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Metasomatized mantle sources for orogenic gold deposits hosted in high-grade metamorphic rocks: Evidence from Hg isotopes

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ABSTRACT

Investigation of Hg isotope ratios of gold-related sulfides and penecontemporaneous mafic dikes from four orogenic gold provinces on the margins of the North China Craton and Yangtze Craton identifies three orogenic gold deposit (OGD) groups from different tectonic regimes. Ore-related sulfides of group 1 OGDs and mafic dikes from the craton margin reworked via oceanic subduction mostly have positive Δ^{199}Hg values. The group 2 OGDs and mafic dikes from the margin that witnessed complex oceanic and continental subductions have mixed positive to negative Δ^{199}Hg values. The group 3 OGDs on the margin that experienced continental subduction have dominantly negative Δ^{199}Hg values. These isotopic differences indicate subduction histories have a first-order control on the distinct sources for the OGDs. It indicates that OGDs were derived from fluids from the mantle lithosphere metasomatized by contrasting subduction components, not from metamorphic fluids as is widely accepted. Group 1 OGDs and dikes were sourced from metasomatized mantle, which inherited the positive Δ^{199}Hg of both recycled marine sediments and seawater during oceanic subduction, whereas group 3 with negative Δ^{199}Hg was derived from mantle lithosphere metasomatized by subduction of mainly continental components. This genetic model identifies regions with high-grade metamorphic rocks above metasomatized mantle lithosphere as promising new OGD exploration targets.

INTRODUCTION

The genesis of orogenic gold deposits (OGDs), accounting for ~40% of global gold reserves, is highly controversial (Goldfarb and Groves, 2015). The traditional model for the genesis of OGDs is metamorphic devolatilization of crustal rocks at the transition between amphibolite and greenschist facies, considering that most Precambrian OGDs are hosted by greenschist-facies rocks (Phillips and Powell, 2010; Tomkins, 2010; Goldfarb and Pitcairn, 2023). However, this model fails to explain the genesis of OGDs hosted by amphibolite- to granulite-facies metamorphic rocks, the proposed sources of crustal metamorphic fluids (Groves et al., 2020). These deposits include the giant OGDs in the North China Craton (Wang et al., 2022), Yangtze Craton (Zhao et al., 2019; Fig. 1), and Dharwar craton (Kolb et al., 2015). Contrasting models, including devolatilization of deeper crustal amphibolite- to granulite-facies rocks (Bark et al., 2021), devolatilization of a subducting slab and overlying sediment wedge (Groves et al., 2020), or devolatilization of metasomatized mantle lithosphere (Deng et al., 2020; Zhao et al., 2022a), have been proposed (Fig. 2).

Mercury isotopes that undergo both mass-dependent fractionation (MDF) and mass-independent fractionation (MIF) provide a potential proxy for probing the ore source (Bergquist and Blum, 2007). Hg-MDF is recorded from systems that have undergone various physical, chemical, and biological processes, whereas Hg-MIF is generated mainly by Hg(II) photoreduction in the atmosphere-ocean system (Blum et al., 2014). The primitive mantle has a Δ^{199}Hg of ~0‰ (Moynier et al., 2021), whereas continental components and marine systems have negative and positive Δ^{199}Hg values, respectively (Blum et al., 2014). Large variations of Δ^{199}Hg in hydrothermal systems from various geological settings reveal that Hg recycling in plate subduction or magmatic processes can be traced (Smith et al., 2008; Yin et al., 2019; Deng et al., 2021). Therefore, Hg isotopes may serve as a metallogenetic tracer for evaluating the contentious metal sources for the OGDs.

GEOLOGICAL SETTING

Tectonic Setting

The North China Craton was formed by amalgamation of several Archean microcontinents from 2.1 to 1.8 Ga (Zhao and Cawood, 2012). In the Neoproterozoic, the Yangtze Craton was affected by oceanic subduction (Zhou et al., 2002). The northern and southern margins of the North China Craton were subjected to oceanic subduction in the Paleozoic until ca. 250 Ma. In the Triassic, collision between the North China and Yangtze Cratons formed high-pressure metamorphic rocks in the Dabie and Sulu Belts (Fig. 1B; Zheng et al., 2018). Extensive Cretaceous lithospheric thinning occurred in the eastern North China Craton in response to the westward subduction of the Paleo-Pacific oceanic plate (Zheng et al., 2018).

Gold Mineralization

The sampled OGDs in the North China and Yangtze Cratons are mainly mesozonal (Groves et al., 1998) quartz-vein and disseminated-veinlet deposits that formed in four episodes

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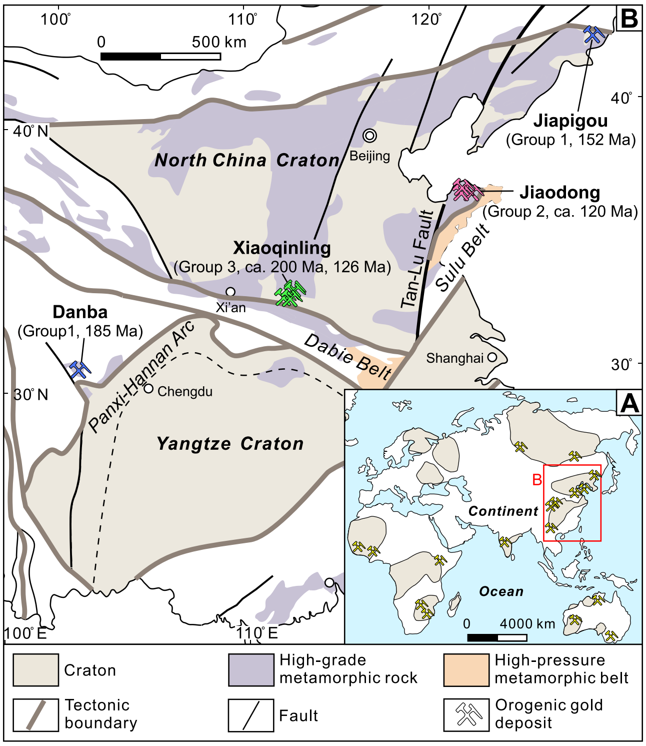

Figure 1. (A) Distribution of orogenic gold deposits (OGDs) along craton margins (after Groves et al., 2020). (B) Locations of the sampled three groups of OGDs in the North China and Yangtze Cratons (Wang et al., 2022).

(Fig. 1B): (1) Triassic OGDs such as Dahu in the Xiaoqinling province, along the southern margin of the North China Craton; (2) the Early Jurassic hypozonal Danba deposit on the western margin of the Yangtze Craton (Zhao et al., 2019); (3) Late Jurassic OGDs including Jiapigou on the northern margin of the North China Craton; and (4) the giant Early Cretaceous Jiaodong and Xiaoqinling gold provinces on the eastern and southern margins, respectively, of the North China Craton (Deng et al., 2020). Mafic dikes broadly coeval with gold mineralization are associated with most gold deposits, with their source magmas considered to have originated from low-degree partial melting of metasomatized mantle lithosphere (Deng et al., 2020). Reliable geochronology indicates that OGDs in the Yangtze Craton formed much later than the Neoproterozoic oceanic subduction, and those in the North China Craton formed later than the Triassic continental collision and coeval to the Mesozoic oceanic subduction (Wang et al., 2022).

SAMPLES AND ANALYTICAL METHODS

A total of 150 samples, including 48 ore-related sulfides, 6 wall-rock sulfides, 14 mafic rocks, 54 granites, and 28 metamorphic rocks, were collected from four gold provinces (Fig. S1 in the Supplemental Material[1]). Total Hg concentrations (THg) and Hg isotope ratios were analyzed at the Institute of Geochemistry, Chinese Academy of Sciences, following previous methods (see Text S1).

Hg-MIF is reported in Δ notation, which describes the difference between the measured δ^{xxx}Hg and the theoretically predicted δ^{xxx}Hg, in units of per mil (‰):

$$\Delta^{xxx}\text{Hg} \approx \delta^{xxx}\text{Hg} - \delta^{202}\text{Hg} \times \beta, \qquad (1)$$

where *xxx* is 199, 200, or 201, and β is 0.2520 for ^{199}Hg, 0.5024 for ^{200}Hg, and 0.7520 for ^{201}Hg (Blum and Bergquist, 2007).

RANGES AND GROUPS OF Hg CONCENTRATIONS AND ISOTOPIC RATIOS

Mercury concentrations and isotopic ratios of the samples are summarized in Table S1 and shown in Figure S2 (see Text S2). Hg-MIF relates mainly to photochemical processes with little contribution from complex biogeochemical cycling and mineralization processes. Therefore, Hg-MIF values of ore-related sulfides are indicative of Hg sources, as emphasized in the following sections.

THg concentration varies by orders of magnitude among ores (4.99–2740 ppb), with less fluctuation in wall rocks (55.6–275 ppb) and mafic rocks (0.397–4.45 ppb). However, the 54 granites and 28 metamorphic rocks from the Jiaodong and Xiaoqinling provinces have extremely low THg concentrations (<0.1 ppb), which inhibit Hg isotope determination (Fig. S2).

[1]Supplemental Material. Detailed analytical methods, Figures S1–S3, and Table S1. Please visit https://doi.org/10.1130/GEOL.S.24424462 to access the supplemental material; contact editing@geosociety.org with any questions.

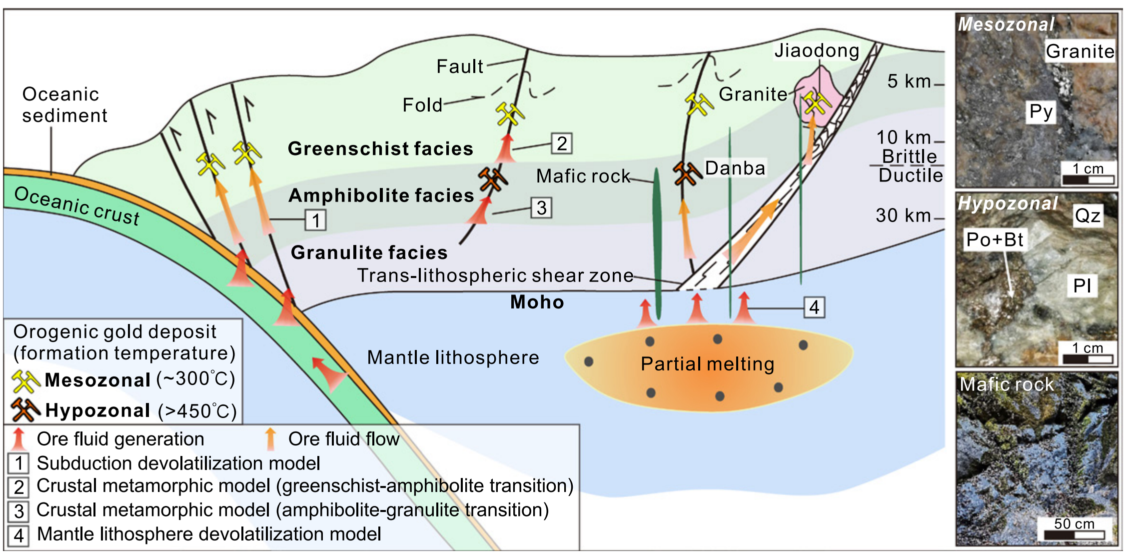

Figure 2. Competing crustal and sub-crustal genetic models for orogenic gold deposits (OGDs) showing geological setting for selected OGDs in this study. Mineral abbreviations in subfigures: Bt—biotite; Pl—plagioclase; Po—pyrrhotite; Py—pyrite; Qz—quartz.

$\Delta^{199}Hg$ of the samples exhibit large ranges of −0.22‰ to 0.29‰, given the analytical uncertainty of ±0.06‰ (2SD). There is a positive correlation between $\Delta^{201}Hg$ and $\Delta^{199}Hg$ values (Fig. 3), consistent with that of the atmosphere-ocean system (Blum et al., 2014). The $\Delta^{199}Hg$ values of OGDs from this study differ from those of Carlin-style gold deposits in China, which collect Hg from magmatic or regional wall-rock sources and display mainly negative to near-zero $\Delta^{199}Hg$ values (Fig. 3B; Yin et al., 2019). The values also deviate from those of epithermal gold deposits, which derived from mantle metasomatized by oceanic subduction (Deng et al., 2022b). This implies the OGDs have more complicated Hg sources.

According to $\Delta^{199}Hg$ signals, the OGDs from the four gold provinces are divided into three groups: (1) those in the Yangtze Craton and the northern margin of the North China Craton, including the Danba and Jiapigou deposits, with mainly positive $\Delta^{199}Hg$ (Danba Craton: 0.03‰ to 0.24‰, median = 0.11‰; Jiapigou: −0.03‰ to 0.29‰, median = 0.12‰; Fig. 3); (2) those in the Jiaodong province with a mixed signal of $\Delta^{199}Hg$ (−0.22‰ to 0.24‰, median = 0.03‰); and (3) those in the Xiaoqinling district, on the southern margin of the North China Craton, which display predominantly negative values (−0.19‰ to 0.09‰, median = −0.08‰).

Notably, $\Delta^{199}Hg$ values of group 1 and group 2 OGDs deviate from those of local Precambrian basement, which has mainly negative $\Delta^{199}Hg$ values (Fig. 3B). Mafic dikes from group 1 OGDs have mostly positive $\Delta^{199}Hg$ values ranging from −0.11‰ to 0.29‰, whereas those from group 2 OGDs are dominated by more negative $\Delta^{199}Hg$ values ranging from −0.17‰ to 0.18‰. The ranges of $\Delta^{199}Hg$ values for mafic dikes from group 1 and group 2 OGDs are broadly consistent with those of gold ores from the two groups (Fig. 3A). The $\Delta^{199}Hg$ values of Danba ore-related pyrrhotite (0.03‰ to 0.24‰) contrast with those of wall-rock pyrrhotite (−0.02‰ to 0.12‰).

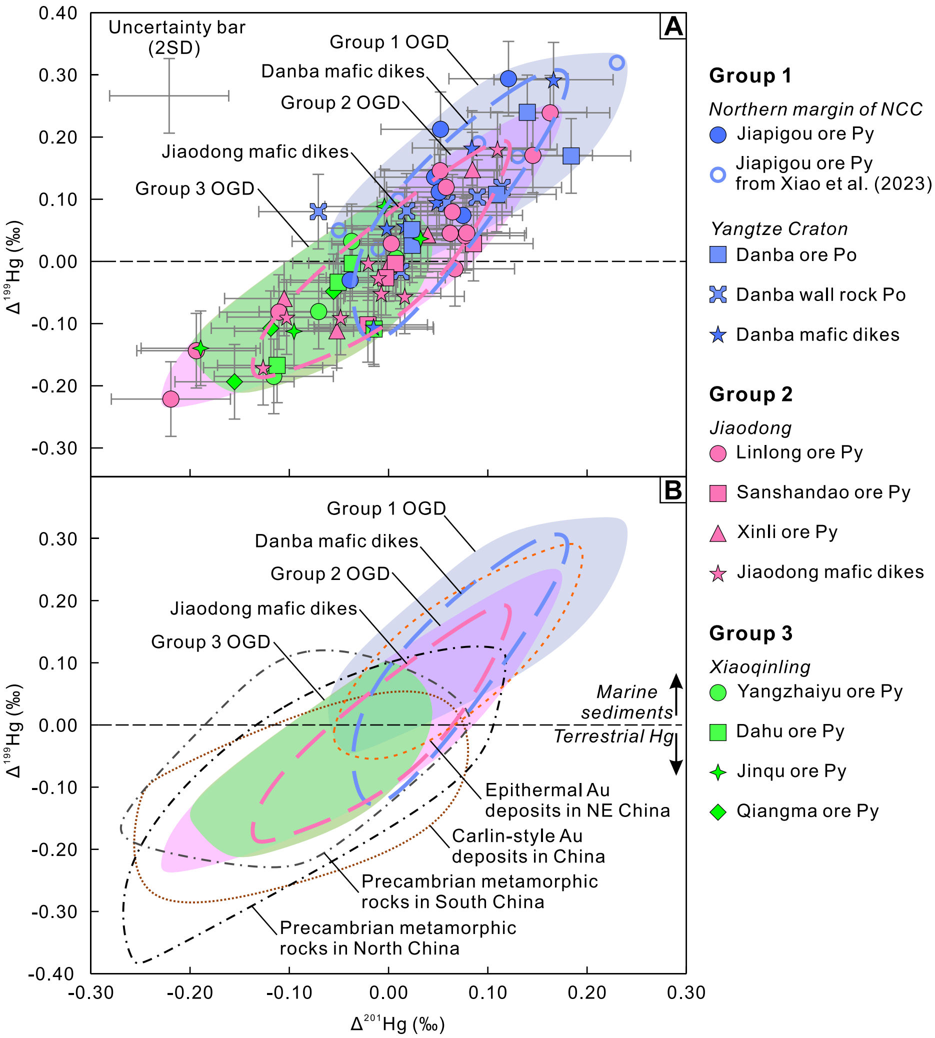

Figure 3. $\Delta^{199}Hg$ versus $\Delta^{201}Hg$ diagrams for the studied orogenic gold deposits (OGDs) and mafic dikes (A) with the reference of various Hg reservoirs (B). Data sources: terrestrial Hg—Blum et al. (2014); marine sediments—Yin et al. (2015), Meng et al. (2019); Precambrian metamorphic rocks in South China and North China—Deng et al. (2022b), Xiao et al. (2023); Carlin-style Au deposits—Yin et al. (2019); epithermal Au deposits in NE China—Deng et al. (2021). Abbreviations: NCC—North China Craton; Py—pyrite; Po—pyrrhotite.

Lack of Contribution of Hg from Wall Rocks

The disseminated-veinlet ores in the Jiaodong gold deposits are suggested in previous studies to have formed via sulfidation during interaction between ore fluids and granite wall rocks (Wang et al., 2022). However, granites have near-zero $\Delta^{199}Hg$ (0.02‰ ± 0.08‰) and extremely low THg (<0.1 ppb here and 4.12 ± 4.43 ppb from Deng et al. [2022a]), indicating that wall-rock reactions cannot have contributed enough Hg to affect ore Hg isotope ratios. This is supported by contrasting $\Delta^{199}Hg$ ranges between gold ores and wall rocks at Danba (Fig. 3A).

Oceanic Subduction Delivers Positive Hg-MIF Signatures for Group 1 OGDs

Meteoric water displays positive $\Delta^{199}Hg$ values (Chen et al., 2012), which could be responsible for the $\Delta^{199}Hg$ values of group 1 OGDs (Fig. 3A). However, this is excluded based on: (1) OGDs have a primary deep-sourced fluid source (Goldfarb and Groves, 2015); and (2) meteoric water has extremely low Hg contents (1–10 ng/L; Chen et al., 2012). In addition, leaching of Hg from Precambrian basement rocks by meteoric water or devolatilization of Precambrian basement rocks fails to explain the Hg-MIF signatures of group 1 OGDs, given that most basement rocks have negative $\Delta^{199}Hg$ values (Fig. 3B). The compatibility of dominantly positive $\Delta^{199}Hg$ values between group 1 OGDs and associated mafic dikes, however, reflects a Hg source from metasomatized mantle lithosphere with positive $\Delta^{199}Hg$ values induced via earlier oceanic subduction (Fig. 4A).

The craton margins hosting group 1 OGDs were subjected to oceanic subduction in both the Neoproterozoic and Paleozoic (Zhou et al., 2002). Involved marine sediments and seawater have positive $\Delta^{199}Hg$ values (0‰ to 0.3‰: Štrok et al., 2015; Yin et al., 2015; Meng et al., 2019), leaving $\Delta^{199}Hg$ fingerprints in the mantle lithosphere (Deng et al., 2022a) and derived group 1 OGDs (Xiao et al., 2023). Noble gas and halogen compositions of ore fluids at Danba and Jiapigou also indicate derivation from recycled altered ocean crust (Zhao et al., 2022a).

Combined Oceanic and Continental Subduction Delivers Positive to Negative Hg-MIF Signatures for Group 2 OGDs

The Precambrian basement underlying Jiaodong is relatively homogenous, with εNd(t) at ~0% (Wan et al., 2021) and generally negative $\Delta^{199}Hg$ values (Fig. 3B), which are incapable of

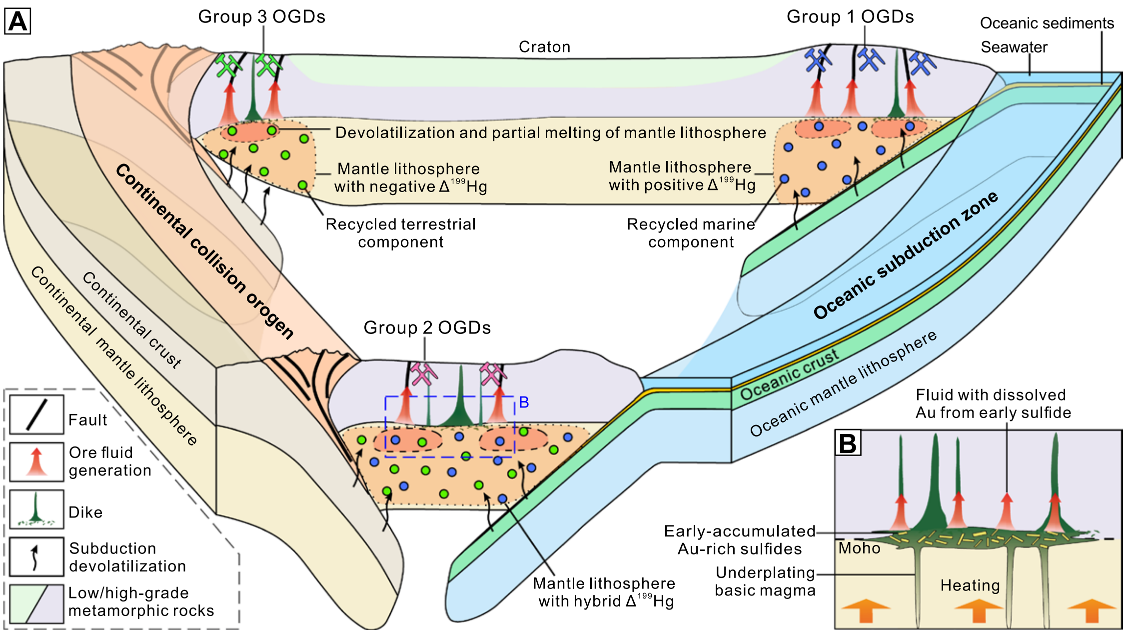

Figure 4. Schematic genetic model for orogenic gold deposits (OGDs) hosted by high-grade metamorphic rocks along craton margins. A is the preferred model, with B a potential alternative model.

explaining the positive to negative Δ^{199}Hg values in group 2 Jiaodong OGDs. Given that group 2 OGDs display a similar range of Hg-MIF values to the regional mafic dikes (Fig. 3) and both have similar Pb isotope ratios (Tan et al., 2012), metasomatized mantle lithosphere is a viable source for the OGDs. This is consistent with complex subduction below the Jiaodong province in which mantle lithosphere was affected by continental subduction of the Yangtze Craton and oceanic subduction of the Paleo-Pacific plate, as also indicated by Os-Sr-Nd isotope compositions of mafic rocks (Zheng et al., 2018). Such subduction would have caused metasomatism of mantle lithosphere with introduction of both positive and negative Hg-MIF signatures, elegantly explaining the Hg isotope heterogeneity of group 2 OGDs (Fig. 4A). It is also possible that delamination of deep metamorphic rocks into the mantle lithosphere during lithospheric extension on the North China Craton margin could have contributed to the negative Δ^{199}Hg signatures of some group 2 OGDs.

Continental Subduction Delivers Negative Hg-MIF Signatures for Group 3 OGDs

Two different Hg sources can potentially account for the overall negative Δ^{199}Hg values of group 3 OGDs in the Xiaoqinling province: (1) a source from mantle lithosphere metasomatized primarily by a continental component; or (2) direct derivation from regional metamorphic continental rocks via devolatilization. Given that metasomatized mantle lithosphere provided the fluid and Hg source for the group 1 and 2 OGDs on the craton margins (Figs. 1B and 4A), the first source is the most logical based on the principle of Occam's razor that assumptions should not be multiplied without necessity. This is supported by the tectonic setting of the Xiaoqinling province, which experienced Triassic continental subduction of the Yangtze Craton (Zheng et al., 2018) and is located farther inboard than the Jiaodong province and thus experienced a lower proportion of oceanic crust from Paleo-Pacific subduction. A crustal metamorphic source is inconsistent with the sulfur and lead isotope ratios of ore sulfides ($\delta^{34}S = \sim 8‰–28‰$ and $^{207}Pb/^{204}Pb = \sim 15.2‰–15.7‰$) in the Xiaoqinling province, which differ from those isotope ratios of the regional metamorphic rocks and the granites that formed via melting of lower crustal rocks (Zhao et al., 2022b).

FORMATION OF OGDs FROM METASOMATIZED MANTLE LITHOSPHERE

The new Hg isotope evidence is consistent with previous evidence for a sub-crustal source that includes (1) the occurrence of gold mineralization under retrograde metamorphic conditions in high-grade metamorphic terranes during which significant devolatilization was absent (Groves et al., 2020); and (2) a range of compatible geochemical and isotope evidence for a mantle lithosphere source (Zhao et al., 2022a). As discussed above, the new Hg isotope data for gold-related sulfides support not only a mantle lithosphere source but one that was metasomatized by various oceanic and continental components. A combination of compatible ore Δ^{199}Hg values and mafic rock Sr-Nd isotope ratios lend further support to this interpretation (Fig. S3) and to the indirect connection between broadly synchronous gold ores and basic magmatism.

The evolution of OGDs and basic magmas is illustrated in Figure 4. In this model: (1) early metasomatism of mantle lithosphere by spatially separated oceanic and continental subduction zones induced contrasting positive and negative Hg-MIF signatures, respectively; and (2) later tectono-thermal events, such as ca. 120 Ma asthenosphere upwelling in the North China Craton (Deng et al., 2020), induced devolatilization and partial melting of metasomatized mantle lithosphere that released voluminous auriferous ore fluid and basic magma, respectively. The basic magma likely ponded beneath the lower crust to potentially degas and release additional ore fluid (Fig. 4B; Hou et al., 2022; Holwell et al., 2022), with generation of additional auriferous fluids by direct degassing of metasomatized mantle lithosphere (Wang et al., 2022).

This study also indirectly supports models for derivation of fluids and metals for OGDs and other gold deposit types, including iron oxide copper-gold and others, such as epithermal deposits, from metasomatized mantle lithosphere as revealed by Os isotope ratios of Patagonia gold deposits in Argentina (Tassara et al., 2022).

CONCLUSIONS AND IMPLICATIONS

Three OGD groups and penecontemporaneous mafic dikes from different tectonic settings on the craton margins have distinctive but contrasting Hg isotope ratios. Gold-related sulfides of group 1 OGDs and mafic dikes from the craton margin reworked via oceanic subduction mostly have positive Δ^{199}Hg values, whereas those OGDs of group 3 on the margin that experienced continental subduction have contrasting negative Δ^{199}Hg values. The group 2 OGDs and mafic dikes that formed more inboard of the craton margins have mixed Δ^{199}Hg values. These isotopic differences indicate distinctly different sources for the OGDs on or near craton margins with varying subduction histories, suggesting that gold ores were derived from fluids from mantle lithosphere metasomatized by contrasting subducted oceanic and continental components, not primarily from crustal metamorphic fluids. This genetic model defines regions with high-grade metamorphic rocks above metasomatized mantle lithosphere as promising OGD exploration targets in new exploration spaces.

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