pre-Cenozoic basement consisting of Early Paleozoic crystalline rocks intruded by Mesozoic granites and partly covered by Mesozoic coal-bearing deposits; (7) Cenozoic ore occurrences: (a) metalliferous coal deposits, (b) metalliferous coal occurrences; (c) ore occurrences in Cenozoic volcano-sedimentary procks (outside coal seams); (d) ore occurrences in the argillized dikes and breccias; (8) main metals: (a) Ge, (b) REE, (c) U, (d) Nb–Ta–Zr(Hf)–REE–Ga, (e) Au (PGE); (17) Nb–Ta–Zr(HF)–REE–Ga, (e) Au (PGE); (17) Nb–Ta–Zr(HF)–REE–Ga occurrence; (9) Paleozoic fluorine-rare metal deposits including the Voznesenka district of F, Sn, W, and Nb–Ta deposits (larger) and Poperechka Nb–Ta deposit (smaller).

The geochemistry of the volcanic rocks in these fields indicates distinctive contributions from continental crust, Indian-type (I-type) asthenospheric mantle, and subducted plate materials (Flower et al., 1998; Rasskazov et al., 2003; Okamura et al., 2005; Chashchin et al., 2007; Guo et al., 2009; Chashchin et al., 2011). These relationships are illustrated in Fig. 4 (143Nd/144Nd vs 87Sr/86Sr plot) where the volcanic rocks of Primorye and NE China clearly overlap the fields for arc and continental adakitic rocks (Castillo, 2012; and references therein), MORB and fossil spreading center lavas (Flower et al., 1998), Yellowstone basalts (indicating some plume input; Hanan et al., 2008; Ellis et al., 2013) and the Dabie Precambrian metamorphic complex (Ma et al., 1998). Additionally on this figure, Oligocene-Miocene volcaniclastics from the southern Primorye coal basins overlap the field for the Palaeozoic gabbro-granite and related fluorine-rare metal mineralization from southern Primorye (Kupriyanova et al., 2005) suggesting incorporation of this material. It is important for our study that the EM-1 enrichment (DUPAL anomaly) is characteristic of the Indiantype asthenoshere of the southern oceans in contrast to the North Pacific (Hart, 1984). Based on this criterion, Flower et al. (1998) traced the mantle flows from the India-Eurasia collision zone far to the east and northeast using geochemical data on Late Cenozoic basalts from East Asia and the West Pacific. As can be seen in Fig. 4, the Cenozoic basaltic and andesitic rocks of S Primorye and NE China (Okamura et al., 2005; Popov et al., 2008; Martynov and Khanchuk, 2013), including adakitic rocks (Guo et al., 2009; Chashchin et al., 2011), encompass a field between a subducted plate component (i.e. arc adakites) and the East Asia continental crust. The EM1 component suggesting the mantle flows from the India-Eurasia collision likely shifted this field slightly down.

In general, the Cenozoic volcanic rock assemblages in East Asia indicate reworking of the continental lithosphere, which resulted from asthenospheric upwellings above the deeply subducted oceanic slab and upper mantle flows induced by the India-Eurasia collision, rather than a mantle plume derived from the core-mantle boundary (Flower et al., 1998; Pirajno and Santosh, 2014). This is also supported by deep seismic images that show hot upper mantle below modern volcanoes, for example the Changbai volcano (Fig. 3), and above a colder slab stagnating in the mantle transition zone (Li and van der Hilst, 2010; and references therein). The Cenozoic magmatic evolution of the region shows an increasing influence of a within-plate component and a decreasing contribution from the subducted plate component with time (Martynov and Khanchuk, 2013). The Late Cenozoic (15 Ma and younger) within-plate volcanics, which may only be considered as plume-related, consist of voluminous plateau basalts, with ages from 15 to 13 Ma, and small-volume younger volcanics, the composition of which varies from continental tholeiite to phonolite and other alkaline rocks. Sapphire-zircon placers are commonly associated with these volcanics (Graham et al., 2008; Nechaev et al., 2009). It should also be

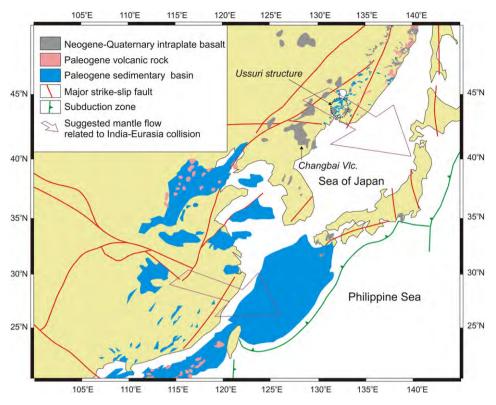


Fig. 3. Distribution of Cenozoic volcanic and sedimentary complexes in East Asia (compiled from Ren et al., 2002; Pavlyutkin and Petrenko, 2010; Martynov and Khanchuk, 2013) showing the suggested mantle flows related to the India–Eurasia collision (Flower et al., 1998).

mentioned that some alkaline rocks of the modern Changbai volcano (Fig. 3) are enriched in Zr (up to 2404 μ g/g) and Nb (up to 217 μ g/g) (Popov et al., 2008), suggesting that this magmatic complex can produce rare-metal mineralization. Meanwhile, some interbeds of the Oligocene–Miocene felsic volcanoclastics also contain moderately high concentrations of rare metals (Zr up to 1160 μ g/g, Nb up to 145 μ g/g). Their plots in Fig. 4 clearly overlap the field for Paleozoic gabbro–granite complex and related fluorine-rare metal mineralization of S Primorye, demonstrating that they are derived from the local crust, which is rich in rare metals. A characteristic and very distinctive feature of the volcanoclastic source is wide variation in 87 Sr/ 86 Sr but almost uniform

 $^{187}\rm Nd/^{186}\rm Nd$ ratio. Other magmatic and metamorphic formations of East Asia show obvious decreases in $^{187}\rm Nd/^{186}\rm Nd$ with increasing $^{87}\rm Sr/^{86}\rm Sr$ (Fig. 4).

Thus, the Ussuri concentric structure should be considered as a local feature of the more extensive volcanic belt and associated pull-apart basins at the back of the convergent plate boundaries. Based on the radial fault system, especially manifested in the present-day river network, the structure is a gently sloping dome.

The above-mentioned East Asian volcano-sedimentary belt started forming in the Paleocene–Eocene, as indicated by small intrusions of deep adakitic magmas in NE China (55–58 Ma, Guo et al., 2009 and

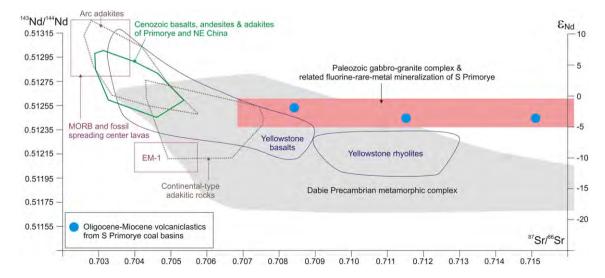


Fig. 4. ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd ratios for Cenozoic volcanic rocks of Primorye and adjacent regions. The reference fields are shown after Castillo (2012), Guo et al. (2009), Ma et al. (1998), Kupriyanova et al. (2005), Hanan et al. (2008), Popov et al. (2008), Ellis et al. (2013), Martynov and Khanchuk (2013). The data for Cenozoic felsic volcanoclastics are from Maksimov and Sakhno (2011) (one analysis) and two unpublished analyses contributed by S.O. Maximov (Far East Geological Institute).

references therein) and S Primorye (Rb–Sr age 46.2 \pm 0.5 Ma, Rasskazov et al., 2003; K–Ar age 46.7 \pm 1.1 Ma, Seredin, 2010; and 45.52 \pm 1.1 Ma, Chashchin et al., 2011), and the initiation of the first coal-bearing basins (Uglovka and Rettikhovka) at their margins (Pavlyutkin and Petrenko, 2010).

2.1.2. Evolution of the Ussuri concentric structure

The evolution of the Ussuri structure can be divided into six stages (Fig. 5):

- Early–Middle Eocene adakitic rocks of the Poiskovaya and Amba basins (Fig. 2; Seredin, 2010; Chashchin et al., 2011) and argillized andesite–dacite lavas of the Rettikhovka Basin (Seredin and Chekryzhov, 2012a);
- (2) Late Eocene-Early Oligocene argillized rocks (mafic lavas?) of the Pavlovka (Pavlovka-2 and Vostochny open-casts) Basin (Seredin and Chekryzhov, 2012a);
- (3) Late Oligocene-Early Miocene mafic volcanics of the Amba (Popov et al., 2007) and Siny Utes (Popov et al., 2005) basins;
- (4) Late Oligocene–Late Miocene felsic tuffs and small intrusions of K-rich basic rocks in the Siny Utes, Pavlovka, Pushkinsk, Uglovka, and other basins (Sakhno et al., 2004; Popov et al.,

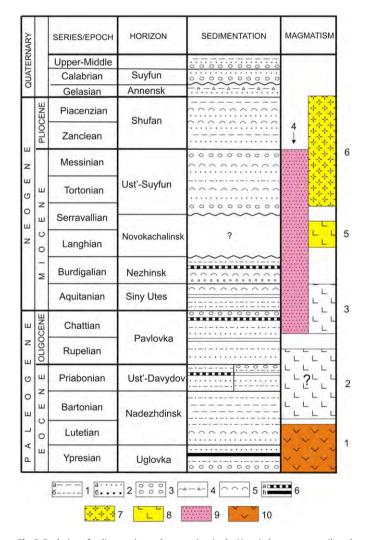


Fig. 5. Evolution of sedimentation and magmatism in the Ussuri plume structure (based on the stratigraphic scheme of Pavlyutkin and Petrenko (2010). 1 -Claystone (a) and siltstone (b); 2 - sandstone (a), and conglomerate (b); 3 - pebble conglomerate; 4 - rock debris in clay; 5 - tuff and tuffite; 6 - coal (a), carboniferous siltstone and lignite (b); 7 - alkaline and associated volcanics; 8 - basalt; 9 - felsic tuff; 10 - adakitic and associated rocks.

2006; Chekryzhov, 2009; Chekryzhov et al., 2010; Pavlyutkin and Petrenko, 2010);

- (5) Middle Miocene (15–13 Ma) plateau basalts of the Shufan and Shkotovo Plateaus (Rasskazov et al., 2003; Maksimov and Sakhno, 2008b) and the final formation of the concentric structure (small plume?) above the subducted slab; and
- (6) Late Miocene–Pliocene (11–3 Ma) basaltic and alkaline volcanics of the Shufan and Shkotovo plateaus (Chashchin et al., 2007).

During stages 1–3, the upper continental crust was broken into pullapart depressions separating basement highs. These depressions later became sites for accumulation of coal-bearing sediments. The largest basins (Pushkinsk, Pavlovka, Glukhovka) were formed in the central part of the area. Volcanism was concentrated almost completely within and adjacent to the depressions, and was diverse in composition: bimodal volcanics (38–32 Ma) and Ti-rich basalts of the Amba Basin and alkaline (K-rich) basalts, andesite, and rhyodacite (24–22 Ma) of the Siny Utes Basin.

Stages 4–6 started with extensive eruptions of felsic tuffs and K-rich basic rocks, followed by within-plate basalts. The previously-formed organic-rich sediments were abruptly replaced by coarse terrigenous and volcanogenic deposits (Pavlyutkin and Petrenko, 2010). Widespread felsic volcanism of the Ust' Suyfun Formation during stage 4 is considered to have resulted from melting of the upper crust under the influence of the ascending mantle diapir (plume?), the main source of the later within-plate basalts (Popov et al., 2007). According to their position below the flood basalts and the similar distribution of their $\frac{87}{5}$ Sr/ $\frac{86}{5}$ r and $\frac{143}{144}$ Nd ratios (Fig. 4), these may be compared to the central Snake River Plain rhyolite ignimbrites of the Yellowstone plume (Ellis et al., 2013). Further, Yellowstone hydrothermal fluids have partly leached REE from the volcaniclastic rocks and redistributed them in the area (Lewis et al., 1995), making this plume environment even more similar to that of South Primorye in the Oligocene-Miocene time (Seredin et al., 2006). This similarity, however, merely indicates a significant crustal contamination (Hanan et al., 2008; Ellis et al., 2013) and should not be considered as solid evidence for a deep plume origin.

2.1.3. Ore mineralization of the Ussuri structure

There were at least six volcanic pulses during the development of the Ussuri structure, with associated extensive hydrothermal activity and significant related mineralization (Table 1). Two large wellknown (Spetzugli and Shkotovo) and many smaller coal-hosted Ge deposits of hydrothermal origin occur in this region. In addition, numerous occurrences of REE mineralization in coal and host rocks have been found in this area, including the unique Abramovka occurrence (Seredin, 1998; Seredin et al., 2009). The Rakovka uranium deposit of hydrothermal origin (Kokovkin, 2006) and associated mineralization also occur in this region. It is possible that additional U-REE deposits will be found with further investigation, although, to date, mineralization consisting only of Ga, Li, Zr, and Nb (which are commonly associated with REE) mineralization has been reported by the local geological survey. The Zr-Nb-REE association is of the tuffaceous and mixed tuffaceous-hydrothermal mineralization styles (Seredin et al., 2011; Seredin and Chekryzhov, 2012a).

It is likely that Cenozoic ore mineralization in the South Primorye coal basins is related to the extensive uppermost Paleogene to Miocene volcanoclastics and small alkaline (K-rich) extrusions, showing a significant crustal contribution and initiated by mantle flows related to the India-Eurasia collision (Flower et al., 1998). This magmatism and associated hydrothermal activity might have redistributed the rare-metal and fluorine crustal mineralization that is well-known in South Primorye, where it is hosted by the Paleozoic granite and gabbromonzonite complexes (Sato et al., 2003; Kupriyanova et al., 2005).

Table 1 Ore types of Cenozoic depres

Ore types of Cenoz	zoic depressions in	n South Primorye.
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Metal	Host rock	Shape of ore body	Ore grade	Mineralogy	Depression
Ge	Coal, carbonaceous mudstone and	Mantle- and	50–3000 μg/g	OC, Ism	P, R, Sh, Am
	sandstone	dome-like	(in coal)		А
	Fine sandstone	Stratiform	20–100 µg/g	Not studied	Р
	Clay (argillized Paleozoic rocks) in the basement	Cutting-linear	10–100 µg/g	Not studied	Р
REE	Coal	Mantle- and dome-like	0.1–1.5% (in ash)	Crb, Hcrb, Ohlg, Ox, Psp, Alpsp, OC	P, R, Rt, G
	Sandstone	Stratiform	0.1-0.2%	Crb, Alpsp	Р
	Clay (argillized Paleozoic rocks) in the basement	Mantle- and dome-like	0.05-0.2%	Crb, Alpsp	Р
	Clay	Stratiform	0.3-3.5% (in FMN)	Ox	Р
	Argillized dykes and breccias, fault gouge	Cutting-linear and tube-like	0.1–11%	Crb, Hcrb, Psp, Alpsp, Ox	P, R
Au (Ag, PGE) ^a	Coal and carbonaceous mudstone	Mantle- and dome-like(?)	n*0.01–n*1.0 µg/g (in ash)	Native Au, Pt, Ag, Au–Ag, Au–Cu, Pt–Fe, Pt–S, Ag–S, Ag–Cl, OC(?)	P, R, Sh
	Clay (argillized Paleozoic rocks) in the basement	Stratiform(?)	n*0.01–1.1 μg/g	Native Au	Р
	Sand and gravel	Stratiform	<i>n</i> *0.01– <i>n</i> *1.0 µg/g	Native Au, Au–Ag, Au–Hg, Au–Cu, Au–Cu–Sn, Au–Pb, Pd, Pd–Pt, Pt–Fe	Р
	Clay	Stratiform	<i>n</i> *0.01– <i>n</i> *1.0 µg/g	Native Au, Pd, Pd–Pt	Р
	Breccia	Cutting-linear and tube-like	Up to 150 µg/g	Native Au, Au–Ag, Au–Hg, Au–Cu, Au–Cu–Sn, Au–Sn–Pb–Ni, Pd, Pd–Pt	P, Rt
U	Coal	Mantle- and dome-like	0.1-0.4% (in ash)	Ox, Psp, OC	P, R, Rt, D, 0
	Sandstone	Stratiform	0.1-0.2%	Ox, Psp	P, R,
	Clay (argillized Paleozoic rocks) in the basement	Mantle- and dome-like	0.05-0.3%	Ox, Psp	P, R, D, A
	Argillized dykes and breccias, fault gouge	Cutting-linear and tube-like	0.1-1.5%	Ox, Psp, Sl	R
Nb(Ta)-Zr(Hf)-REE	Volcaniclastic interlayer	Stratiform	0.2-0.25% (total)	Psp, IS	Pu

Depressions: (Am) Amba, (A) Arsen'evka, (D) Danilovka, (G) Glukhovka, (P) Pavlovka, (Pu) Pushkinsk, (Sh) Shkotovo, (R) Rakovka, and (Rt) Rettikhovka.

Mineralogy: (Crb) carbonate, (Hcrb) halogenecarbonate, (Ohlg) oxohalogenid, (Ox) oxide, (Sl) silicate, (Psp) phosphate, (Alpsp) aluminophosphate, (OC) organic compound, (Ism) isomorphic admixture, (IS) Ion-sorption admixture, and (FMN) ferromanganese nodule.

^a The table presents contents of Au. Contents of Ag are up to 50–100 µg/g, Pt and Pd – up to a few µg/g.

A substantial additional investigation, however, including geochemical (especially isotopic) studies, is required to test this hypothesis.

2.2. South China

2.2.1. General characteristics of the Emeishan mantle plume

Rare-metal bearing coal deposits of Late Permian age are widely distributed in southern China, with a distribution controlled, like those in the South Primorye region, by a large (approximately 550 km in diameter) concentric structure. The distribution of rare-metal coal deposits in this region coincides with the extent of the Emeishan Large Igneous Province (ELIP; Fig. 6).

The ELIP is distributed in Yunnan, Sichuan, and Guizhou provinces in southwestern China (covering an area of more than 250,000 km²), and mainly consists of massive flood basalts and numerous contemporaneous mafic and felsic intrusions (Xu et al., 2001; He et al., 2010).

In spite of numerous studies, the origin of the ELIP is still controversial. It is conventionally considered to be a mantle plume-derived LIP (Xu et al., 2001; He et al., 2003, 2007; Xu et al., 2013). However, the latest review by Shellnutt (2014) concludes that such a model is far more complicated than previously suggested, and that the ELIP is likely to be derived from a plume-like upwelling of asthenospheric magmas. In addition, isotopic data suggest some crustal contamination and the presence of a sub-lithospheric mantle component in the system (Xu et al., 2001, 2008, 2010; Shellnutt, 2014; and references therein). Accordingly, the terms "plume" or "mantle plume" in this work mean a plume-like setting, although a "really deep" plume may not be completely excluded. The southwestern and northwestern flanks of the structure are cut by the Red River Fault and the Longmenshan Thrust, respectively (Fig. 6).

Despite these late collisional deformations, the main tectonic features of the Emeishan mantle plume overall remain as an intact whole; the western part of the province has the thickest volcanic succession, possibly reaching 5 km in thickness, whereas in the east the thickness is reduced to less than 100 m (Xu et al., 2008; Y. Sun et al., 2010; Xu et al., 2010; and references therein). The most common rock type is tholeiitic basalt, which occupies more than 95% by volume of the ELIP. The vast majority of the lavas have between 4 and 7 wt.% MgO, and additional volcanic rocks include picrites at various stratigraphic levels, mafic alkaline lavas at the base, and trachytes or rhyolites at the top of the sequence (Chung and Jahn, 1995; Xiao et al., 2004; Song et al., 2008). Intrusive rocks include mafic and ultramafic dikes (Shellnutt et al., 2008; Zi et al., 2008) and large layered intrusions (e.g., Zhou et al., 2005; Wang et al., 2006; Song et al., 2008).

The structure of the Emeishan mantle plume consists of three concentric zones (Fig. 6):

- (1) Central inner zone: the core was elevated at the time of the Middle/Late Permian boundary and surrounded by gravitational diamictites and coarse alluvial deposits.
- (2) Intermediate zone: the intermediate zone contains the greatest volume of Late Permian volcanics, with U–Pb ages between 267 and 252 Ma (Liu and Zhu, 2009). These comprise mafic lavas and basaltic tuffs, including those of high-Ti basalt, as well as small-volume lavas and tuffs of rhyolite and trachyte compositions at the top of the volcanic succession.
- (3) Outer zone: the distal volcanics mainly comprise fine-grained mafic tuffs and lesser felsic volcanic ash, which lie at the base of the coal-bearing strata. The distal volcanic ashes also form thin tonstein layers within the coal beds (Dai et al., 2011). Three types of tonsteins (felsic, mafic, and alkali) have been identified, based on their lateral correlation over a large coalfield area and the overall similarity of corresponding mineralogical and

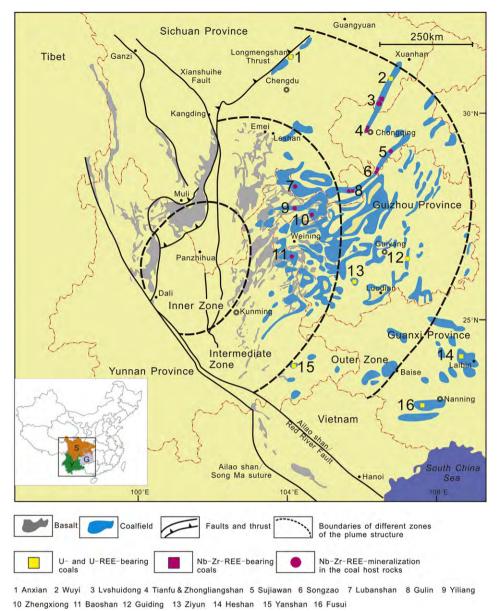


Fig. 6. Distribution of the Late Permian coal basins and metalliferous ore deposits in South China. 1 – Emeishan basalt; 2 – coal basins; 3 – normal fault and thrust; 4 – Emeishan plume structure boundary; 5–7 – rare-metal mineralization in coal basins; 5 – U and U-REE in coal; 6 – Zr–Nb–REE in coal; 7 – Nb–REE in tuff and tuffaceous sedimentary rocks. Data compiled from Xu et al. (2001), Y. Sun et al. (2012), and China Coal Geology Bureau (1996).

chemical compositions (Zhou et al., 1982; Burger et al., 1990, 2000; Zhou et al., 2000; Dai et al., 2011, 2014a). However, only the alkali tonsteins contain high Nb (Ta), Zr (Hf), REY, and Ga that have potential economic significance.

The Emeishan lavas overlie the late Middle-Permian Maokou Formation and are overlain by the Late-Permian Xuanwei/Longtan Formation, suggesting that the age of the Emeishan lavas is constrained to the boundary between the Middle and Late Permian (Xu et al., 2013). However, this boundary is only poorly constrained because the extrusive rocks are thermally overprinted and represent an open system unsuitable for ⁴⁰Ar/³⁹Ar geochronology (Boven et al., 2002; Lo et al., 2002; Ali et al., 2004; He et al., 2007; Zhong et al., 2011; Xu et al., 2013). Based on ID-TIMS (isotope dilution-thermal ionization mass spectrometry) data on mafic, ultramafic and granite intrusions, the Emeishan lavas were constrained to 258–259 Ma (Xu et al., 2013). Zhong et al. (2014) precisely dated the age of the felsic ignimbrites in the uppermost part of the Emeishan lava succession (259.1 \pm 0.5 Ma) using high-resolution chemical abrasion-TIMS zircon U–Pb techniques, and interpreted this as the terminal age of the Emeishan flood basalts.

Based on petrographic, elemental, and Sr–Nd isotopic compositions, the Emeishan basalts can be classified into two major types: 1) a low-Ti type that exhibits low Ti/Y (<500, Fe₂O₃* (<12%), Nb/La (0.6–1.4) and ε_{Nd} (t) (-4.8 to +1.4), but relatively high SiO₂ (48–53%) and Mg (0.52–0.64); and 2) a high-Ti type with high Ti/Y (>500) (Xu et al., 2001). The thick low-Ti lavas are distributed in the western part of the province and may record the main episode of flood basalt emplacement; in contrast, the less abundant overlying high-Ti basalts may imply a waning activity of the plume (Xu et al., 2001). Although the classification of the low- and high-Ti types of the Emeishan basalts remains controversial (Hou et al., 2011; Shellnutt and Jahn, 2011), a recent study by Dai et al. (2014d) showed that roof and floor strata of the coal beds in Xuanwei, Yunnan, were derived from high-Ti alkali basaltic volcanic ashes, further confirming the existence and origin of a high-Ti Emeishan basalt.

2.2.2. Control of ELIP on coal-hosted ore deposits

The control of the ELIP on the late Permian coal-hosted ore deposits, as well as late Permian coal-bearing strata in South China, is reflected by four aspects: (1) the topography (high in west and low in east) that resulted from the ELIP provided favorable sites for peat accumulation (China Coal Geology Bureau, 1996), leading to the majority of the coal-bearing basins distributed in the middle and outer zones of the ELIP (Fig. 6); (2) the Kandian Upland was the dominant sedimentsource region for the late Permian coal-bearing strata (Zhou et al., 1982; China Coal Geology Bureau, 1996; Zhou et al., 2000; Zhuang et al., 2012; Dai et al., 2014d); (3) in some cases, the volcanic ashes resulting from the waning activity of the plume either served as a base for peat accumulation (e.g., as the floor of a coal seam) or terminated peat accumulation (e.g., formed the roof of a coal seam) (Dai et al., 2014d); and (4) the accumulation of organic matter in both the Late Permian coal-bearing basins of South China and in the Cenozoic coalbearing basins of Primorye was accompanied by frequent active volcanism (Zhou et al., 1982; China Coal Geology Bureau, 1996; Zhou et al., 2000; Dai et al., 2011) and related hydrothermal processes (Zhou and Ren, 1992; Ding et al., 2001; Zhang et al., 2002, 2004; Yang, 2006; Dai et al., 2008, 2013a,b). Examples indicating such activity include: the frequent occurrence of tonsteins derived from volcanic ashes in many Late Permian coals of southwestern China (Burger et al., 1990, 2000; Zhou et al., 2000; Dai et al., 2011); the input of syngeneic submarine exhalation to the peat mire (Dai et al., 2008, 2015b); and the high percentages of quartz and chamosite in the Late Permian coals of eastern Yunnan, which are considered to be derived from plume mantlerelated hydrothermal fluids during peat accumulation or at an early stage of diagenesis (Dai and Chou, 2007; Wang et al., 2012). The quartz of syngenetic hydrothermal origin in eastern Yunnan is usually fine to ultra-fine grained, ranging in size from nanometers to less than 20 µm, and may be related to high lung cancer rates in some of the townships in Xuanwei County of eastern Yunnan (Tian et al., 2008; Large et al., 2009).

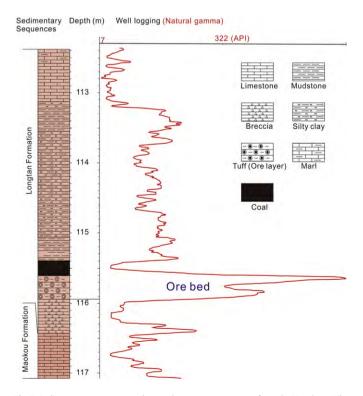


Fig. 7. Sedimentary sequences and natural gamma ray response from the Nanchuan Nb– Zr–REE ore deposit (Drillhole no.: Sujiawan CK6).

2.2.3. Differences of Emeishan and Ussuri structures on coal-hosted ore deposits

One of the major differences between the Emeishan and Ussuri structures is that the former unconformably overlies a carbonate platform sequence, the late Middle Permian Maokou Formation (Fig. 7), which is also the base of the late Permian coal-bearing strata and, in some cases, the tuffaceous Nb–Zr–REE ore deposits in the intermediate and outer zones of the ELIP (Figs. 6 and 7). The Maokou Formation is widely distributed in southern China and is significantly rich in fossils (Wang et al., 1994; China Coal Geology Bureau, 1996; Feng et al., 1997; ECS, 2000). It is dominated by medium-bedded to massive bioclastic limestones and biograin micritic limestones (He et al., 2010). However, where Nb–Zr–REE ore deposits occur, limestone is absent and the ore

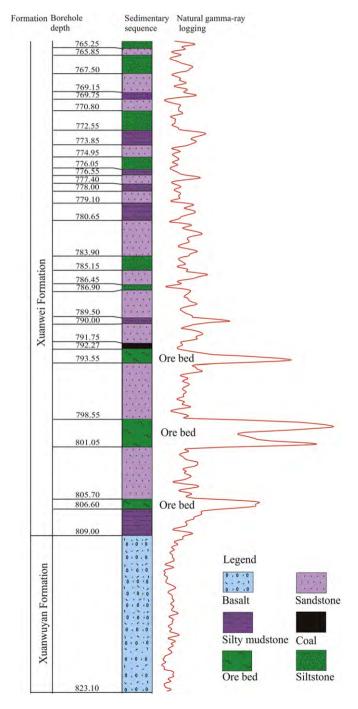


Fig. 8. Sedimentary sequences and natural gamma ray response from the Xuanwei Nb–Zr– REE ore deposit (Drillhole no. 2009-BX-4).

deposit directly overlies the Emeishan basalts and is interbedded with epiclastic sedimentary rocks (Fig. 8). The absence of the Maokou Formation limestone is probably due to rapid crustal uplift or doming related to the mantle plume activity, and subsequent significant weathering and erosion of the limestone (He et al., 2010).

Additionally, periodic marine transgressions occurred during development of the coal-bearing sequence in the outer and intermediate zones of the Emeishan structure. As a result, the coal-bearing strata of South China, in some cases, overlie and are overlain by carbonate rocks (Chen, 1987; Jin and Li, 1987; Huang et al., 1994; Lei et al., 1994; Hou et al., 1995; Shao et al., 2003; Chou, 2004; Zeng et al., 2005; Dai et al., 2008; Chou, 2012; Dai et al., 2013a,b, 2015b), e.g., the Late Permian coals of Yanshan in Yunnan Province, Heshan in Guangxi Province, Chenxi in Hunan Province, and Guiding in Guizhou Province (Figs. 6 and 9). Coals that have been subjected to sea-water invasion during peat accumulation generally have low concentrations of rare metals, close to the averages for world coals (e.g., Song et al., 2007; Wang et al., 2007; R. Sun et al., 2010). However, coals preserved within the marine carbonate successions in South China (Fig. 9) have high concentrations of rare metals, such as U, Se, and Mo (Table 2) and, in some cases (e.g., Yanshan and Chenxi), REY, which are predominantly attributed to hydrothermal activity and the euxinic environment favorable for the preservation of those elements (Dai et al., 2013a,b, 2015b). The U(Mo,Se)-REE ores contain up to 0.05-0.1% U and 0.1-0.3% REE (Dai et al., 2013a,b, 2015b). Studies by Shao et al. (2003) and Zeng et al. (2005) also showed that the Heshan coals preserved within the marine carbonate successions of Guangxi Province are significantly enriched in U, Se, and Mo, close to the values listed in Table 2.

In addition to U(Mo,Se)–REE ore deposits, the other geochemical type of rare-metal mineralization in coal basins of South China is Zr(Hf)–Nb(Ta)–REE (Table 3), which is mainly associated with mafic and felsic alkaline tuffs (Dai et al., 2010) and, in some cases, alkali rhyolite tuffs in coal-bearing basins (Dai et al., 2014a). The Zr(Hf)–Nb(Ta)–REE ores are

Table 2

Concentration of rare metals in coal ash from coal-hosted U(Mo,Se)–REE ore deposits of southwestern China (μ g/g unless indicated as %).

Ore deposits	Ν	V	Se	Mo	Re	REO	U	Sum-RM (%)
Guiding ^a	16	3998	152	1652	1.49	337	950	0.68
Yanshan ^b	7	2061	91.6	742	1.1	941	556	0.44
Fusui ^c	10	147	22.9	27.3	nd	1170	21.0	0.12
Heshan ^d	14	381	35.5	125	nd	767	126	0.14
Chenxi ^e	11	2120	nd	166	nd	1349	539	0.42
World coal ash ^f	nd	170	10.0	14	nd	534	15	0.074

N, sample number. REO, sum of oxides of rare earth elements and yttrium. Sum-RM, sum of V, Se, Mo, Re, REO, and U.^a, from Dai et al. (2015b).^b, from Dai et al. (2008).^c, from Dai et al. (2013a).^d, from Dai et al. (2013b).^e, from Li and Tang (2013) and Li et al. (2013).^f, from Ketris and Yudovich (2009). nd, no data.

represented by horizons of argillized (Figs. 7 and 8) and, in a few cases, hematitized volcanic-ash, 5-10-m thick, and by metalliferous coals (Fig. 10). Compared to the normal sedimentary rocks in the sequences derived from the sediment source region composed mainly of mafic Emeishan basalts, natural gamma-ray data of the Nb-Zr-REE ore beds and the alkali tonsteins show a significant positive anomaly in well logging (Figs. 7 and 8), which may be considered as a geophysical indicator in Nb-Zr-REE ore prospecting (Dai et al., 2010, 2012). Rare-metal concentrations are up to 1-3% (Zr,Hf)₂O₅, 0.05-0.1% (Nb,Ta)₂O₅, and 0.1-0.5% REO in the Zr(Hf)-Nb(Ta)-REE ores (if coal-hosted ores, raremetal concentrations are in the coal ash) (Dai et al., 2010; Zhang, 2013; Dai et al., 2014a). The geochemical compositions of argillized tuffs show that the rare-metal-enriched tuffs may be at the periphery of the Emeishan Large Igneous Province, and probably resulted from a waning activity of the plume (Dai et al., 2011). The highly enriched Zr(Hf), Nb(Ta), and REY ores may have originated from different mantle sources under various low-degree partial melting conditions, and have undergone fluid fractionation and contamination from lithospheric mantle and crustal material. Their source magmas had an alkali basalt composition and were similar to ocean island basalts (Dai et al., 2011).

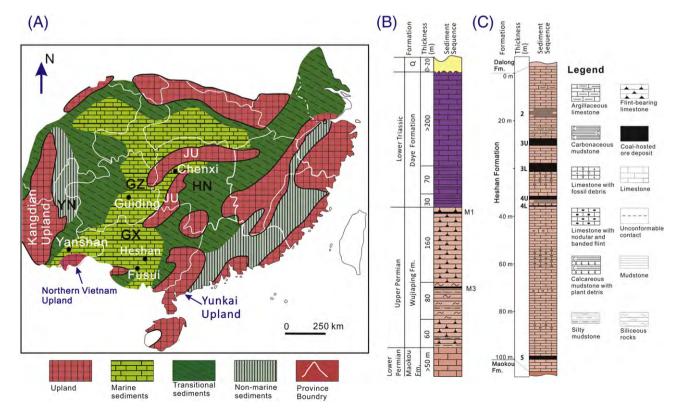


Fig. 9. Paleoenvironment and location of the Guiding, Yanshan, Heshan, Fusui, and Chenxi U(Mo,Se)–REE ore deposit in South China (A), sedimentary sequences of Guiding (B) and Heshan (C) deposits. Data are compiled from Dai et al. (2008, 2015b) and China Coal Geology Bureau (1996).

Table 3

Concentration of rare metals in coal-hosted Zr(Hf)-Nb(Ta)-REE ore deposits of southwestern China (µg/g unless indicated as %).

Ore deposits	Sample	Thickness (m)	(Zr,Hf) ₂ O ₅	(Nb,Ta) ₂ O ₅	REO	Sum-RM (%)
Huayingshan	K1-2p ¹⁾	0.05	2589	433	1972	0.499
	K1-1p2 ¹)	0.07	2569	448	2724	0.574
	K1-1p1 ¹⁾	0.03	2378	618	2289	0.529
	H-Coal ²⁾	2.03	3617	406	1710	0.573
Baoshan Xuanwei	2009-BX-4 ³⁾	2.5	6011	555	1358	0.792
	2009-YN-5 ⁴⁾	16.0	3805	302	1216	0.532
	2009-LY-10 ⁵⁾	3.0	8464	627	1271	1.036
Xinde Xuanwei ^a	301-10 ⁶⁾		3520	558	1248	0.553
	301-13 ⁶⁾		4852	745	1055	0.665
	301-14 ⁶⁾		4703	742	1227	0.667
	301-15 ⁶⁾		4122	673	1263	0.606
	301-16 ⁶⁾		4139	639	1791	0.657

Sum-RM, sum of (Zr,Hf)₂O₅, (Nb,Ta)₂O₅, and REO. 1), Alkali tonsteins; 2), Zr(Hf)–Nb(Ta)–REE-rich coal, based on five samples; 3)–6), tuffaceous Zr(Hf)–Nb(Ta)–REE ore deposit. 3), based on 14 samples. 4), based on 43 samples, 5), based on five samples. 6), Zr(Hf)–Nb(Ta)–REE-rich layer.

^a The total thickness of the ore layers is 2.5 m.

At the same time, the Sr–Nd isotopic data (Fig. 11; Xu et al., 2010; and references therein) clearly demonstrate that the Emeishan plume experienced significant crustal assimilation during its ascent. Additionally, the felsic rocks show a trend of isotopic variations similar to that for South Primorye, characterized by a wide range of ⁸⁷Sr/⁸⁶Sr and uniform ¹⁴³Nd/¹⁴⁴Nd ratios. This suggests a similar mineralization process, strongly influenced by the crustal material. Thus, a crustal source for the Zr(Hf), Nb(Ta), and REY ores of the South China coal basins is highly probable or, at least, cannot be excluded. The hot fluids could have reworked continental crust rich in rare metals and redistributed them through volcanic eruptions and subsequent sedimentation. Such mantle–crust interactions resulting in rare metal mineralization might be caused by either a deep mantle plume or asthenospheric upwellings, as suggested for East Asia in the Cenozoic (Flower et al., 1998) and for the Emeishan plume in the Permian–Triassic (Shellnutt, 2014).

Different mineralization types are distributed in different zones. The U(Mo,Se)–REE ores are located exclusively in the outer Emeishan zone

or outside the Emeishan zone, whereas the Zr(Hf)–Nb(Ta)–REE ores are situated in the middle and outer northwestern Emeishan zones.

3. Rare-metal mineralization in metalliferous coal deposits

3.1. South Primorye

Mineralization with typical REY–Zr–Nb associations has been known in felsic and alkali tonsteins in many coal deposits (Crowley et al., 1989; Hower et al., 1999; Arbuzov et al., 2000; Zhou et al., 2000; Seredin and Finkelman, 2008; Dai et al., 2011; Spears, 2012). However, data on highly-elevated concentrations of REY in the host rocks of coal seams (e.g., roof and floor strata), rather than in the coal seams themselves, are limited to a few coal deposits in the Russian Far East, China, and Uzbekistan (Seredin and Finkelman, 2008). This is not only because little attention has been paid to the study of coal-hosted units and the basement rocks of coal deposits, but also because the rare-metal-

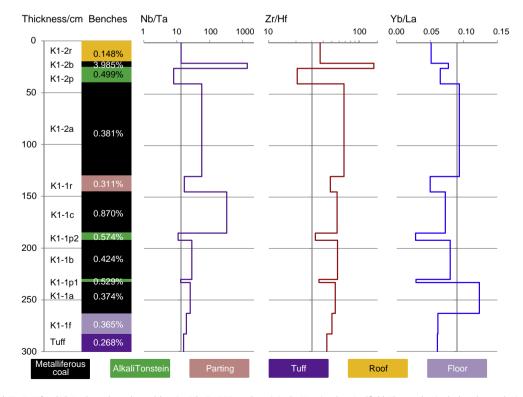


Fig. 10. Variations of Nb/Ta, Zr/Hf and Yb/La throughout the coal-bearing Nb–Zr–REE ore deposit in the Huayingshan Coalfield. The number in the bench samples is the rare metal contents. The vertical lines represent the values for world hard coals.

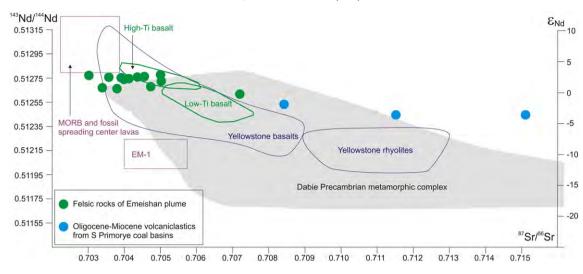


Fig. 11. ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd ratios for Permian–Triassic volcanic rocks of the Emeishan plume (after Xu et al., 2010; see Fig. 4 and text for other references). Data for the Cenozoic volcanoclastics from Primorye are shown to demonstrate that the Sr–Nd isotopic variations of felsic rocks from both regions under consideration show a continuous trend characterized by a wide range of ⁸⁷Sr/⁸⁶Sr but uniform ¹⁴³Nd/¹⁴⁴Nd, suggesting a similar mineralization process influenced by mantle melts interacting with the crust.

bearing intraseam tonsteins are thin (generally 3–10 cm) and, thus, can hardly be considered as raw materials for rare metal recovery. However, the frequent occurrence of such rare-metal-bearing tonsteins provides a basis for predicting the possibility of thick horizons of REY-bearing tuffs outside of coal seams (Seredin and Finkelman, 2008; Dai et al., 2011). This forecast has been successfully realized in Yunnan Province, China



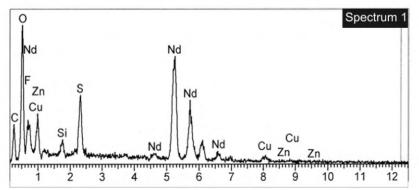


Fig. 12. SEM back-scattered electron image of Nd–F carbonate in tuffaceous clay in the Nezhino deposit and its ED spectrum. The Cu, Zn, and S peaks are most likely due to sulfides which are dispersed on the Nd–F carbonate surface.

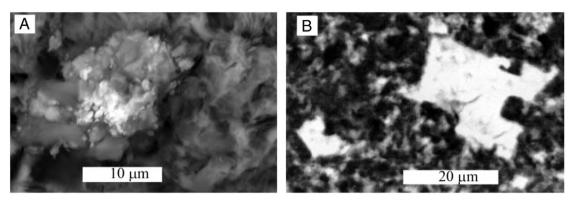


Fig. 13. SEM back-scattered electron images of cerium titanate inclusions (white) in the hollandite ore hosted by Cenozoic sediments (A) and in the Al-lithiophorite veinlet of the Pavlovka coal deposit basement (B).

(Dai et al., 2010) and in the Cenozoic tuffaceous coal-bearing strata in the southern part of the Russian Far East (Seredin et al., 2011). Within these tuffs, the REY content varies from 0.07 to 0.17% with a thickness of up to 10 m. It is believed that similar tuffaceous rare-metal mineralization may be found in many coal basins of various ages worldwide.

A number of different types and modes of occurrence of REY mineralization have been found in the coals and host rocks of the Pavlovka coal deposits in South Primorye (Seredin and Finkelman, 2008). For example, the Nezhino deposit is characterized by tuffaceous–hydrothermal Zr–Nb–REE mineralization in tuffaceous-clayhosting coal, while the Vostochny open-cast mine in the Pavlovka deposit is characterized by a stratiform manganese (hollandite) ore located among the rocks overlying the coal seams (Seredin et al., 2011; Seredin and Chekryzhov, 2011, 2012b; Seredin and Dai, 2012; Seredin et al., 2012).

The tuffaceous clay in the Nezhino deposit contains an unusual, as yet unnamed fluoro-carbonate mineral, which contains Nd but no other rare earth elements (Fig. 12). Similar REE-bearing minerals, e.g., Ge- and Dy-bearing minerals as well as REY compounds with other metals (Sm–Fe–Co, Sn–Ca–Co, Dy–Fe, Y–Ni), have been observed in the coal and fossilized xylem of the Pavlovka deposit, as well as in hydrothermal REE ores in the basement of that deposit (Seredin, 1996; Seredin and Mokhov, 2007). Although the Nd-F carbonate mineral has not been observed in other coal deposits, this occurrence may be considered as evidence for the significant fractionation of the trivalent lanthanides by geological processes, especially in the hydrothermal alteration (argillization) of tuffaceous horizons (Seredin, 1996; Seredin and Finkelman, 2008). The presence of fluorine in this mineralization may also be considered as evidence of Cenozoic redistribution of the Paleozoic rare-metal and fluorine crustal ores that are abundant in the Ussuri district (Sato et al., 2003; Kupriyanova et al., 2005).

A thick horizon of hollandite ore was identified in the Miocene tuffite roof of the coal-bearing strata at the Vostochny open-cast in the Pavlovka deposit. This was initially considered to be a product of

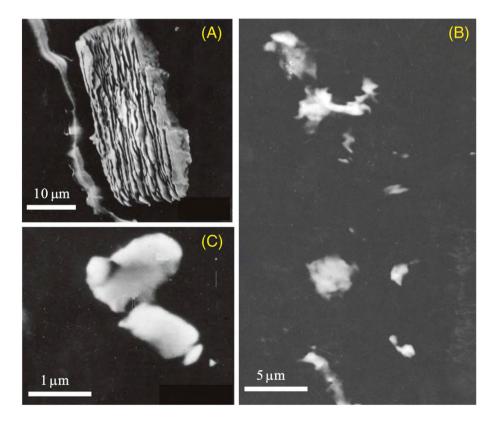


Fig. 14. SEM back-scattered electron images of native Au (A), Ag (B), and Pt (C) from Ge-rich coals of the Pavlovsk deposit, Luzanovka ore field.

supergene processes (Seredin, 1998). However, the identification and modes of occurrence of cerium titanate phases (Fig. 13A) in the stratiform hollandite ore suggest that the ore was of hydrothermal origin (Seredin, 1998). Cerium titanate was also identified in quartzlithiophorite stockworks occurring at the base of the Pavlovka deposit (Fig. 13B). The REE-rich coals in the Pavlovka deposit display similar mineral assemblages to the Ge-bearing coals (include native metals, metal alloys, sulfides, halogenides, and carbonates of various metals), indicating hydrothermal input (Seredin, 1998). This hydrothermal mineralization was formed from gas-saturated H₂O–CO₂ fluids with temperatures of 138–153 °C (Seredin, 1998, 2005).

Although middle Asia contains the majority of U-bearing coals, some U–Mo mineralization has also been observed in coal basins of South Primorye (Seredin and Finkelman, 2008), where it is generally associated with Ge enrichment in the coal.

The typical hydrothermal Ge–U–Mo mineralization style is represented by the Pavlovka coal-hosted Ge ore deposit (Primorye, Russian Far East). The average Ge content in the Ge-rich coal seams is 1025 μ g/g (Dai et al., 2014b). High concentrations of Ge, U (up to 594 μ g/g) and Mo (up to 101 μ g/g) in the Ge-rich coals are related to intense leaching of the underlying granites by N₂–CO₂-mixed hydrothermal solutions (Seredin et al., 2006), which is evidenced by the carbonate mode of U transfer in the hydrothermal solutions (Naumov, 1998) and by the Eu enrichment of the coals and coal-bearing rocks. Eu is generally concentrated in the K-feldspar of the granite; therefore, intense leaching of this abundant mineral by hydrothermal solutions passing through the alkaline basement granites would have led to Eu enrichment in the

solutions (Seredin et al., 2006). Such Eu enrichment has also been observed in hydrothermal water vapors in Yellowstone National Park (Lewis et al., 1995). The Ge, U, and Mo in the coals are organicallybound (Seredin et al., 2006; Seredin and Finkelman, 2008). Relative to the upper continental crust (Taylor and McLennan, 1985), the Pavlovka Ge-rich coals are characterized by heavy-REE enrichment (Dai et al., 2014b), also indicating a hydrothermal input (Seredin and Dai, 2012). In particular, a hydrothermal overprint in the Ge deposit is also evidenced by abnormally high concentrations of As (up to 117 µg/g) and Sb (up to 63.5 µg/g) (Seredin et al., 2006). Along with As and Sb, elements Au, Ag, and Pt are enriched in the Pavlovka Ge-rich coals, occurring in the form of both native metals (Au, Ag, and Pt; Fig. 14), as well as organically-associated Au (Bratskaya et al., 2009).

3.2. South China

The rare-metal ore beds in the coal-bearing basins of South China are mainly argillized tuffs (including those of intra-seam tonsteins) and metalliferous coals (Dai et al., 2010, 2014a). The rare metals mainly occur within secondary minerals (e.g., rhabdophane, florencite, brannerite, and coffinite respectively forming in different ore deposits; Dai et al., 2014a,c) or are associated with clay minerals and organic matter (Dai et al., 2010; Seredin and Dai, 2012; Dai et al., 2013a).

The common REE-, Zr-, and Nb-bearing minerals (e.g., columbite, pyrochlore, samarskite, zircon, and hafnon) are rarely observed in these ore beds, either by XRD or under the optical microscope (Dai et al., 2010). Rare-metal-bearing minerals identified in the argillized

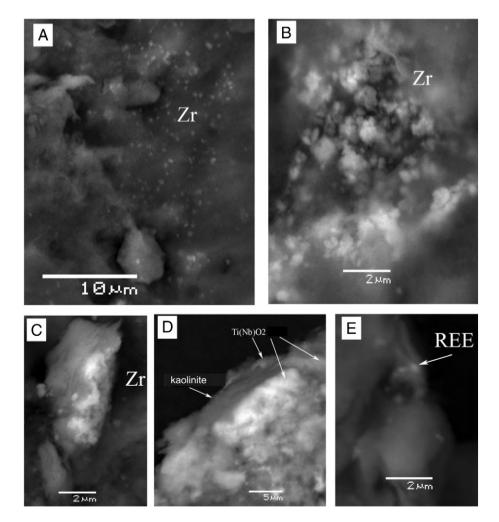


Fig. 15. SEM images of secondary rare-metal minerals in argillized tuffs from Xuanwei, eastern Yunnan of South China: A-C, zircon; D, Nb-rutile; E, rhabdophane.

tuffs include rhabdophane, silico-rhabdophane, Nb-bearing anatase (or rutile), florencite, REE-bearing carbonates, zircon, parisite, and xenotime. They occur as finely-dispersed grains in clay matrices (Fig. 15A), microcrystalline aggregates in the pores of kaolinite (Fig. 15B), or films on kaolinite surfaces (Fig. 15C–E). In some cases, they occur as coarse-grained particles in the ore deposits (Figs. 16, 17, and 18A, B), e.g., distributed in the clay matrix (Fig. 16), filling cavities of hydrothermally-altered primary minerals of magmatic origin (Figs. 17 and 18B), or along the cleavage planes of clay minerals (Fig. 18A). The modes of occurrence of these raremetal-bearing minerals suggest that they are of authigenic origin. However, rare metals rarely occur in the unaltered primary minerals of magmatic origin (Fig. 18C) and do not occur in the secondary chlorite of

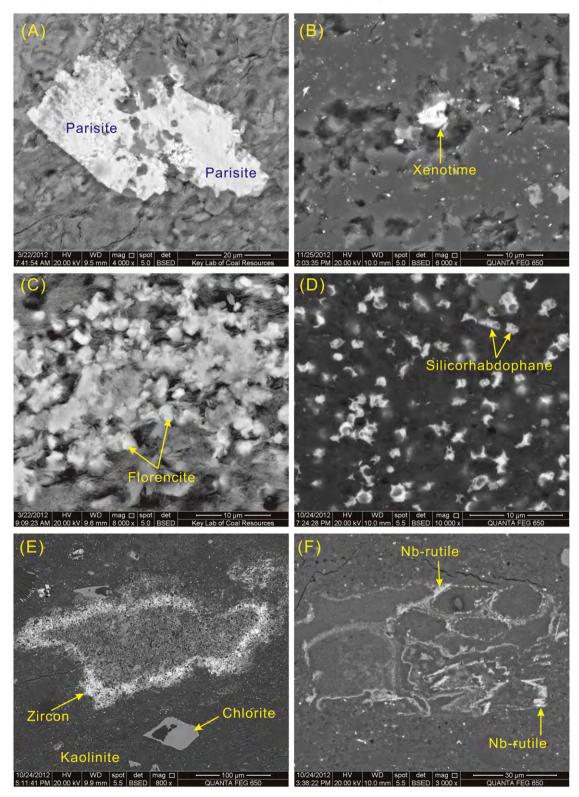


Fig. 16. SEM back-scattered images of rare-metal-bearing minerals distributed in the clay minerals of Zr(Hf)–Nb(Ta)–REE ore deposits in eastern Yunnan, South China. (A), Parisite in borehole 1908#; (B), xenotime in borehole 1908#; (C), florencite in borehole 1908#; (D), silico-rhabdophane in borehole 301#; (E), zircon in borehole 301#; (F), Nb-rutile in borehole 301#.

hydrothermal origin (Fig. 18D), the latter being commonly observed in argillized tuffs in eastern Yunnan.

These authigenic minerals are derived from re-deposition of rare metals leached from the argillized tuffs by hydrothermal fluids (Seredin et al., 2011; Seredin and Chekryzhov, 2012a; Seredin et al., 2012). For example, the mode of rhabdophane occurrence (Fig. 18A) probably indicates re-deposition from leached REY derived from the pre-existing volcanic ash. Figs. 19 and 18B respectively show that REE-bearing apatite and Nb-bearing rutile were partly corroded by hydro-thermal solutions, and were then coated by authigenic kaolinite and filled by authigenic chlorite, respectively. This mineralization process can be classified as a mixed tuffaceous–hydrothermal type. Primary minerals of magmatic origin have rarely been observed (Fig. 18C), because they had been leached-out or altered by the hydrothermal fluids (Figs. 17 and 18B).

Zircon is common in the felsic tonsteins but very rare in the alkali tonsteins (Zhou et al., 2000; Dai et al., 2011) and argillized tuffs (Zhang, 2013), although the Zr concentration in the alkali tonsteins and argillized tuffs (Table 3) is much higher than that in the felsic tonsteins (e.g., 158 and 329 µg/g in two felsic tonstein layers at Xinde, eastern Yunnan, Dai et al., 2014d; and 102-840 µg/g with an average of 433 µg/g in 18 felsic tonstein layers in southwestern China, Zhou et al., 2000). It has been deduced that the relationship between Zr and zircon is poor in alkali tonsteins (Zhou et al., 2000) and in argillized tuffs (Zhang, 2013). Zirconium occurs mainly as zircon in felsic tonsteins (Zhou et al., 1994; Dai et al., 2011, 2014d) but is probably associated with clay minerals in alkali tonsteins, for example, as ion-absorbed froms (Zhou et al., 2000), and in argillized tuffs (Zhang, 2013). However, because the alkali tonsteins and tuffs were generally subjected to leaching by hydrothermal solutions, a large proportion of clayassociated (e.g., ion-absorbed form) Zr could have been leached out by hydrothermal solutions and then re-deposited as secondary minerals (e.g., authigenic zircon in Fig. 20C).

Zircon, as well as rare earth elements and Nb, leached from volcanic ashes and then re-deposited has also been observed by Hower et al. (1999), Crowley et al. (1989), and Dai et al. (2014a). Fig. 10 shows that the REE–Nb–Zr-rich coals from the Huayingshan Coalfield in southwestern China have higher Nb/Ta, Zr/Hf, and U/Th ratios than the alkali tonsteins and host rocks (roof and floor strata). This is attributed to re-deposition of the first elements (Nb, Zr, and Yb) of each element

pair, because of their relatively more active leaching from the tonsteins and host rocks, and then deposition in the underlying organic matter.

The REE-bearing minerals commonly contain only light REE, although, in a few cases, xenotime (Fig. 16B), fluor-carbonates, and phosphates (Fig. 21) that are enriched in yttrium (up to 0.2%) have been found in the tuffs. However, ore horizons characterized by HREE enrichment have been generally observed in the ore-bearing tuffs (Fig. 22).

It has been inferred that the heavy REE in the tuffaceous clays are mainly associated with the clay minerals (e.g., occurring as adsorbed ions, Seredin and Dai, 2012). From the metallurgical point of view, the tuffaceous ores in coal basins with carbonate and ion-adsorption modes of occurrence are favorable for commercial REE utilization. It is probable that REY sources of the currently exploited ion-adsorption ores of weathering crusts (currently the major sources for yttrium in the world, and also for the medium and heavy REEs, the reserves of which in China will soon run out) will be replaced by those of the tuffaceous ores presented in this study.

A number of rare-metal-bearing minerals (e.g., rhabdophane, silicorhabdophane, Nb-bearing anatase (or rutile), florencite, REEbearing carbonates, goyazite, zircon, brannerite, coffinite, apatite, monazite) have been found in metalliferous coals, and these evidently have a hydrothermal origin (Dai et al., 2013a,b; Seredin and Dai, 2012; Fig. 20). There are, however, a few exceptions, represented by monazite, zircon, and rutile that, in some cases, are detrital materials of terrigenous origin (Zhuang et al., 2012). The authigenic REE-bearing minerals are either distributed along bedding planes (e.g., REE-bearing phosphates and silico-phosphates, which in some cases are also rich in U and Zr; Fig. 20A), or in collodetrinite and clay minerals. In some cases, REEbearing minerals occur as cell-fillings or as veinlets cutting across the bedding planes (e.g., REE-carbonate mineral; Fig. 20D). Authigenic brannerite (Fig. 20B) and U-bearing REE phosphate, as well as bedcutting veinlets of kaolinite, quartz, strontianite, and calcite (Dai et al., 2013a,b), have been observed in the coal-hosted U ore deposits of south China, although U has been reported to be mainly associated with organic matter in U-rich coals (Dai et al., 2008; Seredin and Finkelman, 2008; Arbuzov et al., 2012; Dai et al., 2015a,b). In addition, authigenic zircon occurs as veinlet- or cell-fillings coexisting with both kaolinite and calcite (Fig. 20C).

As with the argillized tuffs, all the REE minerals identified in the coals are light REE-bearing minerals, although the host coals themselves

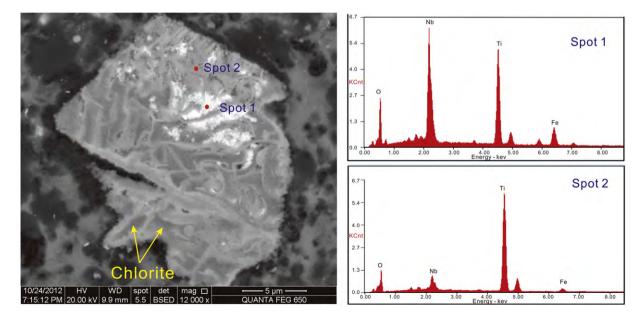


Fig. 17. SEM back-scattered images and EDS data for Nb-rutile in borehole 301# from the Zr(Hf)–Nb(Ta)–REE ore deposits of South China. Chlorite fills in the cavities of the hydrothermally-altered rutile.

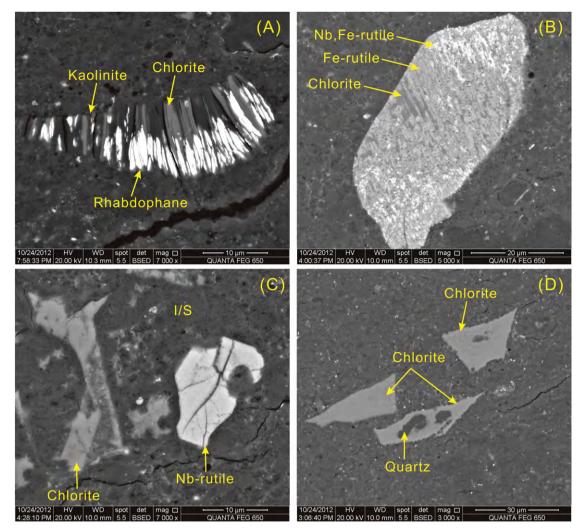


Fig. 18. SEM back-scattered images of minerals in borehole 301# from the Zr(Hf)–Nb(Ta)–REE ore deposits of South China. (A), Rhabdophane distributed along cleavage planes of clay minerals and chlorite; (B), Nb,Fe-rutile and chlorite in hydrothermally-altered rutile; (C), rutile of magmatic origin and chlorite of hydrothermal origin; (D), chlorite derived from vitroclastics by hydrothermal fluids, with authigenic quartz filling in the chlorite cavities.

are rich in heavy REE (Fig. 22). However, heavy REE-bearing minerals are absent and, thus, the heavy REE are deduced to be associated with the organic matter of the coal. The different carriers of LREE and HREE

suggest a sharp fractionation between light and heavy REE groups during hydrothermal processes in these organic-rich environments. Experiments on sorption of REEs on xylain and humic acid by Eskenazy

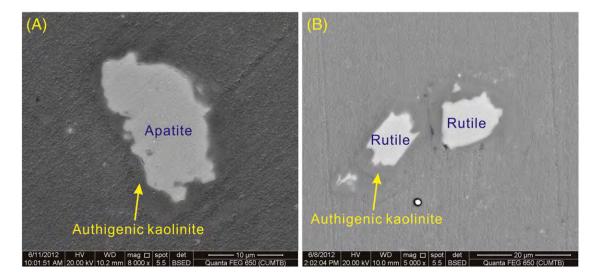


Fig. 19. Altered REY-bearing apatite and Nb-bearing rutile in the Fusui ore deposit, South China. SEM back-scattered electron images.

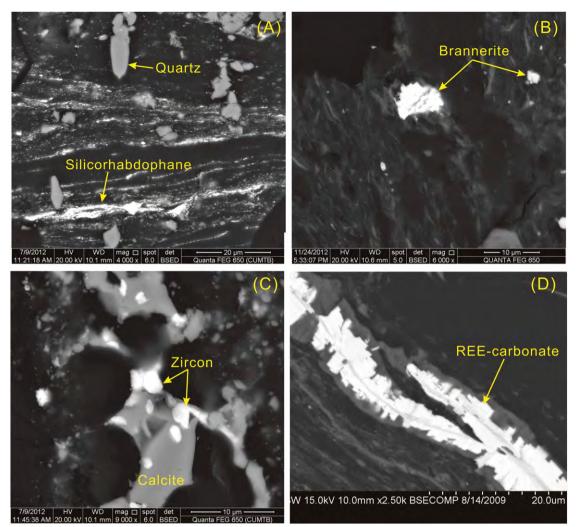


Fig. 20. SEM back-scattered images of authigenic rare-metal-bearing minerals in the Late Permian coal-hosted ore deposits of South China. (A), silico-rhabdophane distributed along bedding planes in coal sample K1-2b of the Huayingshan Coalfield. (B) Brannerite in coal sample GC-3-1 of the Guiding coal-hosted U(Se, Mo) deposit. (C) Authigenic zircon in veinlets in coal sample K1-2b of the Huayingshan Coalfield. (D) REE-carbonate minerals in veinlets in coal sample th-k2b-4 of the Songzao Coalfield, southwestern China. Images C and D are from Dai et al. (2014a) and Zhao et al. (2013), respectively.

(1999) showed that the HREE have a stronger ability to form complexes with organic compounds than the LREE. A greater organic affinity for HREE than for LREE in coal has also been found by Querol et al. (1995), Palmer et al. (1990), and Goodarzi (1987). Seredin and Shpirt

(1999) showed that 50% of the REY in metalliferous coals occur in the form of humic complexes and could be leached by a 1% NaOH solution, further indicating that the REY are associated with organic matter. Organically-associated REY are also found in metalliferous coals of the

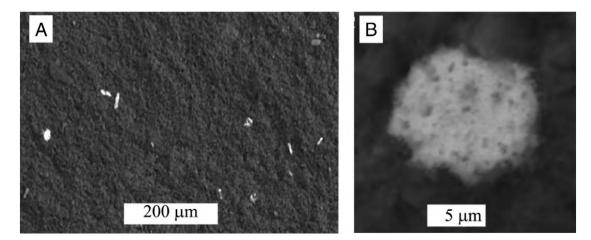


Fig. 21. SEM back-scattered images of dispersed REE-F-bearing carbonates (white) in argillized tuff from Xuanwei, eastern Yunnan, southwest China. A, general view; B, in detail.

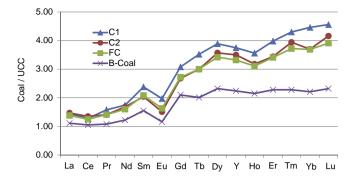


Fig. 22. REY distribution patterns of the REY-rich No. 1 Coal in the Fusui Coalfield, South China. C1 and C2 are both channel samples of No. 1 Coal; B-Coal, weighted averages by thickness of bench sample interval; FC, overall averages of channel and bench samples. REE data from Dai et al. (2013a). REY are normalized to Upper Continental Crust (UCC) (Taylor and McLennan, 1985).

Vanchin Graben, Primorye, Russia (to the east of the Ussuri structure), where REY-bearing minerals are absent in the REY-rich coals, although REE phosphates and fluor-carbonates have been identified in the host rocks (Seredin and Shpirt, 1999). Organically-associated REY in coal, especially as humic complexes, have also been found by Arbuzov and Ershov (2007), Arbuzov et al. (2012), Shpirt and Seredin (1999), and Finkelman et al. (1990).

4. Conclusions

- (1) The major rare-metal coal deposits in East Asia, mainly containing Zr(Hf)-Nb(Ta)-REE and U(Mo,Se)-REE ores, in particular those of South Primorye and South China, initially resulted from the evolution of plumes ascending from the deep mantle and/or asthenospheric flows, both of which included reworking of the continental crust.
- (2) This mantle-crust interaction not only led to coal-basin formation but also played a significant role in the production of extensive volcanism and ore-generation via associated hydrothermal activity.
- (3) Three mineralization styles can be identified for the metalliferous coal deposits of East Asia: tuffaceous, hydrothermal-fluid, and mixed tuffaceous-hydrothermal fluid types. The mineralization process occurred not only in the coals but also in the associated host rocks, and not only during peat accumulation but also after the coalification stage.
- (4) The rare metals in the metalliferous coal deposits are generally associated with clay and organic matter, or occur as secondary minerals derived from decomposition of the primary rare-metal bearing magmatic minerals.
- (5) It is expected that similar rare-metal mineralization in coal deposits, associated with extensive volcanism and hydrothermal activity and of potential economic significance, may be discovered elsewhere in Russia (e.g., Kuznetsk, South Yakutian, etc.), China and other coal basins of the world, after further geochemical and mineralogical investigations.

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