

# **Metasomatized lithospheric mantle for Mesozoic giant gold deposits in the North China Craton**

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## 1. Analytical methods

Visibly altered surfaces of basalts were removed with a rock saw. Cut surfaces were abraded with silica emery paper and washed with 18.2Ω Milli-Q (Nanopure) water, and subsequently dried overnight. Rock pieces of about 100 grams were crushed into small chips and processed to fine powder using an agate disc mill. This procedure avoids possible metal contamination from sample processing.

We modified previous analytical methods for gold and PGE contents (Fischer-Gödde et al., 2011) and have established in Wuhan for different types of geological rocks (Cheng et al., 2019). The bulk rock gold and PGE contents of xenoliths and basalts were determined after Carious tube digestion in reverse aqua regia and chromatography separation: PGE by isotope dilution methods and gold by two independent methods of standard addition and internal standardization of gold to platinum (Cheng et al., 2019). After addition of a suitable amount of mixed  $^{191}\text{Ir}$ - $^{99}\text{Ru}$ - $^{194}\text{Pt}$ - $^{105}\text{Pd}$  and  $^{185}\text{Re}$  spike solutions, about 2 grams of powder of each sample was digested by reverse aqua regia (5 ml of 14 M  $\text{HNO}_3$  and 2.5 ml of 9 M  $\text{HCl}$ ) in pre-cleaned Carius tubes at 240 °C for 3-4 days. The digested sample solutions were transferred to centrifuge tubes, and 6 M  $\text{HCl}$  was used to rinse the Carius tube twice, and then combined with the sample solution. After centrifugation, the supernatant solution was completely transferred to weighed Teflon beakers and 6 M  $\text{HCl}$  was further used to rinse the centrifuge tubes and transferred to the Teflon beakers. These processes ensure complete recovery of Au and PGEs, which is important for quantitative determination of gold if the standard addition method is applied (Cheng et al., 2019).

About one quarter of each sample solution was weighed and converted to chloride for chemical separation. Chemical separation followed techniques described previously (Cheng et al., 2019; Fischer-Gödde et al., 2011), and gold, Ir, Ru, Pt, Pd and Re were separated from the matrix by cation exchange chromatography using 10 ml of pre-cleaned Eichrom 50W-X8 (100-200 mesh) resins. The samples were dissolved in 10 ml of a mixture of 0.5 M  $\text{HCl}$  - 40% (V/V) acetone and loaded on the cation resin. 20 ml 0.5 M  $\text{HCl}$  - 40% (V/V) acetone was

collected for complete recovery (> 99%) of Au and PGEs. Afterwards, the eluent was dried down to 0.5-1 ml.

The chemical purification procedure removed most matrix, except for some Cr, which can be abundant (hundreds of ppm or more) in mantle xenoliths and basalts. To avoid the potential effect of remaining matrix, two independent methods were used to determine the Au contents: 1) internal standardization of gold to platinum, which is precisely determined by isotope dilution and 2) standard addition which eliminates the effect of remaining matrix. For the standard addition method, the sample solution was quantitatively separated into three fractions and 1.25 M HCl and/or the gold standard solution with known concentration was added (Cheng et al., 2019).

The samples in 1.25 M HCl solutions were measured by high sensitivity sector-field inductively coupled plasma mass spectrometry (Thermo Scientific® Element II) at the State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences, Wuhan. The detection limit of instrument was about 0.8 ppt for Au. The monitored intensity of  $^{181}\text{Ta}$  was low and the potential oxide interference of  $^{181}\text{TaO}$  on  $^{197}\text{Au}$  (monitored  $^{181}\text{Ta}/^{197}\text{Au} < 0.001$ ) was negligible in the routine measurement conditions with Ta/TaO ratio of 0.002. The total procedural blanks were very low for gold ( $\approx 5 \pm 5$  pg, 2sd,  $n=20$ ), which resulted in low detection limits of our analytical methods. The abundances of Ir, Pt and Re by isotope dilution were obtained with Au from the same aliquots. The sample solution was further purified by anion resin for Ru and Pd (Cheng et al., 2019).

## **2. Data quality of gold abundances and sample heterogeneity**

Accurate analysis of geological samples with low gold concentrations is very challenging. Application of aqua regia and Carius tubes achieved low total procedural blanks (Cheng et al., 2019; Fischer-Gödde et al., 2011). In our methods, the total procedural blanks were only  $5 \pm 5$  (2sd) pg for gold and resulted in low detection limits (Figure DR1). The data quality of gold abundances was extensively evaluated in this study.

Gold concentrations were obtained by two independent methods: internal standardization and standard addition. The values from the two methods are consistent within

a few percent (Table DR1-2; Figure DR2a). Geological reference materials with variable compositions and their replicates such as TDB-1, BHVO-2 and GPt-2 have been analyzed and the results showed the high precision and accuracy of the analytical methods (5-15%, 2sd) (Cheng et al., 2019). For example, the replicates of TDB-1 by our method displayed a mean value of  $6.1 \pm 0.7$  ppb (2sd, n=20), similar to the certified  $6.3 \pm 1.0$  ppb (2sd) and those analyzed by nickel sulfide-fire assay ( $6.2 \pm 0.6$  ppb, 2sd, n=20) (Richardson and Burnham, 2002) and HF-acid digestion ( $6.3 \pm 1.7$  ppb, 2sd, n=21) (Pitcairn et al., 2015). UB-N was determined using internal standardization methods, and the gold content ranged from 1.0-1.7 ppb with a mean value of  $1.49 \pm 0.52$  ppb (2sd, n=11) (Fischer-Gödde et al., 2011). Our results were 1.1-1.4 ppb with a mean of  $1.2 \pm 0.16$  ppb (2sd, n=4). The gold in BHVO-2 is 1.1-1.4 ppb with a mean of  $1.3 \pm 0.3$  ppb (2sd, n=7). Overall, the typical analytical uncertainty for gold contents is about 5-10 % (2sd) but can be larger (20-50%) for samples with very low gold contents of < 0.1 ppb (Table DR1-2; Figure DR2b).

Sample heterogeneity is often thought as a critical issue for Au and PGE contents in ultramafic and mafic rocks. Thus, we have measured replicates of many peridotites and basalts. For five basalts and three mantle xenoliths, the aqua regia solutions from the same digestion were repeatedly purified via column separation and measured, and the results show identical results, indicating excellent reproducibility of the gold data (Table DR1-2; Figure DR3a). Replicates of eleven basalts and three mantle xenoliths were measured and they show consistent results within uncertainty, indicating the negligible effect of sample heterogeneity (Figure DR3b). PGE data have been available for most mantle xenoliths (Chu et al., 2009; Liu et al., 2011) and our new PGE data are mostly similar to previous values, except for a few samples with very low PGE contents, indicating the comparability of the PGE and Au data between previous work and this study (Table DR1-3).

### **3. Supplementary notes**

#### **3.1. Gold contents in mantle xenoliths of the NCC**

Mantle peridotites and pyroxenites, as well as experimental data (Brenan et al., 2016; Mungall and Brenan, 2014), have constrained the magmatic behavior of PGEs, Au, Re, Cu

and S during partial melting, melt transport and refertilization, with sulfide melt-silicate melt partition coefficients decreasing in the order: PGE > Au > Cu > S  $\approx$  Re (e.g., Fischer-Gödde et al., 2011; Lorand et al., 2008; Wang and Becker, 2015a, b; Wang et al., 2013). High-degree melting would lead to high contents of Os-Ir-Ru (compatible) in refractory peridotites but low Pd, Au, Cu and S contents with an increasing depletion in the order of Pd < Au < Cu < S (variably incompatible), e.g., low Pd/Ir and Au/Pd (e.g., Becker et al., 2006; Fischer-Gödde et al., 2011; Lorand et al., 2013; Luguet et al., 2007). Subsequent melt refertilization with sulfide precipitation often enriches the refractory peridotites in incompatible Pd, Cu, Au and S, leading to the elevation of Pd/Ir and Au/Pd (Fischer-Gödde et al., 2011; Lorand et al., 2008; Lorand et al., 2013; Maier et al., 2012). A higher extent of enrichment of Au relative to PGE is expected if strong melt/fluid metasomatism is involved, as reflected by peridotite xenoliths from the Kaapvaal craton (e.g., Finsch and Venetia samples with high S contents of 280-1240 ppm and high Au/Pd<sub>N</sub> of > 1 to 13, Maier et al., 2012).

The SCLM under the NCC metasomatized mainly by subduction materials is often assumed to be the main gold source for the 130-120 Ma giant gold deposits (e.g., Goldfarb and Groves, 2015; Li et al., 2012; Tan et al., 2018; Zhu et al., 2015). Gold contents in the SCLM are of fundamental importance for understanding the formation of Mesozoic gold deposits and could be directly represented by mantle xenoliths hosted in kimberlites and basalts. The gold and PGE contents of many peridotite xenoliths hosted in Paleozoic kimberlites (Mengyin and Fuxian) (Li et al., 2011; Zhang et al., 2008), Mesozoic basalts (Xinyang) (Zheng et al., 2005) and Cenozoic basalts, e.g., Hebi, Shanwang (Zheng et al., 2005), Wangqing (Orberger et al., 1998) and Hannuoba (Chu et al., 1999; Fischer-Gödde et al., 2011) have been reported before. These data show a large range from 0.5-38.3 ppb Au with a median value of 3.5 ppb (Figure DR4 and DR5), which is clearly higher than the median 1.2 ppb of mantle peridotites worldwide (n=508, typical range of < 0.6 to 2 ppb) (Saunders et al., 2018).

Mantle xenoliths from Hebi (Liu et al., 2011; Sun et al., 2012; Zheng et al., 2005; Zheng et al., 2007), Mengyin and Fuxian (Zhang et al., 2008) are mostly highly refractory harzburgites with low Al<sub>2</sub>O<sub>3</sub> contents and high Mg# and show Neoproterozoic to

Paleoproterozoic Os model ages. These features reflect that they are the relics of ancient lithospheric mantle after high-degree melt extraction. However, these harzburgites display strong enrichment of light REE (Zheng et al., 2005), radiogenic Sr-Nd isotopes and high S contents, suggesting that they have experienced subsequent extensive metasomatism with sulfide precipitation. Previous data by NiS fire assay-ICPMS show high gold contents and high Au/Pd<sub>(N)</sub> in these refractory xenoliths (Zhang et al., 2008; Zheng et al., 2005). Mineral separates of olivine and chromite from Mengyin xenoliths display surprisingly high gold contents up to 6.2-16.9 ppb and high Au/Pd<sub>(N)</sub> of 16-21 (Zhang et al., 2008). The Mesozoic basalt-hosted Xinyang peridotite xenoliths (Zheng et al., 2005) and Cenozoic basalt-hosted Shangwang (Zheng et al., 2005) and Wangqing xenoliths (Orberger et al., 1998) similarly show high and variable gold contents (Figure DR5). These results could be interpreted as evidence for an inherently gold-rich SCLM beneath the NCC from Archean to present (Griffin et al., 2013), distinct from other mantle domains worldwide, including those with strong mantle metasomatism, like Kaapvaal mantle xenoliths and other locations (Fischer-Gödde et al., 2011; Maier et al., 2012; Saunders et al., 2018).

However, the Cenozoic Hannuoba basalt-hosted peridotites, which contain many sulfides (Gao et al., 2002) show low gold contents of mostly 1-2 ppb (Chu et al., 1999; Fischer-Gödde et al., 2011). Considering the highly variable gold contents in previous data and their great importance, the well-characterized mantle xenoliths from Hebi, Mengyin and Shanwang with published S, Cu and PGE contents (Chu et al., 2009; Liu et al., 2011) were analyzed for gold in this study. The selected Mengyin peridotite and pyroxenite xenoliths show strong enrichment of light REE and high S contents (Chu et al., 2009; Li et al., 2011), which are the result of metasomatism between the Archean and 480 Ma. However, our new data show only 0.06-0.50 ppb Au, as well as low Pd, Cu and Re contents (Chu et al., 2009), suggesting limited recharge of Pd, Au and Cu by sulfur-bearing metasomatism. These gold contents are significantly lower than the previously reported 3-6 ppb in peridotite xenoliths from the same location. Similarly, Hebi harzburgites with ancient Os model ages and elevated La/Yb<sub>(N)</sub> also display low gold contents of 0.03-0.11 ppb (Figure 2). Note that in this study, the low gold contents in Mengyin and Hebi harzburgite xenoliths are coupled with low Pd, Cu

and Re contents in the same samples (Figure 2). The elevated Au/Pd in some samples suggests that metasomatism did replenish some chalcophile elements by sulfide precipitation, but the low Au contents indicate that the amount was limited.

As for peridotite xenoliths from other Cenozoic basalts in the eastern NCC, Shanwang xenoliths show Os isotopic compositions similar to abyssal peridotites (representing present-day convecting mantle) (Chu et al., 2009). Combined with Sr-Nd-Hf isotopes, they suggest that the lithosphere sampled by Shanwang peridotite xenoliths was derived from convecting upper mantle during the Mesozoic or Cenozoic, after the destruction of the NCC (Chu et al., 2009). These samples do not show enrichment of Pd and Re relative to compatible PGE such as Os, Ir and Ru (Chu et al., 2009). Our new results for gold contents are 0.02-1.78 ppb with a median value of 0.7 ppb (n=19), lower than previous values (3.4 to 6.7 ppb). Importantly, the new gold analyses are also consistent with Pt, Pd and Cu contents. The contents and patterns of PGE, Au, Cu and S are indistinguishable with fertile mantle (Figure 2, DR4).

Therefore, our high-precision new data from Hebi, Mengying, Shanwang mantle xenoliths, as well as the previous gold data from Hannuoba mantle xenoliths (reflecting the metasomatized ancient SCLM and juvenile SCLM under the NCC) indicate no substantial enrichment of Au relative to Pd and Cu contents in the same samples. They suggest that the NCC is not inherently rich in gold and that mantle metasomatism between the Archean and 480 Ma and replacement by juvenile lithospheric mantle did not lead to strong enrichment of gold in the SCLM.

### **3.2. Discrepancy between previous and new values on peridotites**

The PGEs contents in this study are generally consistent with previous values but gold displays obvious discrepancy. The gold and PGE contents in peridotite xenoliths from Hebi, Mengyin and Shangwang have been measured before and also by this study (Figure DR5). This gives us an opportunity to better compare the obtained data. As discussed above, sample heterogeneity is not the issue because the replicates show repeatable gold and PGEs contents (Figure DR3). Instead, the different data quality for gold contents may be the main reason. Previous gold data from North China Craton were highly variable and mostly higher than

other regions worldwide (mostly 3-16 ppb, Figure DR5). However, the peridotite xenoliths from Hannuoba show low gold contents of 1-2 ppb, which are more reasonable for fertile mantle rocks (Fischer-Gödde et al., 2011). The high values were mainly obtained by NiS fire assay pre-concentration method, but the latter was by aquia-regia solution with rather low blanks (a few pg). It is well known that the NiS fire assay method often shows variable levels of procedural blanks for Au (e.g., from <0.1 ng to several ng), sometimes similar or even higher than samples (e.g., Barefoot and Van Loon, 1999; Gros et al., 2002; Oguri et al., 1999). It implies that it is difficult to accurately measure samples with low gold contents sometimes. Given the inconsistent values in previous studies, we thus have set up the robust analytical methods and used a lot of international geological reference materials to test our data quality (Cheng et al., 2019). Our new methods led to very low gold blanks of a few pg, which makes it possible to measure samples with rather low gold contents. The gold contents in this study were also obtained by two independent methods for one sample, and they are consistent with a few percent (Figure DR2).

### **3.3. Gold contents in mantle-derived basalts**

The basalts in this study include high Mg# (71-75), hydrous, isotopically enriched 130-120 Ma basalts, and isotopically depleted < 110 Ma basalts (Figure DR6). The former are Yixian, Sihetun, Fangcheng and Feixian basalts (Gao et al., 2008; Huang et al., 2017; Liu et al., 2008). They are from the northern (Yixian, Sihetun) and southern (Feixian, Fangcheng) margins of the eastern NCC, where pre-existing Paleozoic and Triassic subduction occurred and probably affected the SCLM.

The 130-120 Ma basalts from SCLM of the North China Craton have been intensively studied in the past (Gao et al., 2008; Geng et al., 2019a; Geng et al., 2019b; Huang et al., 2017; Liu et al., 2008; Meng et al., 2015; Xia et al., 2013; Zhang et al., 2002). They are characterized by high water contents (Geng et al., 2019b; Xia et al., 2013), arc-like trace element patterns (Figure DR7) and radiogenic Sr-Nd-Hf-Os isotopes (Gao et al., 2008; Huang et al., 2017; Liu et al., 2008; Meng et al., 2015). These features have been interpreted to be the accumulated consequences of multiple stages of metasomatism of the SCLM (e.g., Meng



et al., 2015; Wu et al., 2019; Zhang et al., 2002). Although crustal contamination could lead to similar isotopic features, but it is not a viable explanation for 130-120 Ma basalts from the North China craton. The main lines of evidence are: 1) Mesozoic mantle-derived rocks (130-120 Ma), irrespective of basalts (Meng et al., 2015; Zhang et al., 2002), lamproites (Ma et al., 2016) and gabbros (Xu et al., 2004) all show similar Sr-Nd isotopes (Wu et al., 2019); 2) such Sr-Nd isotopes hardly change for different rocks with variable SiO<sub>2</sub> (e.g., Ma et al., 2014; Wan et al., 2019); 3) Ratios of trace elements such as Nb/U and Ce/Pb are similar to MORBs rather than mixture with crustal materials and also do not support the model of crustal contamination (e.g., Ma et al., 2016); 4) 130-120 Ma basalts contain high abundances of water and other volatiles (Geng et al., 2019a; Geng et al., 2019b; Xia et al., 2013) which cannot result from the high-grade metamorphic crust. All these results support that the 130-120 Ma were originated from metasomatized and hydrated mantle.

The primitive magmas were derived from melting of metasomatized and hydrated SCLM and erupted around 120-125 Ma, coeval with or slightly before the peak formation of giant gold deposits (130-120 Ma). The high PGE contents (this study) and radiogenic Os isotopes (Gao et al., 2008; Huang et al., 2017) suggest that these primitive basalts dominantly resulted from the metasomatized and fusible components of the metasomatized SCLM. Discrete gold grains may be present in veins of metasomatized peridotites (Tassara et al 2017), and such fusible rocks could be entered into the melts. Therefore, the 130-120Ma basalts are of great importance in constraining the degree of gold enrichment in the metasomatized and hydrated SCLM, and as an analogue for primitive mantle-derived, ore-forming magmas to understand the potential for the almost coeval giant gold deposits that display substantial inheritance of mantle volatiles (Mao et al., 2008; Tan et al., 2018).

The < 110 Ma basalts occur at Fuxin (erupted at 100 Ma), Shanwang (18 Ma) and Hebi (4 Ma). Based on their depleted Sr-Nd-Hf isotopes, they are interpreted to be derived from the Mesozoic and Cenozoic convecting asthenospheric mantle (Meng et al., 2015; Zhi et al., 1994; Zhu et al., 2012). The Fuxin basalt is from the northern margin of the eastern NCC and spatially close to the enriched Yixian and Sihetun basalts (Figure 1). The transition from the 130-120 Ma to < 110 Ma basalts in adjacent locations allows comparison of the amount of

gold released from the metasomatized SCLM and asthenospheric mantle, respectively. The Shanwang basalts occur near the trans-lithospheric Tanlu fault and the Hebi basalts from the middle part of the NCC where the extent of craton destruction was far less than the eastern NCC. During their eruption, Shanwang and Hebi basalts brought up the mantle xenoliths studied here.

The major and trace elements of the basalts from the same locations are generally similar, but the Au and PGEs contents sometimes display a large variation (Figure 2 and DR8). This variation seems not to result from sample heterogeneity as replicate analyses of basalts show consistent values (Figure DR3b). Instead, it reflects the heterogeneous distribution of gold and PGEs in their mantle sources, because Os isotopic compositions of the basalts from the same locations also show a large range (Gao et al., 2008; Huang et al., 2017).

Gold contents in the 130-120 Ma and < 110 Ma basalts display positive correlations with PGEs (Figure DR8). Overall, the 130-120 Ma basalts (except those from Fangcheng) show higher Au and PGE contents than the < 110 Ma ones. The low Au contents in the Fangcheng basalts could be attributed to sulfide segregation during magmatic petrogenesis (Sun et al., 2013), but more likely reflect the intrinsic depletion of the mantle source because other chalcophile elements such as Cu and PGEs are also low and Cu/Pd are constant (Figure DR8). The data of Fangcheng basalts thus was excluded to calculate the mean values of 130-120 Ma basalts. The Au/Pd<sub>(N)</sub> ratio in the Yixian and Sihetun basalts (erupted at 125 Ma) is slightly higher than that of the adjacent Fuxin basalts (erupted at 100 Ma, Figure DR9), likely suggesting slight enrichment of Au relative to PGEs in the metasomatized SCLM.

The average gold contents from the former are 2.2 ppb, a factor of 3-4 times higher than the 0.7 ppb of the latter, indicating a higher contribution of Au from the metasomatized SCLM, although the metasomatized SCLM was not rich in gold (< 1-2 ppb, Figure 4). The Shanwang basalts located near the trans-lithospheric Tanlu fault contain < 1 ppb Au. The SCLM near Hebi was probably not thinned or thinned to a limited extent (Liu et al., 2011) and the Hebi basalts display similarly low Au contents of < 1 ppb (Figure 2). These results

suggest a low Au concentration of the juvenile SCLM or lower extraction efficiency of Au from their mantle source than the metasomatized SCLM.

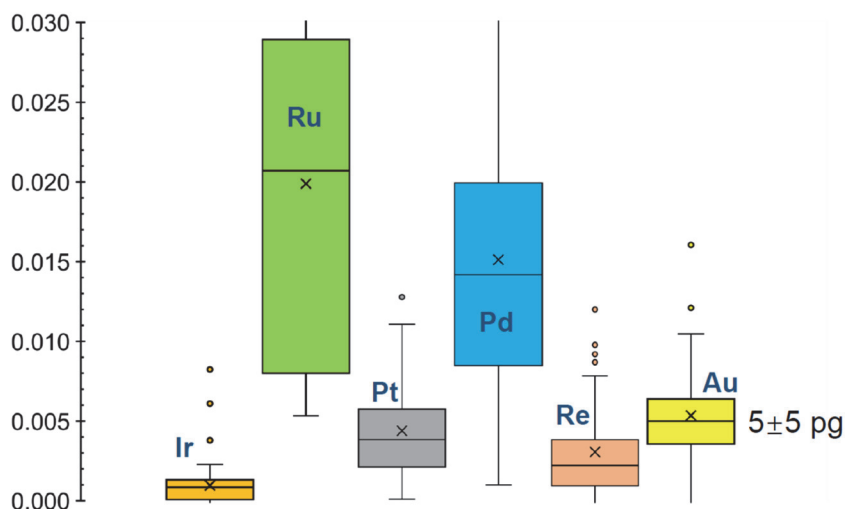
### **3.4. High contents of water and volatiles in the 130-120 Ma basalts**

The Feixian and Fangcheng basalts are from the southern margin of the eastern NCC, and Yixian and Sihetuan basalts from the northern margin (Figure 1). These 130-120 Ma basalts have been well studied, and were thought to be primitive melts of the metasomatized and hydrated SCLM (Gao et al., 2008; Huang et al., 2017; Liu et al., 2008; Meng et al., 2015; Sun et al., 2013; Xia et al., 2013; Zhang et al., 2002). Previous work has constrained these basalts to be hydrous and volatile-rich (e.g. water, S, C). For example, the Fangcheng basalts show many carbonate and sulfide inclusions and carbon-bearing melt inclusions (Sun et al., 2013). The primitive Feixian basalts contain a few percent water (Xia et al., 2013), suggesting > 1000 ppm water in the metasomatized SCLM source, far higher than the MORB source (50-200 ppm), the Kaapvaal cratonic mantle (120 ppm) (Xia et al., 2013) or the juvenile SCLM of the NCC (Xia et al., 2017). The 130-120 Ma basalts that are from the northern margin of the eastern NCC: the Yixian basalts are also rich in water and volatiles (Geng et al., 2019a; Geng et al., 2019b). Many melt inclusions in olivine phenocrysts display high volatile contents (Figure DR10). Mantle fluids and volatiles promoted gold extraction from mantle during melting and led to elevated gold contents in the hydrous mantle magmas (Botcharnikov et al., 2011; Pokrovski et al., 2013). Given the very high partition coefficients for Au between fluids and hydrous magmas (Pokrovski et al., 2013), fluid exsolution during later magmatic-hydrothermal stage would highly enrich gold in the S, C, Cl and noble gas-bearing fluids. They eventually evolved to auriferous fluids for hydrothermal lode gold deposits. These processes are consistent with the broad similarity in fluid/volatile compositions including C, Cl, and water between hydrous mantle magmas and ore fluids (Qiu et al., 2002; Zhu et al., 2015), and also with the inheritance of a mantle isotopic signature of volatiles and fluids (Mao et al., 2008; Tan et al., 2018).

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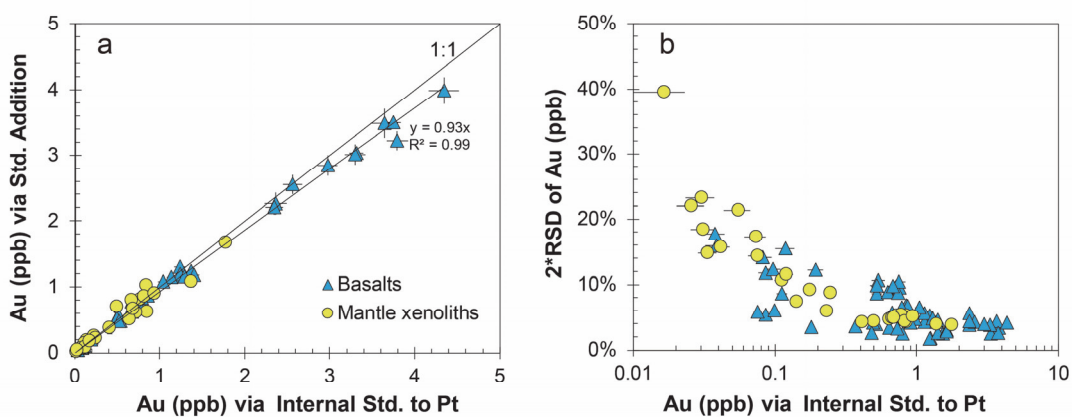
#### 309 4. Figures DR

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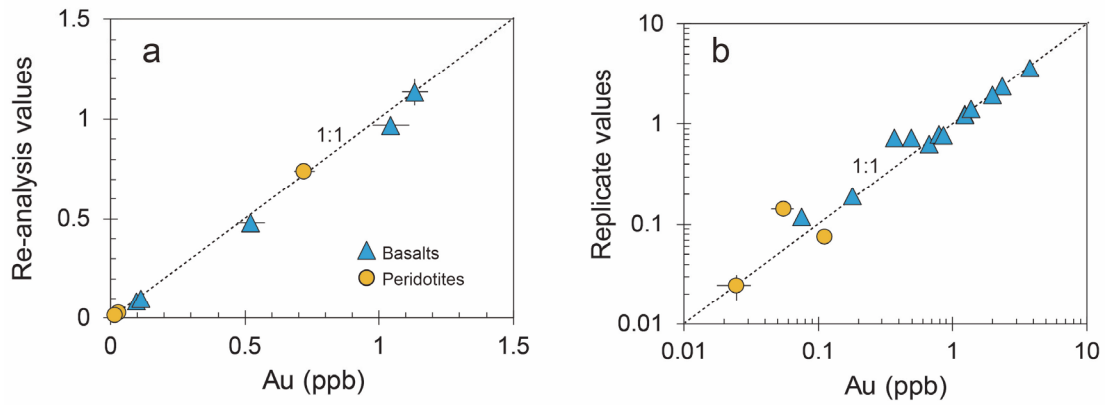


311 **Figure DR1.** Total procedural blanks of Au and PGEs in this study (ng), which are negligible  
 312 for most samples except samples with low values (e.g., < 0.1 ppb Au). The mean (horizontal  
 313 line) and median (cross) values are shown.

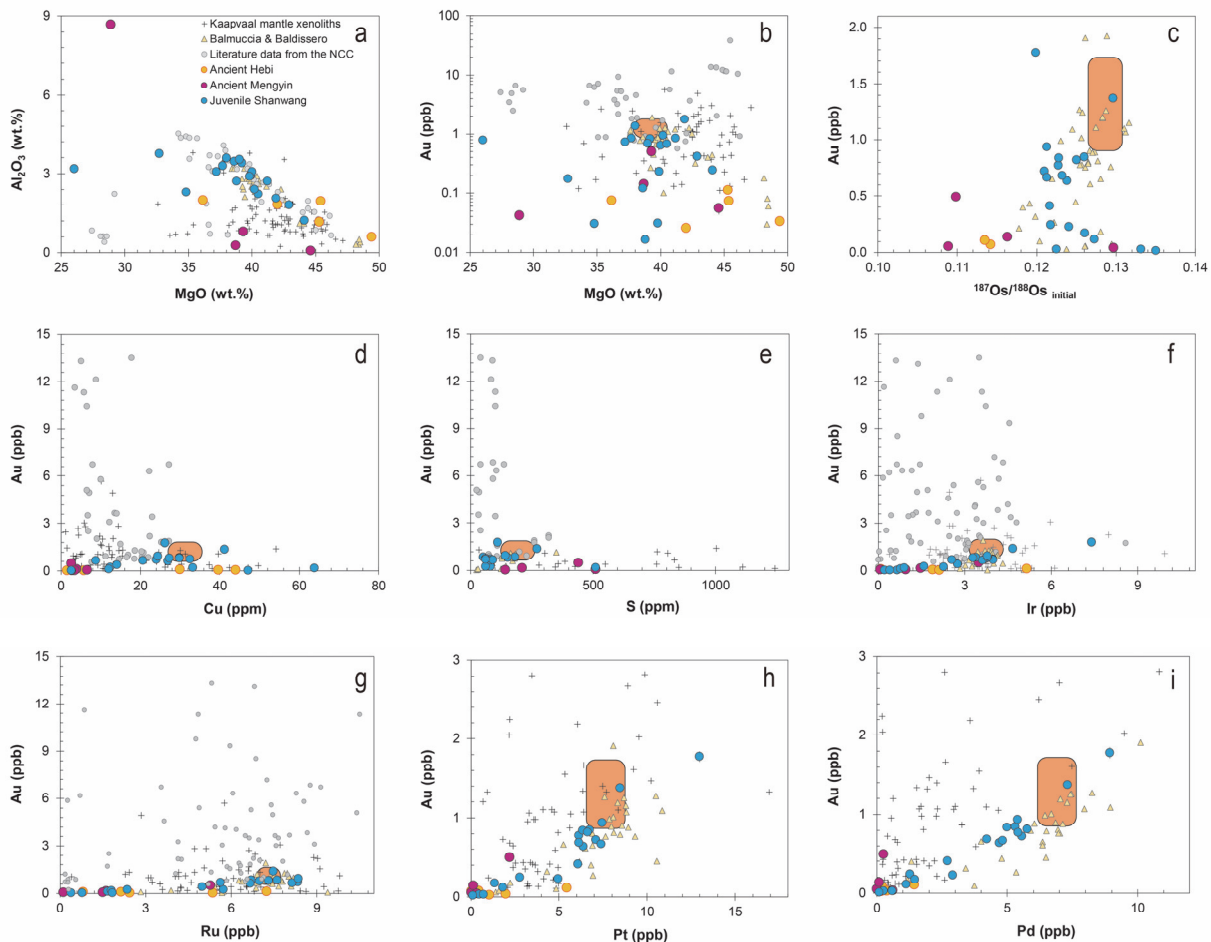
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315 **Figure DR2.** Comparison of the gold contents obtained by two independent methods: 1)  
 316 internal standardization (X axis) and 2) standard addition (Y axis). These two methods  
 317 yielded consistent results within a few percent (a). The typical uncertainty is 5-10 % (2sd) for  
 318 gold and could be larger for samples with very low contents of < 0.1 ppb (b).



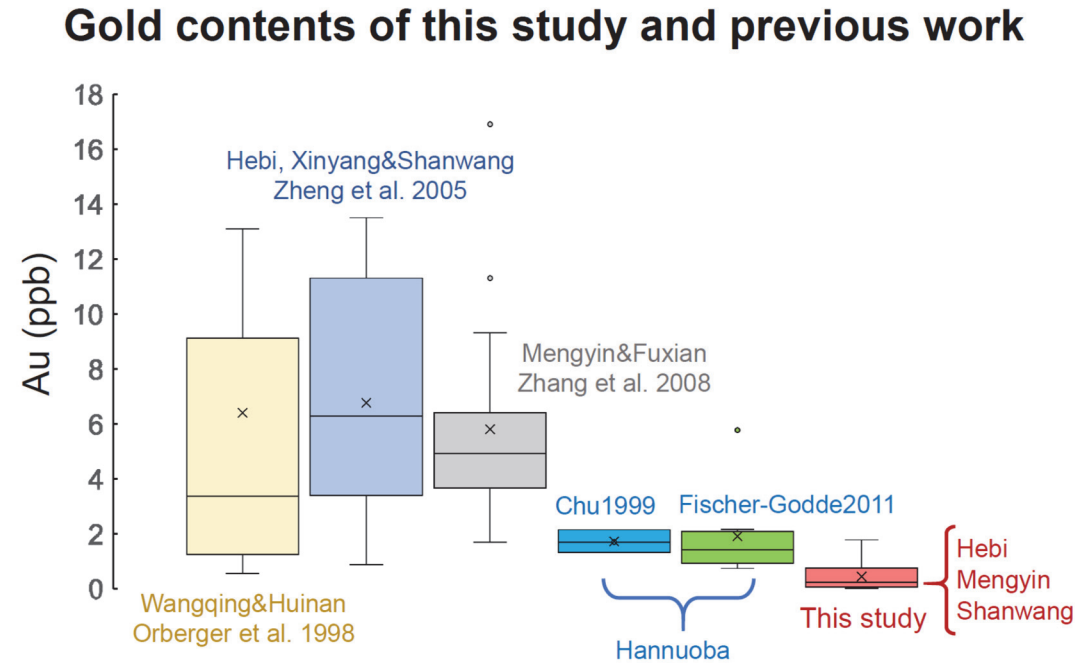
**Figure DR3.** Evaluation of data quality of gold contents in this study. a) Re-analyses of different fractions of aqua regia solution from the same digestion of basalts and peridotites show reproducible values within analytical uncertainty. b) gold contents of replicates of eleven basalts and three peridotites which were digested from different fractions of sample powder, indicating limited effect of sample heterogeneity.



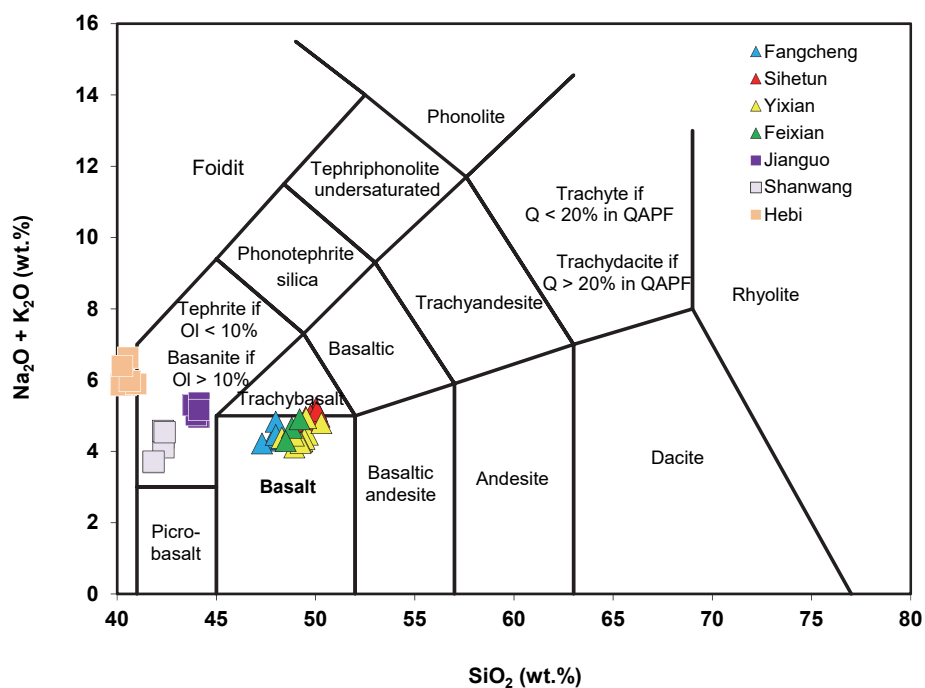
**Figure DR4.** Gold and PGE contents in mantle xenoliths from Mengyin, Hebi and Shanwang.

Kaapvaal mantle xenoliths (Maier et al., 2012) and massif-type peridotites from Ivrea Zone (Wang et al., 2013) which were metasomatized or refertilized by sulfide-bearing components, as well as the estimated range of the primitive mantle ((Becker et al., 2006; Fischer-Gödde et al., 2011; McDonough and Sun, 1995), orange rectangular) are shown for comparison. The gold contents, as well as S, Cu, PGEs, of mantle xenoliths from the NCC have reported before (Chu et al., 1999; Fischer-Gödde et al., 2011; Orberger et al., 1998; Zhang et al., 2008; Zheng et al., 2005). However, literature gold contents show a large range and overall, noticeably higher than the new values in this study and those from Kaapvaal, Ivrea Zone and other locations (Saunders et al., 2018), and do not correlate with S, Cu and PGEs. Note that Kaapvaal mantle xenoliths show very high S contents but gold contents are near-constant (e). Literature values from the NCC are not shown in h and i to highlight the similarity in Au and

PGE contents between this study and the typical refertilized mantle peridotites. The gold contents in mantle xenoliths of the NCC show good correlations with PGEs.



**Figure DR5.** Comparison of gold contents of this study and previous work on mantle xenoliths from the NCC (Chu et al., 1999; Fischer-Gödde et al., 2011; Orberger et al., 1998; Zhang et al., 2008; Zheng et al., 2005). The literature gold contents show a large range; data from Hannuoba tend to be lower than other locations. The new data on Hebi, Mengyin and Shanwang xenolitihs in this study are all lower than older data from the same locations. This study and Fischer-Godde et al (2011) used the similar analytical methods with low total procedural blanks (a few pg). Note that the values in literature (Orberger et al., 1998; Zhang et al., 2008; Zheng et al., 2005) were obtained by NiS fire assay pre-concentration method.



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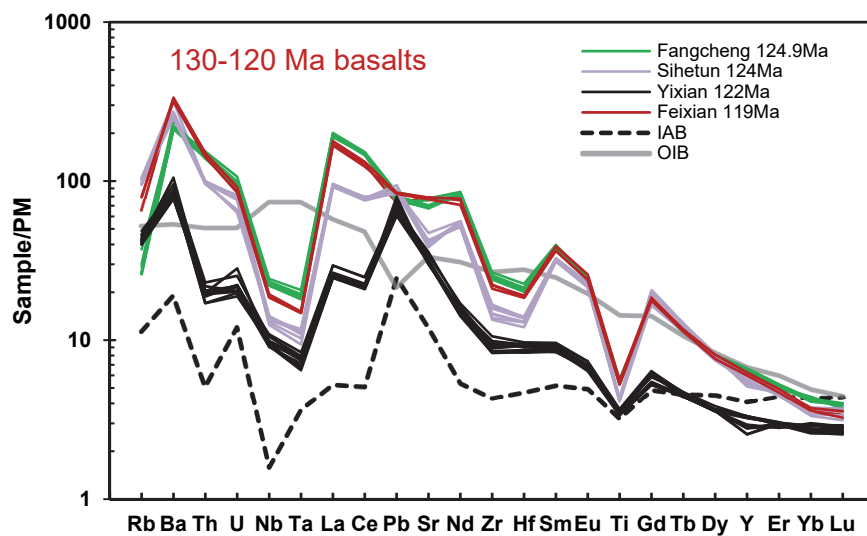
360 Figure DR6. Variations in  $\text{Na}_2\text{O}+\text{K}_2\text{O}$  vs.  $\text{SiO}_2$  (TAS, Le Maitre et al., 1989) for 130-120 Ma

361 and < 110 Ma basaltic samples studied in this study.

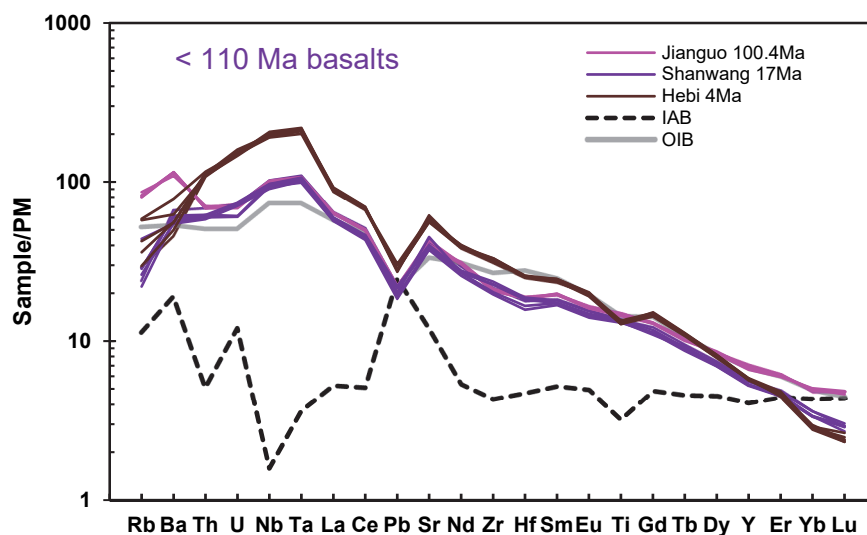
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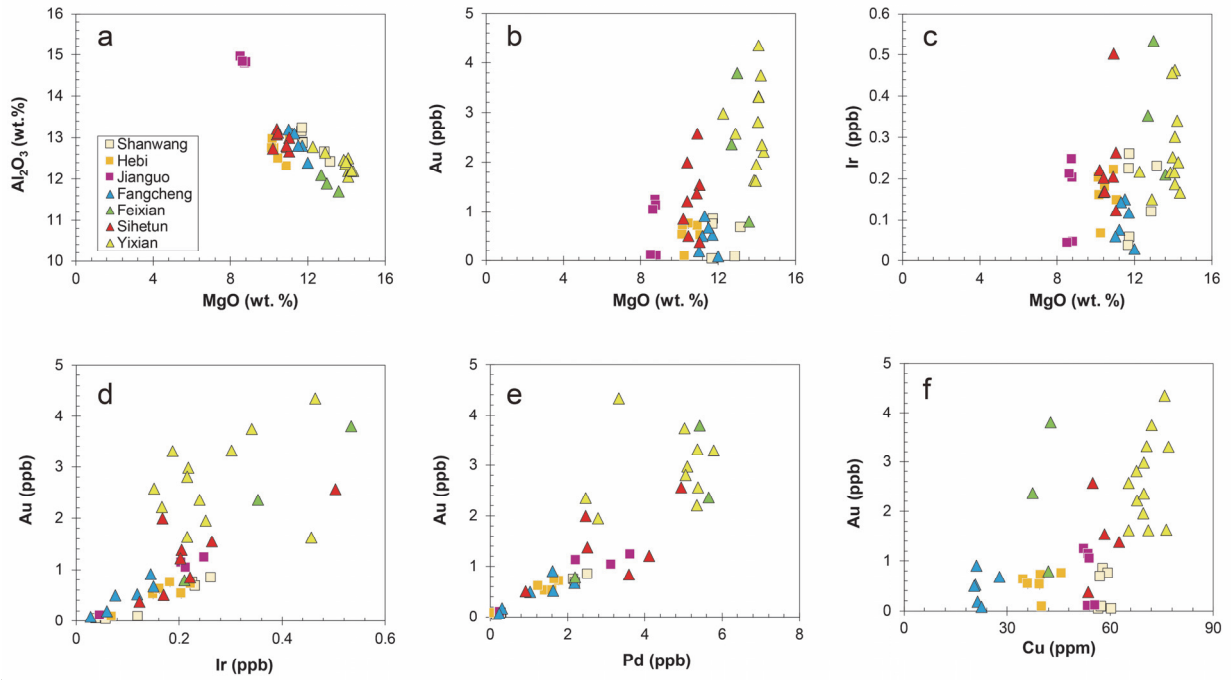


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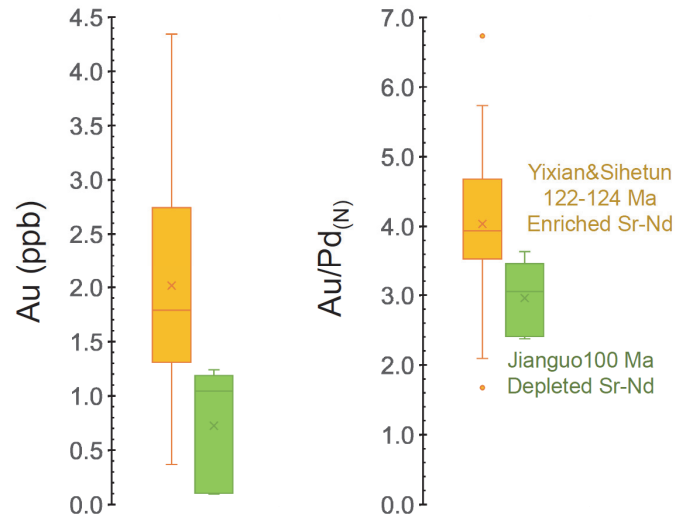


365

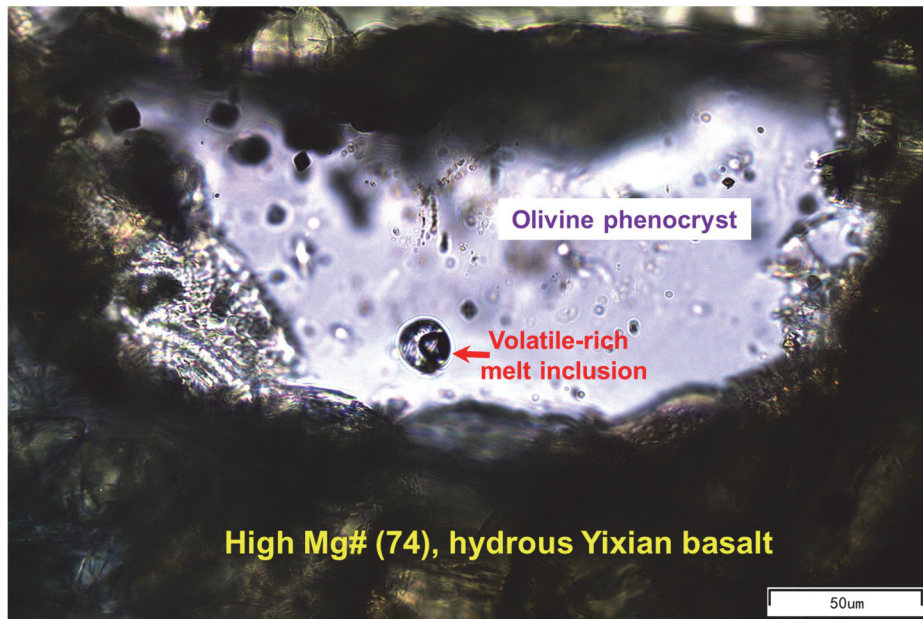
366 **Figure DR7.** Primitive mantle-normalized trace elements patterns of the 130-120 Ma and <  
 367 110 Ma basalts. The 130-120 Ma basalts from the metasomatized SCLM show an island arc-  
 368 basalt like pattern (depletion of Nb-Ta); whereas the < 110 Ma basalts show an OIB-like  
 369 pattern (no depletion of Nb-Ta).



**Figure DR8.** Gold and PGE contents of the 130-120 Ma and < 110 Ma basalts. The gold contents show broadly positive correlations with PGE and Cu contents. Overall, the Au and PGE contents in the 130-120 Ma basalts from the metasomatized SCLM (triangles) are higher than those of the < 110 Ma basalts from the juvenile mantle (squares).



**Figure DR9.** Comparison of Au contents and Au/Pd<sub>(N)</sub> in spatially adjacent basalts that originated from mantle sources with enriched (Yixian and Sihetun) and depleted (Jianguo) Sr-Nd isotopes, respectively. The enriched basalts show higher Au contents and Au/Pd<sub>(N)</sub> than the depleted basalts.



**Figure DR10:** Volatile-rich, hydrous 130-120 Ma basalts derived from the metasomatized SCLM. Volatile-rich melt inclusions are often present in the early crystallized olivine phenocryst of the primitive, hydrous, isotopically enriched basalts (erupted at 125 Ma). Here shown is the Yixian basalt YX-25 with bulk rock Mg# of 74 and olivine Mg# of 88-91 (Geng et al., 2019a) and bulk rock gold of 3.3 ppb.

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