

Mixing matters: resolving the fluid sources of metasediment-hosted auriferous veins in the Grampian Terrane of the Northern British Isles

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Abstract. Throughout the Grampian Terrane of Britain and Ireland, there are multiple occurrences of vein-hosted gold deposits, some of which have commercial value. However, it is unclear whether the fluid source of several auriferous veins in the Grampian Terrane is magmatic or metamorphic. The veins are structurally controlled and mineralogically simple, although the fluid inclusion data and regional geochemical signatures are typical of magmatic mineralization processes. To evaluate the genesis of these more unusual occurrences, we conducted sulfur and lead isotope studies on gold deposits around Loch Tay, which is situated in Central Scotland, and compared the results to regional data from the Grampian Terrane. Pb isotope data from galena in several Scottish gold deposits support a mantle-derived source, although the sulfur isotope profiles suggest that at some localities mantle-derived magmatic fluids have mixed with crustal fluids, obscuring the isotopic signature. The results from our study represent an important case study for other researchers of the challenges that can arise when interpreting isotopic datasets associated with mineralization processes involving fluid mixing, which has been influential in determining the isotopic signatures of gold deposits in Scotland.

1 Introduction

Auriferous quartz \pm carbonate veins in metamorphic terranes have been the focus of much debate, particularly regarding the role of magmatic fluids in the mineralization process (Goldfarb and Pitcairn 2023). There are several examples of 'orogenic' veins globally where opinion is split over whether the fluid source is metamorphic or magmatic (Goldfarb and Pitcairn 2023). To investigate this issue, we collected

sulfur and lead isotope data from a gold-bearing vein system around Loch Tay, situated in the Scottish Grampian Terrane (Fig. 1). Here, there are several veins resembling 'orogenic' gold deposits with characteristics of both magmatic and metamorphic mineralizing fluids, meaning it is an ideal setting to investigate whether veins resembling 'orogenic' gold deposits can arise as a result of magmatic-hydrothermal processes. Whilst gold has been mined by artisanal methods in the Grampian Terrane for centuries, commercial operations (e.g. Cavanacaw and Cononish; Fig. 1) have only been initiated in recent decades.

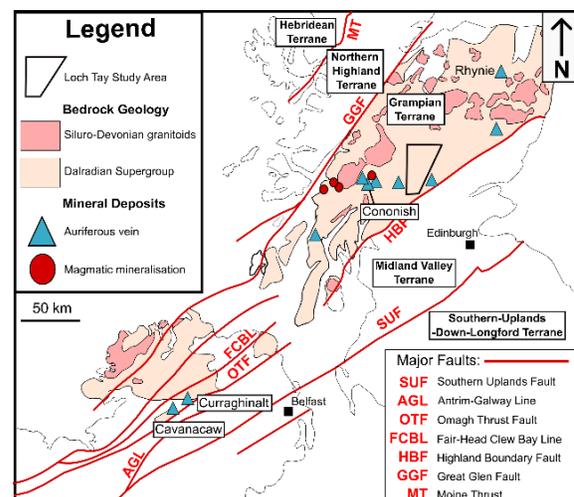


Figure 1. Occurrences of gold mineralization in the Grampian Terrane (adapted from Webb et al. 2025).

The Loch Tay region hosts several gold-bearing veins (Fig. 2) that consistently trend 120-140°, potentially indicating they are genetically

related (Corkhill et al. 2010). Whilst the mineralogy (simple gold-bearing quartz ± carbonate veins), host rocks (Phanerozoic metasediments) and structural controls are typical of 'orogenic' gold-bearing veins attributed to metamorphic processes, fluid inclusion studies show high salinities that are rare in metamorphic fluids and more similar to magmatic fluids (14 wt % NaCl; Naden 2010). Furthermore, soil sampling has discovered regionally elevated Bi concentrations (Corkhill et al. 2010). The veins display textures that are typical of brittle deformation at shallow (< 5 km) depths (syntaxial quartz, cockade textures, cataclastic sulfides) (Webb 2024). To resolve the genesis of the mineral system, we derived sulfur and lead isotope datasets in our study and interpreted the results within the context of the known geological history of the area and previous isotopic studies on other geological features within the wider Grampian Terrane.

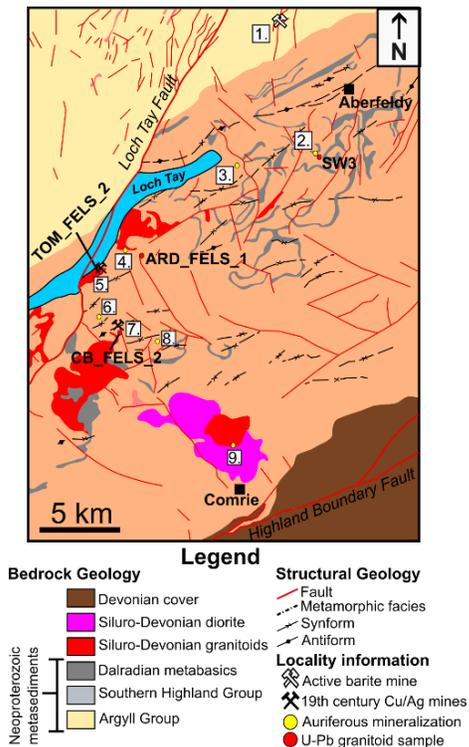


Figure 2. Geology of the Loch Tay region (Webb et al. 2025). 1 = Foss, 2 = Calliachar-Urilar, 3 = Tombuie, 4 = Lead Trial, 5 = Tomnadashan, 6 = Finglen Vein, 7 = Coire Buidhe, 8 = Glen Almond Vein, 9 = Comrie Pluton. CA-ID-TIMs dating of zircons from several granitoids (red dots) yielded ages of c. 420-418 Ma (Webb, 2024).

2 Methodology

Ninety-three $\delta^{34}\text{S}$ analyses on mineral separates were conducted at the University of Leeds in an Elementar PYROcube coupled to an IsoPrime continuous flowmass spectrometer. At the University of Glasgow, fifty-four in-situ sulfur isotope analyses were performed on polished blocks via a laser ablation system. Forty-two Pb isotope measurements from thirty-three samples were conducted at the British Geological Survey and University College Dublin using a Thermo Fisher Scientific Neptune Plus MC-ICP-MS coupled with a Jet Interface.

3 Sulfur isotope results

Tomnadashan and Lead Trial record $\delta^{34}\text{S}$ values of +1 to +3 ‰, whilst the other Loch Tay veins are isotopically heavier (+3 to +12 ‰; Fig. 3). Although the sulfur isotope data from Lead Trial and Tomnadashan are consistent with a magmatic-hydrothermal source (Webb et al. 2025), the signatures of the metasediment-hosted veins are more typical of crustal sources of sulfur (+5 ‰; Hutchison et al. 2020).

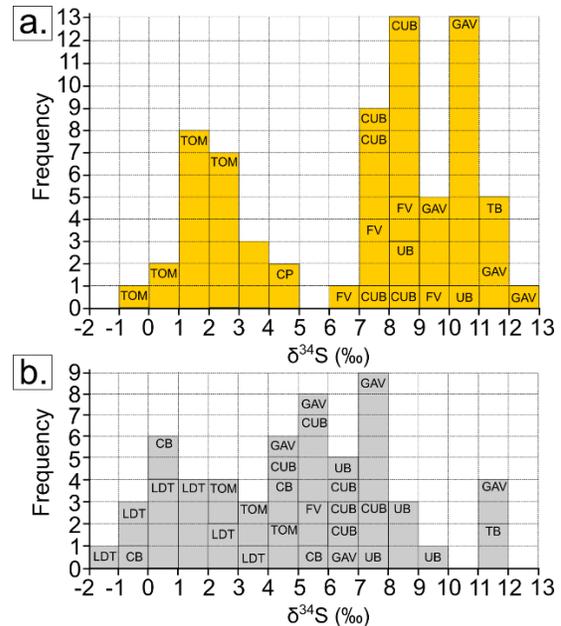


Figure 3. $\delta^{34}\text{S}$ data from sulfides around Loch Tay (Webb et al. 2025). **a.** Pyrite. **b.** Galena (usually paragenetically later). Abbreviations are as follows: TOM = Tomnadashan, CP = Comrie Pluton, FV = Finglen Vein, UB = Urilar, CAL = Calliachar, GAV = Glen Almond Vein, TB = Tombuie, LDT = Lead Trial.

4 Pb isotope results

The Pb isotope studies demonstrate that Lead Trial is the least radiogenic locality around Loch Tay, although most veins show a close alignment with the Zartman and Doe (1981) curve for Pb evolution in the mantle (Fig. 4).

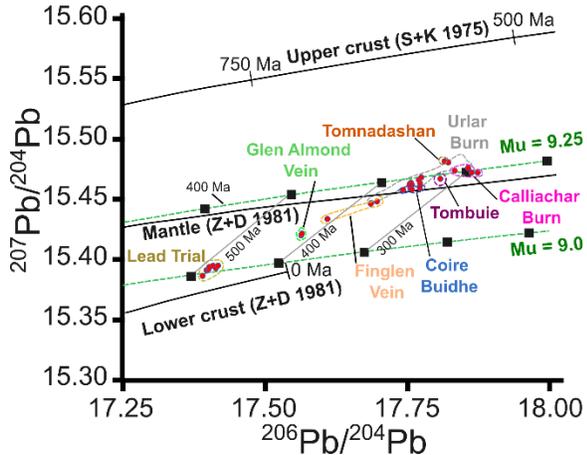


Figure 4. $^{207}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ isotope ratios of galena from Loch Tay (Webb et al. 2025). Pb evolution curves (Stacey and Kramers, 1975; Zartman and Doe 1981) have also been added to the graph.

5 Assessing fluid and metal sources of Grampian gold deposits

Throughout Scotland, there are several instances of coeval gold mineralization and magmatism, including at Loch Tay; Re-Os dates of c. 425–417 Ma have been reported from molybdenite at Tomnadashan, which is consistent with the age of several local intrusions (c. 420–418 Ma; Webb 2024). Furthermore, Cononish and Rhynie (Fig. 1) are spatially associated with granitoids and record mineralization ages (408–407 Ma; Rice et al. 2012) that are coeval with Late Caledonian magmatism (c. 430–390 Ma; Oliver et al. 2008). Collectively, the evidence from geochronology strongly indicates a link between magmatism and gold mineralization in some parts of Scotland, with the mineralization ages postdating the peak metamorphic event (c. 475–465 Ma; Rice et al. 2016) by > 50 Ma. The Pb isotope data support this interpretation; the Pb values from the auriferous veins within the Grampian Terrane are considerably less radiogenic than those reported from Phanerozoic orogenic gold deposits, which

tend to show alignment with crustal sources of Pb (Mortensen et al. 2022).

However, the sulfur isotope profiles from several gold deposits in the Grampian Terrane (Fig. 5a) show affinities with both magmatic-hydrothermal and crustal sulfur sources. In some instances, magmatic-hydrothermal fluids may have mixed with and assimilated other fluids in the upper crust. Potential candidates include meteoric, basinal and metamorphic fluids, depending on the specific deposit in question. Nonetheless, a crustal fluid contribution during the mineralization process can explain the hybrid characteristics of several auriferous veins in the Grampian Terrane, including those at Loch Tay. The Pb isotope values of the SEDEX horizons also overlap with those from the Loch Tay galena samples (Fig. 5b), which may suggest that local SEDEX layers have contributed Pb to the mineralization locally. However, other metasedimentary Pb sources (Fig. 5b) do not overlap with Pb from the auriferous veins in the Grampian Terrane (Fig. 5b), potentially indicating that Pb was not sourced from other non-SEDEX lithologies in the Dalradian Supergroup during the mineralization processes.

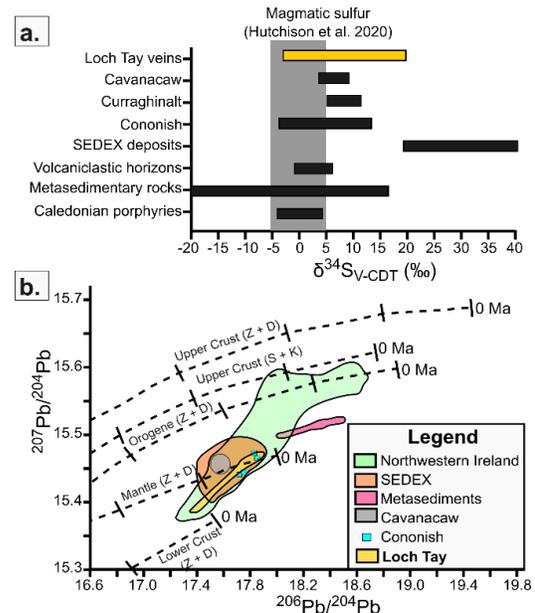


Figure 5. Sulfur and lead isotope results from this study presented alongside regional data from Scotland and Ireland. **a.** $\delta^{34}\text{S}$. **b.** Pb isotope data. Adapted from Webb et al. 2025.

A genetic model involving the mixing between magmatic-hydrothermal fluids and crustal sulfur sources (Fig. 6) has been described at Cononish (Hill et al. 2015), and our study hereby extends this scenario to Loch Tay (Fig. 1).

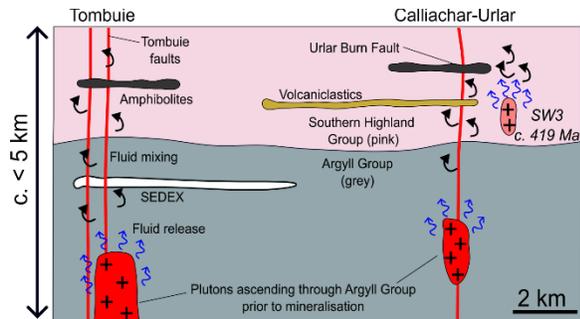


Figure 6. Genetic processes implicated in gold mineralization at Cononish and Loch Tay (Hill et al. 2015; Webb et al. 2025), presented using an approximated NW-SE cross-section through Tombuie and Calliachar-Urilar (Fig. 2).

6 Conclusions

Based on our study, it is possible to make the following conclusions:

1. Several gold deposits in Scotland record Pb isotope values that are typical of mantle or lower crustal sources.
2. $\delta^{34}\text{S}$ signatures typical of magmatic or mantle-derived sources, in addition to coeval magmatism at several localities, may suggest that mantle-derived magmatic fluids are a typical causative agent in Grampian gold deposits.
3. The sulfur isotope data of vein-hosted deposits in the Grampian Terrane are indicative of mixing between magmatic-hydrothermal and crustal fluids, a process that has been particularly influential in central Scotland.

Fluid mixing may have resulted in the incorrect classification of several deposits globally as 'orogenic'. Future research should consider this possibility when evaluating isotopic datasets from auriferous veins hosted in metamorphic terranes, which in some cases may be mistakenly interpreted as having a metamorphic fluid source.

Acknowledgements

Thanks to National Museums of Scotland and National Museum of Wales for loaning samples.

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