



An atmospheric source of S in Mesoarchaeoan structurally-controlled gold mineralisation of the Barberton Greenstone Belt



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ABSTRACT

The Barberton Greenstone Belt of southern Africa hosts several Mesoarchaeoan gold deposits. The ores were mostly formed in greenschist facies conditions, and occur as hydrothermal alteration zones around extensional faults that truncate and post-date the main compressional structures of the greenstone belt. Ore deposition was accompanied by the intrusion of porphyries, which has led to the hypothesis that gold may have been sourced from magmas. Because the transport of Au in the hydrothermal fluids is widely believed to have involved S complexes, tracing the origin of S may place strong constraints on the origin of Au. We measured multiple S isotopes in sulfide ore from Sheba and Fairview mines of the Barberton Greenstone Belt to distinguish “deep” S sources (e.g. magmas) from “surface” S sources (i.e. rocks of the volcano-sedimentary succession that contain S processed in the atmosphere preserved as sulfide and sulfate minerals). Ion probe (SIMS) analyses of pyrite from ore zones indicate mass-independent fractionation of S isotopes ($\Delta^{33}\text{S} = -0.6\text{‰}$ to $+1.0\text{‰}$) and the distribution of the analyses in the $\Delta^{33}\text{S}$ – $\delta^{34}\text{S}$ space matches the distribution peak of previously published analyses of pyrite from the entire volcano-sedimentary succession. Notwithstanding that the H_2O – CO_2 components of the fluids may have been introduced from a deep source external to the greenstone belt rocks, the fact that S bears an atmospheric signature suggests the hypothesis that the source of Au should also be identified in the supracrustal succession of the greenstone belt. Our findings differ from conclusions of previous studies of other Archaean shear-hosted Au deposits based on mineralogical and isotopic evidence, which suggested a magmatic or mantle source for Au, and imply that there is no single model that can be applied to this type of mineralisation in the Archaean.

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1. Introduction

The Palaeoarchaeoan Barberton Greenstone Belt of southern Africa hosts some of the oldest gold mineralisation known (Anhaeusser, 1976; de Ronde et al., 1991; Dirks et al., 2013; Dziggel et al., 2010) (Fig. 1). These deposits have proved to be an important source of Au since their discovery in the 1880s, and have produced more than 345 tons of Au (Anhaeusser, 1976; Dirks et al., 2009) (Fig. 1). Most deposits are hosted in greenschist facies rocks, where gold mineralisation is structurally controlled and occurs along extensional faults cross-cutting the main compressional structures of the greenstone belt, which extend for several tens of km along strike (Dirks et al., 2013). The ore is dominated by pyrite and arsenopyrite, and gold is mostly finely dispersed in sulfides,

either present in the mineral structure or as sub-microscopic inclusions (“invisible gold”) (Craig et al., 1998). In this kind of structurally-controlled gold deposits, the mineralising fluids are typically aqueo-carbonic and have low to moderate salinity (Goldfarb et al., 2001; Mikucki and Ridley, 1993), and are interpreted to originate from a deep source (Salier et al., 2005). The origin of mineralising fluids is controversial, especially in Archaean deposits, and mineralogical, elemental and isotopic evidence seems to point towards either metamorphic or igneous sources, or a combination of these (Hutti mine, India; Rogers et al., 2013; Western Australia, Doublier et al., 2014; Wang et al., 1993). Propagation of these fluids along crust-scale structures is believed to be responsible for the formation of deposits in a single region over a range of depths and temperatures (from <200 to >500, and possibly ≤ 700 °C) (“crustal continuum model”; Barnicoat et al., 1991; Groves, 1993; Phillips and Powell, 2009; Kolb et al., 2015). Deposition of Au would have occurred by reaction of the mineralising

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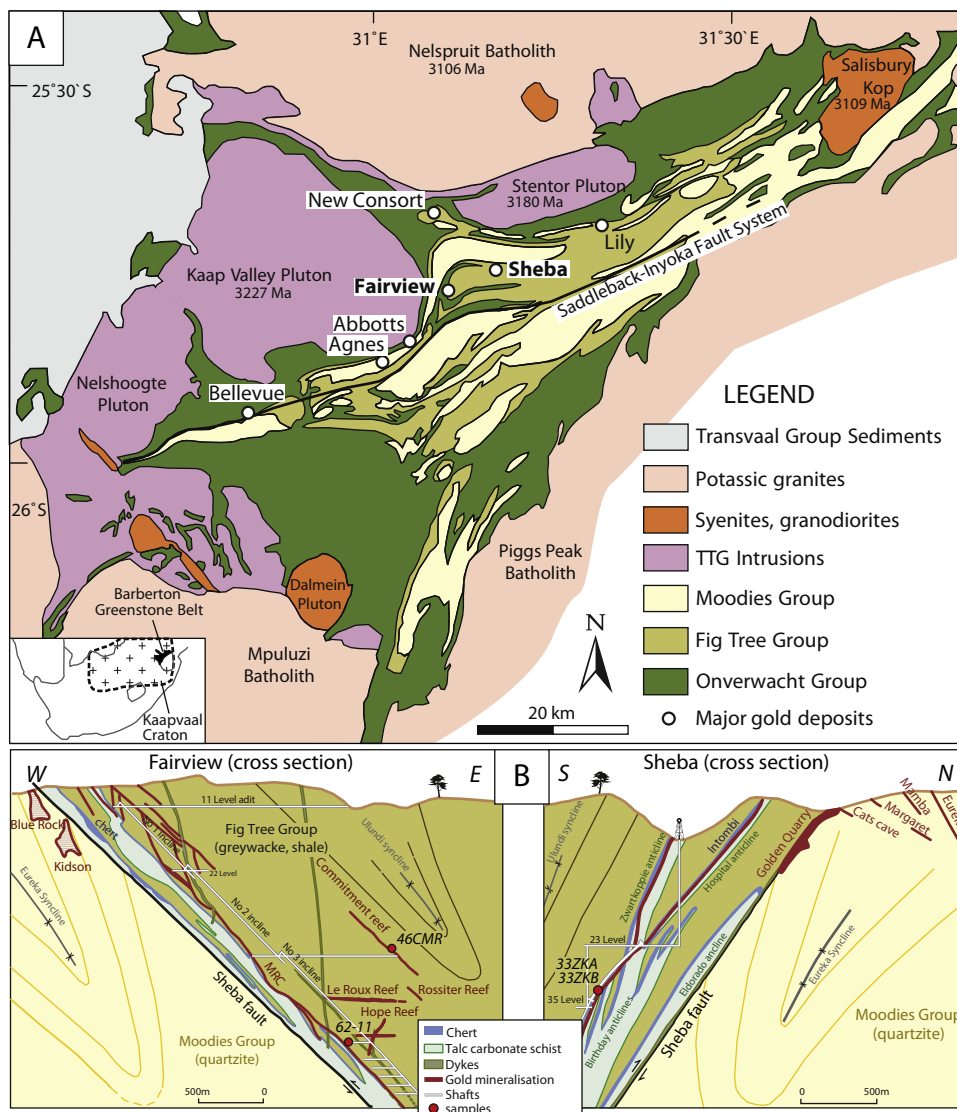


Fig. 1. (A) Geological map of the Barberton Greenstone Belt and distribution of the main gold deposits (modified from de Ronde et al., 1992). (B) Cross sections of Fairview and Sheba mines (modified from Barberton gold mines, 2014).

fluid with the host rocks or by fluid mixing (Bateman and Hagemann, 2004; Evans et al., 2006), or vapour separation (de Ronde et al., 1992; Mikucki and Ridley, 1993). The origin of S is an important aspect in the study of deposits hosted in Archaean greenstone belts and other structurally controlled Au deposits, since hydrosulfide complexes [Au(HS)₂ and AuHS] are believed to be the main Au transporting agents (Benning and Seward, 1996; Pokrovski et al., 2014; Seward, 1973; Simon et al., 1999). Therefore, identifying the source of S can help constrain the origin of Au in these deposits, which has so far remained elusive (Tomkins, 2013; Gaboury, 2013; Kendrick et al., 2011; Pitcairn et al., 2006).

In order to distinguish deep (magmatic- or mantle-related) from sedimentary sources of sulfur, multiple S isotope analyses can be used. Mass-independent fractionation of S isotopes (MIF-S) is a common feature of Archaean and early Palaeoproterozoic (>2.4 Ga) sedimentary and diagenetic sulfur minerals (sulfides and sulfates; Ono et al., 2003). This S isotope signature is believed to originate from ultraviolet radiation-induced reactions of S gas species (e.g. SO₂, SO₃) in anoxic atmosphere, and thus to be a distinctively atmospheric signature (e.g. Farquhar et al., 2000). MIF-S

can be expressed as $\Delta^{33}\text{S} = \delta^{33}\text{S} - 1000 \cdot [(1 - \delta^{34}\text{S}/1000)^{0.515} - 1]$, as ‰ variation. Among the products of this reaction, water-soluble sulfate with $\Delta^{33}\text{S} < 0$ and relatively insoluble elemental S with $\Delta^{33}\text{S} > 0$ can then be separated by bodies of water upon deposition on the Earth's surface, incorporated into the sediments, and preserved in the rock record in the form of sulfide and sulfate minerals. In the Barberton Greenstone Belt MIF-S has been described in pyrite and barite from several stratigraphic units (Grosch and McLoughlin, 2013; Montinaro et al., 2015; Philippot et al., 2012; Roerdink et al., 2012, 2013).

Following the discovery of MIF-S in sedimentary environments, MIF-S signal has also been found in Neoproterozoic VMS deposits (Jamieson et al., 2013), in diamond-hosted sulfide inclusions (Farquhar et al., 2002; Thomassot et al., 2009), in the Palaeoproterozoic Rustenburg Layered Suite of the Bushveld complex (Penniston-Dorland et al., 2012) and in olivine-hosted sulfide inclusions in Cainozoic plume-related ocean island basalt magmas (Cabral et al., 2013). These findings have revealed a feedback between surface and deep S cycle, indicating that S processed in the atmosphere during the Archaean can be stored in the crust or the mantle, and be recycled back to the surface through different

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