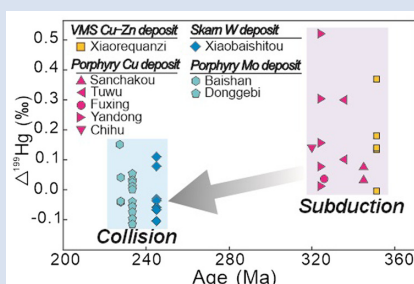


Hg isotopes constrain metallogenic effects of the Wilson Cycle of the Paleo-Asian Ocean

X. He¹, X. Deng^{1*}, M.J. Brzozowski¹, Y. Wu², J. Zhang⁴, C. Wu¹, Y. Chen^{2,5}, R. Yin^{3*}

Abstract



The Central Asian Orogenic Belt (CAOB) hosts a variety of hydrothermal mineralised systems that formed during the opening, development and closure of the Paleo-Asian Ocean. Mercury is an important ore-forming metal in hydrothermal systems. Mercury photoreactions on the surface of Earth produce negative and positive $\Delta^{199}\text{Hg}$ signals in terrestrial and marine sediments, respectively. Here, we observe positive $\Delta^{199}\text{Hg}$ values in 345 to 320 Ma porphyry Cu deposits ($+0.15 \pm 0.15$ ‰, s.d.), and a 355 Ma volcanogenic massive sulfide Cu–Zn deposit ($+0.16 \pm 0.13$ ‰, s.d.) in the CAO, which reflect the recycling of marine sediments during oceanic subduction and circulation of seawater in an extensional tectonic regime, respectively. In contrast, we observe negative $\Delta^{199}\text{Hg}$ values in 232 to 227 Ma porphyry Mo deposits (-0.01 ± 0.07 ‰, s.d.) and a 245 Ma skarn W deposit (-0.02 ± 0.08 ‰, s.d.), which reflect the mixing of recycled Hg from marine and terrestrial sediments during continental collision. This study is useful in understanding the metallogenic effects of the Wilson Cycle and highlights that plate tectonics can cause intensive mobilisation, migration and mineralisation of Hg from subducted oceanic and continental crust to form Hg-bearing hydrothermal systems in long lived accretionary orogens.

Received 22 January 2025 | Accepted 14 April 2025 | Published 20 May 2025

Introduction

The Central Asian Orogenic Belt (CAOB), known as the world's largest Phanerozoic continental accretionary orogen (Sengör *et al.*, 1993), hosts a variety of hydrothermal mineralised systems, including porphyry Cu deposits, volcanogenic massive sulfide Cu–Zn deposits, porphyry Mo deposits, and skarn W deposits (Pirajno *et al.*, 2011). These hydrothermal systems formed in response to the Wilson Cycle (*i.e.* opening, development and closure) of the Paleo-Asian Ocean. In the early to middle stages of the Wilson Cycle, Paleo-Asian Ocean subduction and associated accretionary orogeny led to the occurrence of porphyry Cu deposits and volcanogenic massive sulfide Cu–Zn deposits between 355 to 320 Ma in the Eastern Tianshan segment of the CAO (Mao *et al.*, 2020; Wang *et al.*, 2021). In the late stage of the Wilson Cycle, continental collision following closure of the Paleo-Asian Ocean led to the formation of porphyry Mo deposits and skarn W deposits between 245 and 227 Ma in the Eastern Tianshan segment of the CAO (Deng *et al.*, 2017; Wu *et al.*, 2017). The diverse hydrothermal systems in the CAO reflect the metallogenic effects of the Wilson Cycle.

Mercury (Hg) is an important ore-forming metal in hydrothermal systems. Hg isotopes display unique mass-dependent fractionation (MDF, $\delta^{202}\text{Hg}$) and mass-independent fractionation (MIF, $\Delta^{199}\text{Hg}$) (Blum and Berquist, 2007). MDF of Hg isotopes can

occur during a large number of geochemical processes (*e.g.*, Blum *et al.*, 2014). However, MIF of Hg isotopes is predominantly associated with photoreactions in the land–ocean–atmosphere systems (Blum and Berquist, 2007). Hg(II) photoreduction has resulted in positive $\Delta^{199}\text{Hg}$ values in marine sediments and negative $\Delta^{199}\text{Hg}$ values in terrestrial sediments (Blum *et al.*, 2014). The primitive mantle displays near zero $\Delta^{199}\text{Hg}$ values (Moynier *et al.*, 2021). Considering that mineralogical and petrological processes cannot generate MIF of Hg isotopes, the unique $\Delta^{199}\text{Hg}$ signals across marine, terrestrial and mantle reservoirs allow for identification of the source of Hg and, by extension, other metals in hydrothermal systems (Yin *et al.*, 2022, 2024). Non-zero $\Delta^{199}\text{Hg}$ values have been detected in hydrothermal systems across diverse environments, demonstrating the recycling of terrestrial- or marine-derived Hg through oceanic subduction and continental collision (*e.g.*, Yin *et al.*, 2024).

This study presents the Hg isotope compositions of five porphyry Cu deposits, two porphyry Mo deposits, one volcanogenic massive sulfide Cu–Zn deposit, and one skarn W deposit in the Eastern Tianshan segment of the CAO to clarify the metallogenic impacts of the Wilson Cycle. We demonstrate a large variation of $\Delta^{199}\text{Hg}$ in these deposits (-0.11 to $+0.51$ ‰), which reflects the recycling of marine- and terrestrial-derived Hg related to oceanic subduction and continental collision, respectively. During the Wilson Cycle, plate subduction contributed to mobilisation of

1. School of Earth Science and Resources, Chang'an University, Xi'an 710054, China
2. Xinjiang Research Center for Mineral Resources, Xinjiang Institute of Ecology and Geography, Chinese Academy of Sciences, Urumqi 830011, China
3. State Key Laboratory for Critical Mineral Research and Exploration, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, China
4. State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences, Beijing 100083, China
5. School of Earth and Space Science, Peking University, Beijing 100871, China
* Corresponding authors: Xiaohua Deng; Runsheng Yin (email: dxh198411@126.com; yinrunsheng@mail.gyig.ac.cn)

Hg from subducted oceanic and continental crusts to form various Hg-bearing hydrothermal systems in the accretionary orogen.

Geological Background

The CAOBS is situated between the Siberian Craton to the north, Tarim and North China cratons to the south, and the East European Craton to the west (Fig. 1a; Jahn, 2004). The Eastern Tianshan metallogenic belt along the southern CAOBS comprises the Harlik Belt, the Jueluotage Belt and the Central Tianshan Block, which are bound by the Kalamaili and Aqikekuduke faults (Fig. 1b; Qin *et al.*, 2002; Chen *et al.*, 2012). From north to south, the Jueluotage Belt is divided into the Dananhu-Tousuquan Arc, the Kangguer-Huangshan Shear Zone and the Aqishan-Yamansu Arc, by the Kangguer and Yamansu faults (Fig. 1c).

In the Carboniferous, double-sided subduction of the North Tianshan Ocean (NTO) formed the Dananhu-Tousuquan Arc and Aqishan-Yamansu Arc (Wang *et al.*, 2015). As shown in Figure 1c, the Dananhu-Tousuquan Arc contains numerous 355 to 320 Ma porphyry Cu deposits (*e.g.*, Tuwu, Yandong, Sanchakou, Fuxing and Chihu; Wang *et al.*, 2021;

He *et al.*, 2023) and ~355 Ma volcanogenic massive sulfide Cu-Zn deposits (*e.g.*, Xiaorequanzi; He *et al.*, 2020; Mao *et al.*, 2020). The Aqishan-Yamansu Arc includes numerous 330–300 Ma volcanic-hosted Fe deposits (*e.g.*, Yamansu; Huang *et al.*, 2018). The NTO closed during the Triassic, resulting in the collision of the Dananhu-Tousuquan Arc and Aqishan-Yamansu Arc (Xiao *et al.*, 2004; Mao *et al.*, 2022). This collision formed the Kangguer-Huangshan Shear Zone and numerous 232 to 227 Ma porphyry Mo deposits (*e.g.*, Baishan and Donggebi; Wu *et al.*, 2017). The main deposit in the Central Tianshan Block is the 245 Ma Xiaobaishitou skarn-type W deposit (Deng *et al.*, 2017).

Methods

In this work, 14 granite rocks and 23 sulfide ores were collected from 5 porphyry Cu deposits (Tuwu, Yandong, Sanchakou, Fuxing and Chihu), 1 volcanogenic massive sulfide Cu-Zn deposit (Xiaorequanzi), 2 porphyry Mo deposits (Baishan and Donggebi), and 1 skarn-type W deposit (Xiaobaishitou) in the Eastern Tianshan. The geological characteristics of these ore deposits are summarised in Table 1. The comprehensive details of the samples are provided in Supplementary Information Table S-1.

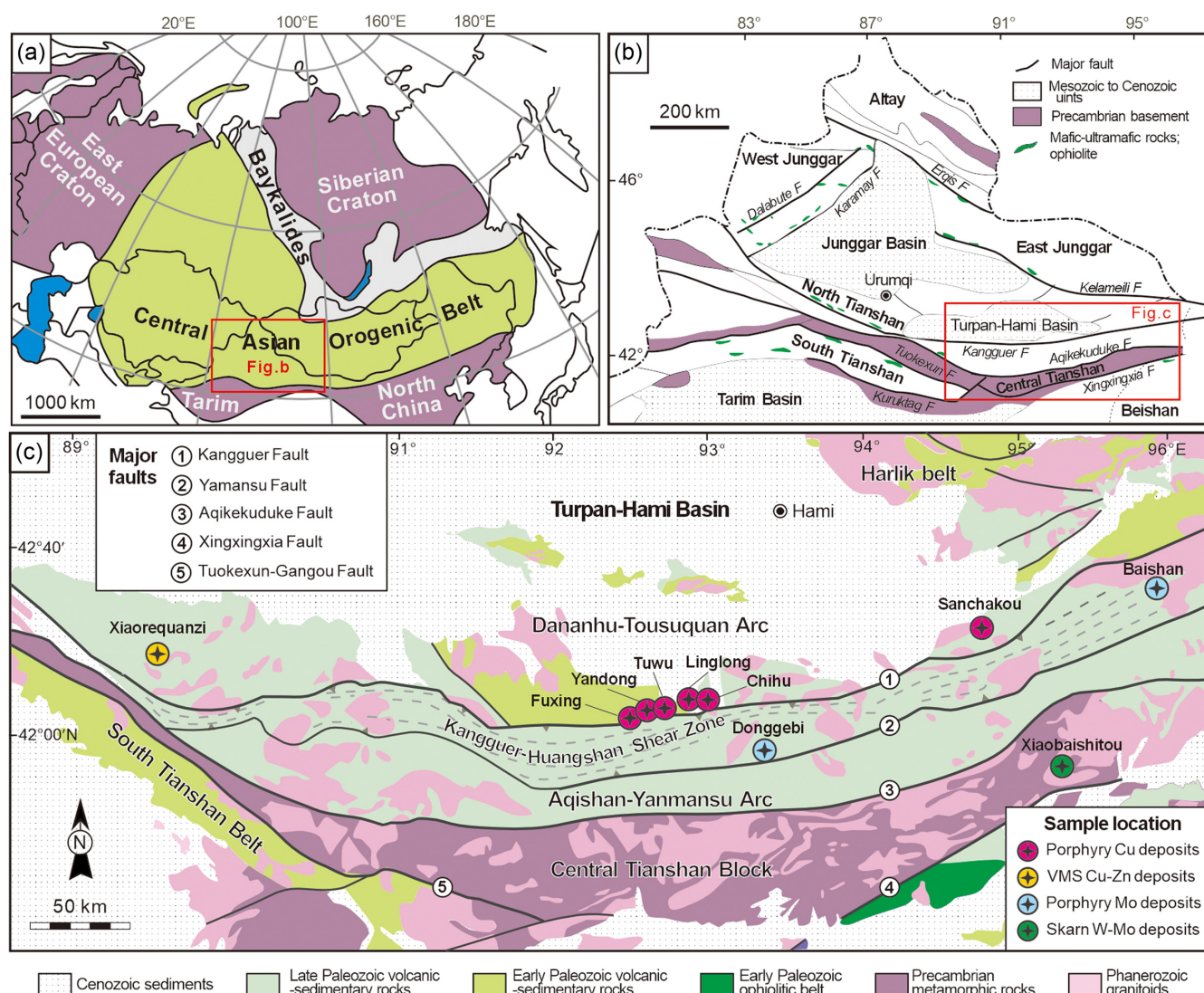


Figure 1 (a) Schematic map illustrating the precise location of the CAOBS and the adjacent cratons (after Şengör *et al.*, 1993). (b) Geological map showing the emplacement of tectonic sections in the southern area of the CAOBS (after Chen *et al.*, 2012). (c) Geological map exhibiting the arrangement of ore deposits in the Eastern Tianshan (after Wang *et al.*, 2006; Deng *et al.*, 2016).

Table 1 Description of the ore deposits studied in the Eastern Tianshan.

Location	Longitude (E°)	Latitude (N°)	Lithology	Tonnage and grade	Deposit type	Age of the granitoids (Ma)		Mineralisation age (Ma)
						Zircon U-Pb SHRIMP or SIMS	Zircon U-Pb LA-ICP-MS	
Xiaorequanzi	89.53	42.29	Tuff	a combined measured reserve of 0.176 Mt Cu, 0.195 Mt Zn	volcanogenic massive sulfide Cu–Zn deposit	Tuff: 352 ± 5		354 ± 11
Sanchakou	94.76	42.39	Quartz diorite	0.5 Mt Cu with an average grade exceeding 0.5 % Cu	porphyry Cu deposit	Quartz diorite: 436 ± 11	Quartz diorite: 440 ± 3	345 ± 5
Tuwu	92.61	42.13	Plagiogranite porphyry	145 Mt Cu with a grade of 0.62 % Cu	porphyry Cu deposit	Plagiogranite porphyry: 334 ± 3	Plagiogranite porphyry: 356 ± 8	335 ± 2
Fuxing	92.33	42.08	Plagiogranite porphyry	Prospect	porphyry Cu deposit	Plagiogranite porphyry: 332 ± 2		
Yandong	92.53	42.1	Plagiogranite porphyry and quartz albite porphyry	372 Mt @ 0.58 % Cu	porphyry Cu deposit	Plagiogranite porphyry: 332 ± 2	Plagiogranite porphyry: 339 ± 2 Quartz albite porphyry: 325 ± 2	325 ± 2
Chihu	93.03	42.17	Plagiogranite porphyry	Prospect	porphyry Cu deposit	Plagiogranite porphyry: 322 ± 10		316 ± 2
Xiaobaishitou	95.17	41.53	Biotite granite	36264 t W at an average grade of 0.78 % WO ₃	skarn-type W deposit	-	Biotite granite: 242 ± 2	245 ± 4
Donggebi	93.38	41.93	Porphyritic granite and granite porphyry	0.508 Mt Mo with an average grade of 0.115 % Mo	porphyry Mo deposit	Porphyritic granite: 236 ± 2	Porphyritic granite: 235 ± 3	232 ± 6
Baishan	95.55	42.31	Granite porphyry	0.72 Mt Mo with an average grade of 0.06 % Mo	porphyry Mo deposit	Granite porphyry: 228 ± 2	Granite porphyry: 230 ± 1	227 ± 3

*Information for porphyry Cu deposits are from He *et al.* (2023) and references therein, for porphyry Mo deposits from Wu *et al.* (2017) and references therein, for the skarn-type W deposit from Deng *et al.* (2017), and for the volcanogenic massive sulfide Cu–Zn deposit from He *et al.* (2020) and Mao *et al.* (2020).

Analyses of Hg concentration and isotopic composition of the samples were done at the Institute of Geochemistry, Chinese Academy of Sciences. Comprehensive analytical methodologies are provided in [Supplementary Information Text S-1](#). Hg-MDF is denoted in $\delta^{202}\text{Hg}$ in parts per thousand (‰), referring to the NIST-3133 Hg standard, which is examined prior and subsequent to each sample:

$$\delta^{202}\text{Hg} (\text{‰}) = \left[\left(\frac{{}^{202}\text{Hg}}{{}^{198}\text{Hg}}_{\text{sample}} \right) / \left(\frac{{}^{202}\text{Hg}}{{}^{198}\text{Hg}}_{\text{standard}} \right) - 1 \right] \times 1000 \quad \text{Eq. 1}$$

Hg-MIF is expressed as Δ , expressing the discrepancies between the observed and anticipated $\delta^{\text{xxx}}\text{Hg}$ values, where xxx = 199, 200 or 201, measured in per mil (‰):

$$\Delta^{\text{xxx}}\text{Hg} = \delta^{\text{xxx}}\text{Hg} - \delta^{202}\text{Hg} \times \beta \quad \text{Eq. 2}$$

β is 0.252 for ^{199}Hg , 0.5024 for ^{200}Hg and 0.752 for ^{201}Hg (Blum and Bergquist, 2007).

Results

The results of all samples are presented in [Supplementary Information Table S-2](#). The Hg concentration varies largely among porphyry Cu deposits (granite porphyry: 4.90 to 27.9 ng/g; sulfides: 0.85 to 7860 ng/g), porphyry Mo deposits (porphyritic granite: 1.20 to 27.6 ng/g; sulfides: 38.3 to 143 ng/g), volcanogenic massive sulfide Cu–Zn deposit (sulfides: 315 to 8990 ng/g), and skarn-type W deposit (biotite granite: 4.30 to 33.9 ng/g; sulfides: 199 to 359 ng/g). The $\delta^{202}\text{Hg}$ values vary significantly among all samples (−3.14 to +1.29 ‰), although there is no systematic difference among the different deposit types (porphyry Cu deposits: -0.56 ± 0.96 ‰; porphyry Mo deposits: -0.78 ± 0.87 ‰; volcanogenic massive sulfide Cu–Zn deposit: -1.25 ± 1.18 ‰; skarn-type W deposit: -0.70 ± 1.03 ‰; average $\pm 1\sigma$). A positive association variation trend exists between $\Delta^{201}\text{Hg}$ and $\Delta^{199}\text{Hg}$ values (Fig. 2a), aligning with the patterns observed in the land–ocean–atmosphere systems (Blum et al., 2014). Based on $\Delta^{199}\text{Hg}$ values (−0.11 to +0.51 ‰), the samples can be categorised into two groups (Fig. 2b). The first group comprises porphyry Cu deposits and the volcanogenic massive sulfide Cu–Zn deposit, which exhibit positive $\Delta^{199}\text{Hg}$ values ($+0.15 \pm 0.15$ ‰ and $+0.16 \pm 0.13$ ‰, respectively) that are within the range of marine sediments. The second group

comprises porphyry Mo deposits and the skarn-type W deposit, which exhibit near zero $\Delta^{199}\text{Hg}$ values (-0.01 ± 0.07 ‰ and -0.02 ± 0.08 ‰, respectively) that are intermediate to that of marine and terrestrial sediments.

Discussion

Recycling of marine sediments and seawater contributed subduction-related hydrothermal systems. Because MIF of Hg isotopes is mainly related to photochemical processes and does not occur during hydrothermal processes (Yin et al., 2024), we utilise $\Delta^{199}\text{Hg}$ to analyse the origin of Hg and, by extension, other ore-forming metals in the investigated deposits. Sulfide samples from the studied porphyry Cu deposits and volcanogenic massive sulfide Cu–Zn deposit exhibit positive $\Delta^{199}\text{Hg}$ values (Fig. 2a, b; $\Delta^{199}\text{Hg}$: $+0.16 \text{ ‰} \pm 0.14 \text{ ‰}$) that are similar to those of marine reservoirs (Fig. 2a; Blum et al., 2014). The positive $\Delta^{199}\text{Hg}$ values of porphyry Cu deposits suggest that Hg originated from recycled marine materials. This is consistent with double-sided subduction of the NTO slab below the Dananhu-Tousuquan Arc and Aqishan-Yamansu Arc between 355 and 320 Ma (Fig. 3a; Jahn, 2004; Wang et al., 2015). Analogous to this interpretation, volcanic arc-associated epithermal Au deposits formed in the circum-Pacific zone display mainly positive $\Delta^{199}\text{Hg}$ values (−0.02 to +0.27 ‰) due to the recycling of marine Hg from the subducted oceanic slab (Deng et al., 2021). The exsolution of fluids from descending oceanic plates has been shown to play a key role in generating porphyry Cu and epithermal Au deposits in volcanic arcs (Richards, 2003). The positive $\Delta^{199}\text{Hg}$ values of marine sediments are likely to be inherited by these fluids released via slab dehydration (Deng et al., 2021). The positive $\Delta^{199}\text{Hg}$ values of volcanogenic massive sulfide deposits can be explained by seawater circulation in an extensional tectonic regime induced by asthenospheric heat, since seawater contains Hg with positive $\Delta^{199}\text{Hg}$ values (Zhu et al., 2020).

Recycling of terrestrial sediments contributed to collision-related hydrothermal systems. The near zero $\Delta^{199}\text{Hg}$ values in porphyry Mo deposits (-0.01 ± 0.07 ‰) and skarn-type W deposits (-0.02 ± 0.08 ‰) are intermediate to those of marine and terrestrial sediments (Fig. 2a). Terrestrial sediments are mainly characterised by negative $\Delta^{199}\text{Hg}$ values, whereas marine sediments mainly display positive $\Delta^{199}\text{Hg}$ values. The overall

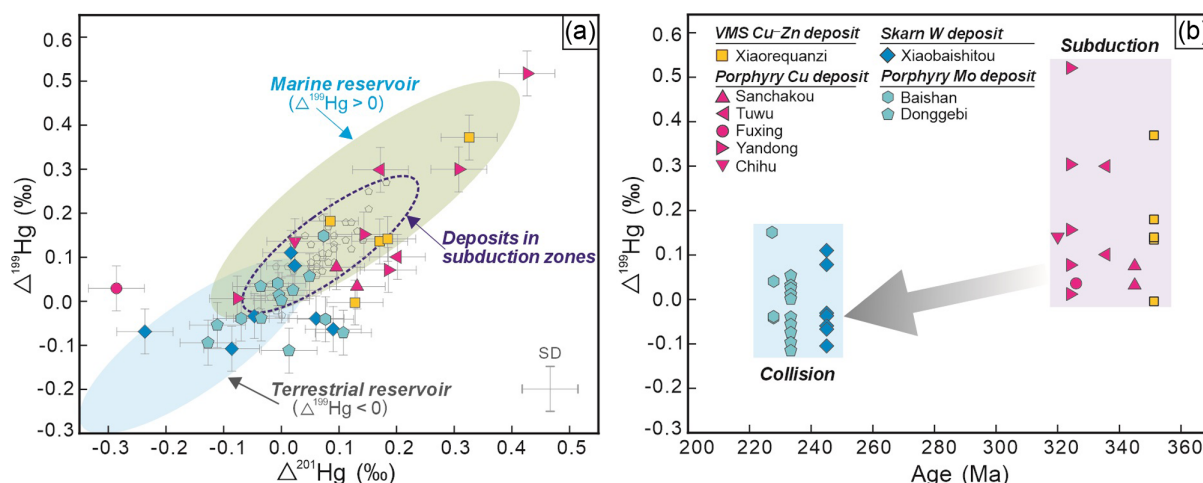


Figure 2 Binary diagrams displaying the variations in (a) $\Delta^{199}\text{Hg}$ – $\Delta^{201}\text{Hg}$ and (b) $\Delta^{199}\text{Hg}$ –Age (Ma) of the analysed ore deposits in the Eastern Tianshan. The area of marine sediments is delineated by Yin et al. (2015) and Meng et al. (2019), terrestrial sediments are characterised by Blum et al. (2014) and references therein, and subduction-related deposits are detailed by Deng et al. (2021).

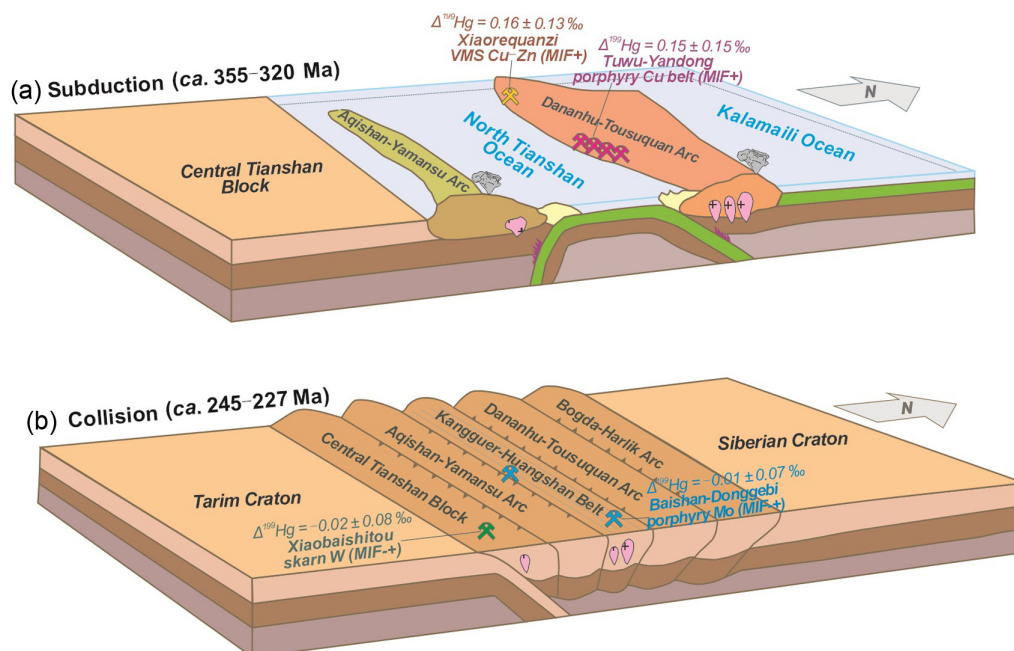


Figure 3 Schematic diagram illustrating the tectonic evolution of the Eastern Tianshan. (a) In the Carboniferous, the double-sided subduction of the NTO plate resulted in the formation of the Dananhu-Tousuquan Arc and Aqishan-Yamansu Arc, during which porphyry Cu deposits and volcanogenic massive sulfide Cu–Zn deposits were generated. (b) During the Triassic, the collision between the Tarim Craton and the Siberian Craton led to the formation of porphyry Mo deposits and skarn-type W deposits.

near zero $\Delta^{199}\text{Hg}$ values of the porphyry Mo deposits and skarn-type W deposit, with a few positive outliers, suggest contributions of Hg from both recycled marine sediments and terrestrial sediments during their formation. This interpretation is consistent with the relatively high $^{87}\text{Sr}/^{86}\text{Sr}_i$ values (0.7060 to 0.7088) of ore-bearing rocks coeval with porphyry Mo deposits and skarn-type W deposits in the Eastern Tianshan, which imply that oceanic subduction fertilised the mantle lithosphere (Deng *et al.*, 2017; Wu *et al.*, 2017). Continental subduction during the collisional stage would not only have released hydrothermal fluids with negative $\Delta^{199}\text{Hg}$ values, but also have caused partial melting and devolatilisation of metasomatised lithospheric mantle, releasing hydrothermal fluids with positive $\Delta^{199}\text{Hg}$ values (Deng *et al.*, 2022). Mercury in these two types of fluids tends to mix during upwards transport, and precipitate in tectonic weak zones. The negative $\Delta^{199}\text{Hg}$ values observed in the biotite granite of porphyry Mo deposits and skarn-type W deposits can be attributed to the closure of the NTO around ca. 245 Ma, and support the presence of continental arc magmatism in a collisional setting (Fig. 3b; Wu *et al.*, 2017).

Implications for the metallogenic effects of Wilson Cycles.

The CAOBS originated from long lived subduction, development and closure of the Paleo-Asian Ocean, during which the accretion of juvenile crust played a significant role in Phanerozoic continental growth (e.g., Jahn, 2004; Xiao *et al.*, 2004, 2018). The observation of positive $\Delta^{199}\text{Hg}$ values in oceanic subduction-related hydrothermal systems (porphyry Cu deposits, volcanogenic massive sulfide Cu–Zn deposits), and near zero $\Delta^{199}\text{Hg}$ values in continental collision-related hydrothermal systems (porphyry Mo deposits, skarn-type W deposits) suggest that Hg from both continental and oceanic crusts can be remobilised by magmatic–hydrothermal fluids and concentrated to form Hg-bearing hydrothermal deposits at convergent plate margins. As shown in Figure 3, mobilisation of Hg via dehydration of subducted oceanic and continental slabs not only releases Hg-bearing hydrothermal fluids, but also transfers substantial quantities of Hg into the overlying mantle wedge, resulting in

non-zero $\Delta^{199}\text{Hg}$ in the metasomatised lithospheric mantle. Partial melting and devolatilisation of the metasomatised lithospheric mantle, triggered by plate subduction, can also contribute metals for the generation of hydrothermal deposits in accretionary orogens.

Conclusions

Based on the newly generated Hg isotope dataset for magmatic–hydrothermal deposits in the Eastern Tianshan, this work demonstrates that oceanic subduction and continental collision can drive large scale recycling of Hg from oceanic and continental reservoirs into various hydrothermal systems within long lived accretionary orogens. In oceanic subduction-related porphyry Cu and volcanogenic massive sulfide Cu–Zn hydrothermal systems, the consistently positive $\Delta^{199}\text{Hg}$ values in sulfides indicate the dominant contribution of Hg from subducted marine sediments and circulated seawater, respectively. In continental collision-related hydrothermal systems (porphyry Mo deposits and skarn-type W deposits), the near zero $\Delta^{199}\text{Hg}$ values in sulfides suggest the contribution of Hg from subducted terrestrial sediments and the oceanic subduction fertilised mantle lithosphere. Overall, this work provides key knowledge into crust–mantle cycling of Hg and the metallogenic effects of oceanic subduction and continental collision during Wilson Cycles.

Acknowledgements

The research was supported by the Natural Science Foundation of China (42222205, 42430804, 41872084), National Key Research and Development Program of China (2018YFC0604006), Xinjiang Key Research and Development Project (2023B03015), and the Third Xinjiang Scientific Expedition Program (2022xjkk1301).

Editor: Horst Marschall

Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2516>.



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Cite this letter as: He, X., Deng, X., Brzozowski, M.J., Wu, Y., Zhang, J., Wu, C., Chen, Y., Yin, R. (2025) Hg isotopes constrain metallogenic effects of the Wilson Cycle of the Paleo-Asian Ocean. *Geochem. Persp. Let.* 35, 7–12. <https://doi.org/10.7185/geochemlet.2516>

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