

## 1 REGIONAL GEOLOGICAL BACKGROUND

2 NE China is situated between the Siberia and North China cratons ([Fig. 1c](#)), at the  
3 intersection of the Paleozoic Central Asian Orogenic Belt and the Mesozoic/Cenozoic  
4 Pacific orogenic belt. It is characterized by multistage subduction of Paleo-oceanic  
5 plates (e.g., the Paleo-Asian, Mongol-Okhotsk, and Paleo-Pacific plates), and  
6 amalgamation of several microblocks during the Phanerozoic. Large-scale  
7 tectonism-magmatism-metallogenesis and a series of NE trending transcrustal faults  
8 developed on a Precambrian crystalline basement, overlain by Paleozoic low-grade  
9 metamorphic marine strata, and Mesozoic-Cenozoic terrestrial sedimentary rocks.  
10 Phanerozoic igneous rocks are widely distributed in NE China ([Fig. 1d](#); [Wu et al.,](#)  
11 [2011](#); [Xu et al., 2013](#)). Especially during the late Mesozoic, large scale (sub)volcanic  
12 rocks developed in an extensional setting in eastern NE China due to the roll-back of  
13 the Paleo-Pacific plate ([Xu et al., 2013](#)), triggering the formation of typical  
14 subduction-related hydrothermal Au deposits (e.g., Sandaowanzi, Tuanjieyou,  
15 Jinchang, and Sipingshan; [Fig. 1b and d](#)).

## 16 SAMPLES AND ANALYTICAL METHODS

17 A total of 48 samples, including six from the Jinchang, four from the Sipingshan,  
18 twelve from the Tuanjieyou, six from the Fuqiang, five from the Sandaowanzi, seven  
19 from the Yongxin, and eight from the Pangkaimen Au deposits, were collected from  
20 mines, pits, and trenches. The locations of these deposits are shown in [Fig. 1d](#).

21 The collected samples include 7 volcanic rock samples from wall rocks and 41  
22 ores. Collected wall rocks are calc-alkaline volcanic rocks, including rhyolite,  
23 andesite, and tuff. These rocks are relatively fresh without significant influences of  
24 hydrothermal alteration, such as pyritization, sericitization, silicification, and  
25 carbonatization. Ores in these Au deposits consist mainly of hydrothermal breccias

26 and quartz veins, with minor silicified volcanic rocks, and disseminated  
27 mineralization types. Four auriferous breccia samples from the Yongxin, twelve  
28 breccia and quartz vein samples from the Tuanjiegou, seven hydrothermal breccias  
29 samples from the Pangkaimen, four pyritization chert samples from the Sipingshan,  
30 five sulfide-bearing quartz vein samples from the Sandaowanzi, five hydrothermal  
31 breccias samples from the Fuqiang, and four quartz-sulfide vein and breccias samples  
32 from the Jinchang Au deposits were collected for Hg isotope analysis. Considering  
33 that pyrite is the main sulfide in these deposits, and is the main Hg-bearing mineral  
34 (occurring as isomorphic substitution), seven ore samples from the Tuanjiegou,  
35 Pangkaimen, Sipingshan, and Sandaowenzi Au deposits were selected and prepared  
36 for pyrite concentration. Pyrites were separated by handpicking under a binocular  
37 microscope after being crushed to within 50-100  $\mu\text{m}$  for magnetic selection. Before  
38 chemical analysis, the samples were powdered to 200 mesh size and homogenized in  
39 an agate mortar.

40 Total Hg (THg) concentrations of the samples were measured using a direct  
41 combustion system (Nippon MA-2) followed by atomic absorption spectroscopy  
42 analysis. Measurements of standard reference material (GSS-4, soil) showed  
43 recoveries of 90-110%, and coefficients of variation for triplicate analyses were <  
44 10%.

45 The samples were prepared for mercury isotope analysis following the  
46 double-stage thermal combustion and pre-concentration protocol (see the detailed  
47 method in [Zerkle et al., 2020](#)). Standard reference material (GSS-4, soil) was prepared  
48 in the same way as the samples. The prepared samples were diluted to 1 ng/mL Hg  
49 and measured by a Neptune Plus multi-collector inductively coupled plasma mass  
50 spectrometer (MC-ICP-MS), following the method by [Yin et al. \(2016\)](#). Hg

concentrations and acid matrices in the bracketing NIST-3133 solutions were matched well with the neighboring samples. Mercury isotopic compositions were reported following the convention recommended by [Blum and Bergquist \(2007\)](#). Specifically, MDF is expressed in  $\delta^{202}\text{Hg}$  notation in units of ‰ referenced to the NIST-3133 Hg standard (analyzed before and after each sample):

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

MIF is reported in  $\Delta$  notation, which describes the difference between the measured  $\delta^{\text{xxx}}\text{Hg}$  and the theoretically predicted  $\delta^{\text{xxx}}\text{Hg}$  value, in units of ‰:

$$\Delta^{\text{xxx}}\text{Hg} \approx \delta^{\text{xxx}}\text{Hg} - \delta^{202}\text{Hg} \times \beta$$

$\beta$  is equal to 0.2520 for  ${}^{199}\text{Hg}$ , 0.5024 for  ${}^{200}\text{Hg}$ , and 0.7520 for  ${}^{201}\text{Hg}$ . Analytical uncertainty was estimated based on the replication of the NIST-3177 standard solution. The overall average and uncertainty of NIST-3177 ( $\delta^{202}\text{Hg}$ :  $-0.5 \pm 0.1$ ;  $\Delta^{199}\text{Hg}$ :  $-0.01 \pm 0.05\text{‰}$ ;  $\Delta^{201}\text{Hg}$ :  $-0.02 \pm 0.07\text{‰}$ ; 2SD, n=11) and GSS-4 ( $\delta^{202}\text{Hg}$ :  $-1.67 \pm 0.14\text{‰}$ ;  $\Delta^{199}\text{Hg}$ :  $-0.34 \pm 0.08\text{‰}$ ;  $\Delta^{201}\text{Hg}$ :  $-0.35 \pm 0.07\text{‰}$ ; 2SD, n=3) agree well with comparable previous studies ([Blum and Bergquist, 2007](#); [Sun et al., 2019](#)). The larger values of standard deviation (2SD) for either NIST-3177 or -4 are used to reflect analytical uncertainties.

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