

Recycling of metal-fertilized lower continental crust: Origin of non-arc Au-rich porphyry deposits at cratonic edges

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ABSTRACT

Recent studies argue that subduction-modified, Cu-fertilized lithosphere controls the formation of porphyry Cu deposits in orogenic belts. However, it is unclear if and how this fertilization process operates at cratonic edges, where numerous large non-arc Au-rich deposits form. Here we report data from lower crustal amphibolite and garnet amphibolite xenoliths hosted by Cenozoic stocks that are genetically related to the Beiya Au-rich porphyry deposits along the western margin of the Yangtze craton, China. These xenoliths are thought to represent cumulates or residuals of Neoproterozoic arc magmas ponding at the base of arc at the edge of the craton that subsequently underwent high-pressure metamorphism ca. 738 Ma. The amphibolite xenoliths are enriched in Cu (383-445 ppm) and Au (7-12 ppb), and a few garnet amphibolite xenoliths contain higher Au (6-16 ppb) with higher Au/Cu ratios $(2 \times 10^{-4} \text{ to } 8 \times 10^{-4})$ than normal continental crust. These data suggest that metal fertilization of the base of an old arc at the edge of the craton occurred in the Neoproterozoic via subduction modification, and has since been preserved. The whole-rock geochemical and zircon Hf isotopic data indicate that melting of the Neoproterozoic Cu-Au-fertilized low-crustal cumulates at 40-30 Ma provided the metal endowment for the Au-rich porphyry system at the cratonic edge. We therefore suggest that the reactivated cratonic edges, triggered by upwelling of asthenosphere, have the potential to host significant Au ore-forming systems, especially non-arc Au-rich porphyry deposits.

INTRODUCTION

Unlike orogenic belts, most cratons have been stable since their formation in the Archean–Proterozoic Eons (Griffin et al., 2013). The initial cratonic crust and subcontinental lithospheric mantle (SCLM) are strongly depleted in magmaphile elements, especially Au and Cu (Rudnick and Gao, 2003), largely due to the liberation of metamorphic fluids from the deep crust (Cameron, 1989) and a high degree of mantle partial melting (Griffin et al., 2013), and have been preserved as a durable, rigid, and buoyant raft. Therefore, the cratons that have not undergone metal fertilization and later activation would be unlikely to host Phanerozoic Cu-Au ore deposits (Groves and Bierlein, 2007). However, recent studies have recognized a suite of Mesozoic–Cenozoic large Au-rich deposits, varying from porphyry Cu-Au (Richards, 2009; Griffin et al., 2013; Hou et al., 2015) to orogenic Au deposits (Goldfarb et al., 2007; X. Sun et al., 2009) that have formed at the margins or in the cratonic interior. They are postulated to be genetically related to non-arc potassic magmas derived from the Proterozoic lithosphere (Lu et al., 2013), metasomatized lower crust (Richards, 2009), SCLM (Griffin et al., 2013), and/or crustal fluids from deep reservoirs (Goldfarb et al., 2007), released during later reactivation. All these non-arc Au-rich deposits are in the category of postcollisional deposits. The occurrence of these Au-rich deposits implies the existence of metal fertilization in cratons during later tectonic episodes (Griffin et al., 2013). However, it is unclear when and how this fertilization process operated, and what factors ultimately control the formation of Au-rich porphyry copper deposits (PCDs) (Richards, 2009).

Here we report the occurrence of Cu-Au–rich lower crustal amphibolite and garnet amphibolite xenoliths, hosted by Cenozoic potassic stocks that are genetically related to the Beiya Au-rich PCD in the western Yangtze craton, China. We suggest that these xenoliths represent direct samples of the Neoproterozoic fertilized cratonic lower continental crust, which played an important role in the genesis of Au-rich PCDs at cratonic edges.

GEOLOGICAL BACKGROUND

The Yangtze craton, southwest China, underwent Neoproterozoic lithospheric accretion and Cenozoic tectonic reactivation with Au-Cu mineralization at its margin (Fig. 1). It is therefore an ideal place to study Au-rich systems at cratonic edges. The remnants of 1000–740 Ma voluminous arc plutons and volcanic rocks in the western part of the craton (Fig. 1B) suggest that the oceanic subduction beneath the craton occurred in the Neoproterozoic (W. Sun et al., 2009). An ~1000-km-long potassic magmatic belt of Eocene–Oligocene intrusive and associated volcanic rocks (40–30 Ma) along the cratonic edge (Fig. 1B) record significant reworking by Indo-Asia collision that started ca. 65 Ma (Lu et al., 2013).

The Beiya Au-rich PCD (304 t Au, 2.4 g/t Au; 0.6 Mt Cu, 0.48% Cu) is the largest among several PCDs associated with the Eocene–Oligocene collision-related intrusive stocks (ca. 37 Ma; He et al., 2015). The Beiya porphyries are thought to have formed by remelting of thickened mafic lower crust, whereas the Liuhe syenite stocks formed by remelting of the metasomatized SCLM during collision (Lu et al., 2013). Geophysical data reveal that asthenosphere upwelling appears along the cratonic edge with 42–45-km-thick crust, and upwelling is thought to trigger melting of the cratonic lithosphere during collision (Lu et al., 2013).

LOWER CRUSTAL XENOLITHS AND THEIR ORIGIN

Abundant xenoliths have been found with Eocene stocks and associated volcanic rocks at six locations exposed along the cratonic edge (Fig. 1B). Amphibolites and garnet amphibolites are the primary types, the former hosted by Liuhe syenites and Beiya monzogranite porphyries (Fig. 2A), and the latter widely occurring in the Liuhe stock (Fig. 2B). Their mineralogical and whole-rock compositions, and zircon, oxide, and sulfide geochemical

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Figure 1. A: Significant orogenic Au deposits (black dots) in the North China craton (NCC, 130–120 Ma; Goldfarb et al., 2007) and Yangtze craton (YC, 38–32 Ma; X. Sun et al., 2009). CAO—Central Asia orogen; TM—Tarim block; CCO—Central China orogen; AHO—Alpine-Himalaya orogen; SGO—Songpan Ganzi orogen; CC—Cathaysia craton. B: Locations of the lower crustal xenoliths (six stars) are shown; xenoliths at Beiya and Liuhe were sampled for study. The tectonic framework and distribution of Cenozoic porphyry Cu-Au and orogenic Au deposits in the western Yangtze craton are shown.

data are listed in Tables DR1–DR11 in the GSA Data Repository¹ and presented in Figure 2 and Figures DR1–DR6 (the Data Repository).

Sulfide phases (<0.3 vol%) in the garnet amphibolites are dominated by pyrrhotite with chalcopyrite rims and pyrites. The pyrrhotite occurs as globules enveloped by garnet grains that lack any fissures and hydrothermal alteration (Fig. 2C). Their globular shape, sharp boundaries, and the coexistence of pyrrhotite with chalcopyrite suggest original entrapment as a magmatic sulfide melt (Nadeau et al., 2010). Magnetite occurs inside and outside of amphibole crystals as globules, suggesting dissolution of the sulfide melt by a volatile phase, which was likely oxidized (Fig. DR1g; Nadeau et al., 2010). Pyrite occurs as an interstitial phase within biotite-orthoclase assemblages in the amphibolites and garnet amphibolites, and is irregular in shape, suggesting a secondary or metamorphic origin. Microprobe analyses of some pyrites show Au enrichment of 170–580 ppm (just above the detection limit of 140 ppm; Table DR2).

The lack of quenched margins (Figs. 2A and 2B) and the occurrence of metamorphic mineral assemblages (Fig. 2D; Figs. DR1a–DR1d) in all of these xenoliths indicate that they are unlikely to be autoliths formed during magma crystallization or enclaves formed by injection of mafic melts into the felsic magma chamber. The garnet amphibolite xenoliths show typical retrograde textures, including (1) symplectite composed of fine-grained diopside, pargasite, and magnetite (Fig. DR1e) formed by the decompressional breakdown of garnet (Zhao et al., 2003) and (2) finegrained assemblages of anhedral albite, pargasite, and magnetite around coarse diopside grains (Fig. DR1f), likely formed by the breakdown of the Ca-Tschermaks components in pyroxene (Core et al., 2006). These



Figure 2. A, B: The xenoliths and their mineral assemblages. C: Electron probe-scanned element distribution of the globular sulfide phases in garnet. Cp—chalcopyrite; Gt—garnet; Po—pyrrhotite. D: Garnet amphibolite (sample LH14–53) consisting of two stages of amphibole (Am), garnet, and minor plagioclase (PI), pargasite, and biotite. Ap—apatite; Rt—rutile; Di—diopside.

observations indicate that the xenoliths underwent high-pressure metamorphism and later retrograde metamorphism during exhumation (Figs. DR1g and DR1h). The clinopyroxene geothermobarometer (Ravna, 2000) and garnet-clinopyroxene Fe-Mg geothermometer (Mercier, 1980) yield temperature-pressure estimates from 642 °C to 675 °C and 1354 MPa to 1560 MPa for the garnet amphibolites (Table DR3); this suggests highpressure eclogitic facies metamorphism at the crustal base (~41–52 km, based on amphibolite density of 3.0 g/cm³). By contrast, the amphibolite xenoliths show massive and gneissic structures and formation at 832 MPa (Al-in-amphibole barometer; Table DR4), corresponding to a metamorphic depth of ~27 km. We therefore argue that the garnet amphibolite and amphibolite xenoliths were derived from the bottom (~45 km) and upper part (~27 km) of the lower continental crust, respectively.

Zircon age populations provide further evidence for the origin of these xenoliths. Two groups of zircons with distinct ages have been recognized in these xenoliths (Fig. DR2; Table DR5). Older inherited zircons show a prominent age cluster ca. 773 Ma in the Beiya amphibolites (Fig. DR2a) and ca. 794 Ma in the Liuhe garnet amphibolites (Fig. DR2b) that approximate the peak of Neoproterozoic arc magmatism in the western Yangtze craton (ca. 813 Ma; W. Sun et al., 2009). These U-Pb ages also coincide with the crustal growth period (1000-740 Ma) of the craton (W. Sun et al., 2009). Younger zircons from the Liuhe xenoliths occur as overgrowth rims around older zircons, or as interstitial grains, both vielding similar U-Pb age clusters at 35.6 Ma (Fig. DR2c). This age group is slightly older than the host syenite $(34.6 \pm 0.4 \text{ Ma}; \text{Fig. DR2c})$, suggesting the growth of younger zircons during igneous remobilization of the xenoliths. Most Neoproterozoic and Cenozoic zircon samples are typical of igneous zircons (Fig. DR3). By contrast, one Neoproterozoic zircon grain (ca. 738 Ma) lacks an Eu anomaly, and has a flat heavy rare earth element (HREE) pattern, suggesting its growth in the presence of garnet (Fig. DR3). This metamorphic zircon suggests that the eclogitic facies metamorphism occurred ca. 738 Ma.

Whole-rock analyses indicate that these xenoliths show a close geochemical affinity with the Neoproterozoic arc plutons (Fig. DR4; Table DR6). They have relatively high Y (>15 ppm) and low Sr/Y (< 30) (Fig. DR4b) and show enrichment in large ion lithophile elements and depletion in high field strength elements (Fig. DR4c), which are characteristics typical of arc magmas derived from the metasomatic mantle wedge. The

¹GSA Data Repository item 2017181, petrography of xenoliths, Figures DR1–DR6, and Tables DR1–DR11, is available online at http://www.geosociety.org /datarepository/2017/ or on request from editing@geosociety.org.

inherited zircons from the garnet amphibolites yield $\varepsilon_{\rm Hf}$ values of +0.7 to +14.8 (Fig. DR5; Table DR7) that overlap with those of the Neoproterozoic arc rocks (Zhao and Zhou, 2007). Identical zircon Hf isotopic arrays for the xenoliths and arc plutons ca. 794 Ma suggest binary mixing between asthenospheric melts and crustal materials during their generation (Fig. DR5). The least-assimilated xenoliths and arc plutons have high $\varepsilon_{\rm Hf}$ values (\geq 5), similar to primitive arc magmas. These data indicate that both the garnet amphibolite and amphibolite xenoliths are comagmatic in origin and that their protoliths are likely lower crustal cumulates and residue of the Neoproterozoic arc magmas, respectively (Fig. 3), which, as juvenile components, led to the crustal growth of the western Yangtze craton.

Au-Cu ENRICHMENT AND DEPLETION IN THE XENOLITHS

The Liuhe xenoliths show variable concentrations of Au, Cu, and S (Table DR6). There is no correlation of Cu-Au contents with proportions of sulfides. Most garnet amphibolite xenoliths have lower metal contents (Au < 3 ppb; Cu < 10 ppm) than normal continental crust (Rudnick and Gao, 2003), and only a few show moderate enrichments of Au (6–16 ppb) and Cu (to 82 ppm). The amphibolite xenoliths have slightly higher Au (5–6 ppb), but remarkably lower Cu (15–18 ppm) contents than calc-alkaline arc magmas (1–2 ppb Au, 50–90 ppm Cu) (Lee et al., 2012).

The Au-poor garnet amphibolites have high modal garnet (>10 vol%) and diopside (>15 vol%; Fig. 2D) (Table DR3). Accordingly, these xenoliths yield lower contents of mobile elements (Figs. DR6c–DR6f) and light REEs than their Au-rich counterparts (Table DR6). This suggests that Au and Cu in the lower crustal cumulates have been in part extracted by fluids liberated during high-grade metamorphism (Cameron, 1989).

By contrast, the Au-rich (6–16 ppb) garnet amphibolites contain low modal garnet and diopside (<10 vol%), and have high Au/Cu ratios (as



Figure 3. Illustrations of the refertilization process and its relationship with the porphyry Cu-Au system at the cratonic edge. The Neoproterozoic arc magmas derived from subduction-modified metasomatic mantle have ponded at the bottom of the cratonic crust, forming the refertilized juvenile lower crust (i.e., garnet amphibolite). Subsequent prograde metamorphism resulted in releasing of Cu-Au-bearing fluids from the juvenile lower crust. The released fluids migrated upward and metasomatized the overlying mafic crust and arc magmatic residuals (i.e., amphibolite). A: During the later reactivation of the craton, invading mafic ultrapotassic melts triggered melting of the refertilized lower crustal cumulates, forming the Au-rich porphyry system at Beiya, China. SCLM—subcontinental lithospheric mantle. B: Mantle-derived mildly alkaline melts directly entrained the fragments of the garnet amphibolites and amphibolites, and ascended upward into the upper crust, forming barren syenite stocks at Liuhe.

high as 7.8×10^{-4}), coupled with relatively high contents of Cu (7.6–82.4 ppm) and Ni (143-318 ppm). Such Au enrichment cannot originate by mass exchange between the xenolith and enclosing magma, because the Liuhe syenites have very low Au contents (~1 ppb; Table DR6). Metasomatism by volatiles escaping from hydrous lamprophyres is also unlikely to explain this Au enrichment, because the Eocene lamprophyres exposed along the western margin of the Yangtze craton contain low Au concentrations (1-3 ppb) and lamprophyre melts have a low Au carrying capacity (Huang et al., 1999). Alternatively, the remnants of globular sulfides shielded by garnet grains in the garnet amphibolite xenoliths demonstrate Au enrichment in the lower crustal cumulates before high-grade metamorphism (Fig. 2C). This is consistent with the expectation that small amounts of sulfide (as melt or crystalline phases) can be present, due to the high sulfur contents in arc magmas that tends to settle out in cumulate zones due to density differences (Richards, 2009). As a consequence, the residual cumulates after high-grade metamorphism would be depleted in Cu-Au, as demonstrated by the low Cu contents of the Liuhe xenoliths.

We therefore argue that ponding of the Neoproterozoic arc magmas with sulfide accumulation at the crustal base had progressively replenished the cratonic lower crust in metal components. The remnants of magmatic sulfides in garnet amphibolite xenoliths and metasomatic refertilization in amphibolite xenoliths indicate that the fluids, released from the lower crustal cumulates during later prograde metamorphism, transported metal Cu-Au and metasomatized the overlying crust, before or during cratonic reactivation at 730 Ma (Fig. 3). The refertilized juvenile lower crust contains >16 ppb Au and >205 ppm Cu, prior to its dehydration and later remelting, based on the Au and Cu contents of the Liuhe Au-rich xenoliths, and considering remobilization of sulfides during later metamorphism.

FORMATION OF AU PORPHYRY BY REMELTING OF THE XENOLITHS

Previous studies indicate that the Beiya porphyries are characterized by high Sr/Y and La/Yb ratios coupled with low Y and Yb (He et al., 2015) (Fig. DR4b), showing geochemical affinity with adakites (Defant and Drummond, 1990). Beiya porphyries are thought to be generated by remelting of the Neoproterozoic lower crustal cumulates such as the xenoliths (He et al., 2015). The breakdown of minor sulfides in the lower crustal cumulates during later remelting would be expected to produce a sulfur-poor magma (Richards, 2009). This is consistent with two facts observed at Beiya: (1) the lack of high-sulfidation alteration, and (2) small mass ratios of sulfides to magnetite (1:10) in the Au orebodies (He et al., 2015). As low sulfur contents only caused minimal sulfide saturation and consequent Cu-Au sequestration from the magma (Chiaradia et al., 2012), we therefore presume that the ore-forming magma has an Au/Cu ratio similar to that of the resultant porphyries at Beiya. The least-altered monzogranite porphyries at Beiya have Au contents of 8-39 ppb and Cu contents of 4-83 ppm (He et al., 2015); the average Au/Cu ratio is 1.4 × 10^{-4} , an order of magnitude higher than that of normal arc magmas (0.4 × 10^{-5} ; Gill, 1981), but identical to that (average = 1.5×10^{-4}) of the Liuhe-Beiya xenoliths. Ulrich et al. (2009) found that the Au/Cu ratio of two giant PCDs is identical to the bulk Au/Cu ratio of primary high-temperature ore-forming fluids, which, in turn, depends on the ratio in the magma source for an S-poor magmatic-hydrothermal system. If this is the case, then the consistency of Au/Cu ratios in the Beiya porphyries and Liuhe Au-rich xenoliths indicates that remelting of the Au-enriched lower crustal cumulates could produce the S-poor porphyry Au-Cu system at Beiya.

The source for Au can be further constrained by mass-balance calculation, based on the total metal content of the Beiya deposit. Assuming that the Liuhe Au-rich xenolith (Au = 16 ppb) represents the source of the Beiya felsic magma, 20% remelting (Fig. DR4b) could produce an unusually Au-rich (>100 ppb) magma with ~9 km³ volume; this is consistent with aeromagnetic evidence and the exposed area of 7 monzogranite stocks at Beiya (He et al., 2015) and with the quantity of magma required for most PCDs (Cline and Bodnar, 1991). To supply the Au content (304 t Au) of the deposit only requires extraction of 38% Au from the same magma. These results suggest that the breakdown of sulfides in Au-fertilized juvenile lower crust during later remelting played a significant role in the generation of Au-rich PCDs at cratonic edges that were once arcs.

Compared to mildly alkaline mantle-derived magmas, ultrapotassic magmas have high H_2O solubility, and can therefore release H_2O to trigger partial melting of metal-fertilized lower crust (amphibolite and garnet amphibolite) to generate fertile adakite-like magmas. The common occurrence of coeval lamprophyres and adakite-like porphyries at Beiya deposit supports this link. By contrast, the mildly alkaline mantle-derived magmas from Liuhe were less likely to trigger extensive lower crustal melting due to low H_2O solubility, and therefore generated sygnitic magmas with sparse mineralization.

IMPLICATIONS FOR EXPLORATION

Our results demonstrate that arc magmas derived from metasomatic mantle have most likely ponded at the crustal base, leading to crustal growth and metal fertilization in the Neoproterozoic. The initial Au-Cu contents and Au/Cu ratios in the fertilized juvenile lower crust likely depend on the amount of accumulated sulfide in the magma ponding at its base (Richards, 2009). These ore-forming components (Au, Cu), although locally remobilized during later metamorphism, could be preserved in the lower continental crust, largely due to the secular stability of the craton. The incubation time between crustal base metal enrichment and reactivation can be short in a successive process from subduction to collision, such as in the Gangdese porphyry deposits (Hou et al., 2015), or it can be delayed until later magmas or fluids ascending upward through cratonic boundaries or strike-slip faults (Lee et al., 2012; Griffin et al., 2013).

There is increasing evidence that such fertilization in the cratonic SCLM and lower crust may play an important role in the formation of some Au-rich deposits worldwide (Lee et al., 2012). Examples include the Cenozoic giant porphyry deposits in the eastern Rocky Mountains, western USA, where Cenozoic melting of SCLM metasomatized by Proterozoic slab fluids provides the metal endowment (Pettke et al., 2010), and the giant Cretaceous Au ore provinces along the margins of the eastern Asian cratons (Goldfarb et al., 2007) (Fig. 1A), where Mesozoic tectonic reactivation resulted in the release of Au from the Proterozoic Au-rich deep crust into magmatic or hydrothermal systems. We therefore suggest that reactivated paleomagmatic arcs at the edge of cratons intruded by later non-arc felsic magmas have the potential to contain Au ore systems, especially porphyry Au deposits.

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