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Mobility of gold during metamorphism of the Dalradian in Scotland

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ABSTRACT

Mobility of Au and related metals during metamorphism has been suggested to be the source of metals enriched in orogenic Au deposits. This study investigates the mobility of Au, As, and Sb during metamorphism of the Dalradian metasedimentary rocks of Scotland. The metamorphic processes in the Dalradian of Scotland are extremely well studied, and the terrane is an ideal area to investigate mobility of these metals. Our results show that of the 25 major and trace elements analysed, only Au, As, Sb, S and volatile contents as shown by loss on ignition (LOI) values show systematic variation with the metamorphic grade of the samples. Average Au concentrations decrease from 1.1 ± 0.55 ppb and 0.72 ± 0.34 ppb in chlorite and biotite zone rocks down to 0.4 ± 0.22 ppb and 0.34 ± 0.13 ppb in kyanite and sillimanite zone rocks. Average As concentrations decrease from 4.8 ppm (range 0.5 to 17.8 ppm) and 1.96 ± 1.9 ppm in chlorite and biotite zone rocks down to 0.24 ± 0.15 ppm and 0.2 ± 0.12 ppm in kyanite and sillimanite zone rocks. Average Sb concentrations decrease from 0.18 ± 0.15 ppm and 0.11 ± 0.10 ppm in chlorite and biotite zone rocks down to 0.04 ± 0.02 ppm in both kyanite and sillimanite zone rocks. Sulphur and LOI concentrations also show significant decreases. Mass balance calculations indicate that compared to chlorite and biotite zone samples, sillimanite zone samples have an average mass loss of 62 ± 14%, 94 ± 4% and 74 ± 14% for Au, As, and Sb respectively. Every 1 km^3 of chlorite–biotite zone mixed psammitic–pelitic protolith rock that is metamorphosed to sillimanite zone conditions would release 1.5 t Au, 8613 t As, 270 t Sb, and 1.02 Mt S. The mobility of these elements is strongly controlled by the paragenesis of sulphide minerals. Pyrite, sphalerite, galena and cobaltite (as well as gersdorffite) decrease in abundance with increasing metamorphic grade in the Dalradian metasedimentary rocks. A critical aspect of the sulphide paragenesis is the transition of pyrite to pyrrhotite. This transition is complete by mid greenschist facies in the Loch Lomond samples but is more gradual at Glen Esk occurring between biotite and sillimanite zones. The Au, As, and Sb content of the sulphide assemblage also decreases with increasing metamorphic grade, and we suggest that this is a controlling factor on the mobility of these metals from the Dalradian metasedimentary rocks during metamorphism. Chlorite may be an important host mineral for As in the greenschist facies rocks. Breakdown of chlorite indirectly drives the mobility of Au, As, and Sb, as this produces the bulk of metamorphic fluid that drives transition between pyrite and pyrrhotite.

We suggest that there is potential for significant undiscovered mineralisation in the Central and SW Highlands of Scotland. However, as the total mass of gold mobilised is lower than observed in other metasedimentary terranes such as the Otago and Alpine Schist's, New Zealand, very efficient fluid focussing and trapping mechanisms would be required to form large deposits in the Dalradian of Scotland.

1. Introduction

The genetic processes that form orogenic Au deposits have long been a source of debate. A key part of the genetic model is constraining the sources of fluids and enriched metals such as Au, As, and Sb. Much recent research, particularly that focussing on Phanerozoic deposits, has favoured a metamorphic dehydration model where metal-rich fluids are produced during prograde metamorphic dehydration reactions (Gaboury, 2013; Goldfarb et al., 2005; Kerrich et al., 2000; Large et al., 2007, 2011; Mortensen et al., 2010; Phillips and Powell, 2010; Pitcairn et al., 2006a, 2010a, 2014a; Tomkins, 2010, 2013). The metamorphic sequence containing the deposits is invariably implied to be the source of metals, but different source lithologies and different scales of metal mobility have been proposed. These include regional-scale devolatilisation of large volumes of metasedimentary rock (Pitcairn et al., 2006a, 2010a, 2014a,b), or metabasaltic rock (Bierlein and Pisarevsky, 2008; Willman et al., 2010) and localised leaching of fertile metasedimentary rock (Large et al., 2007, 2011). One of the few direct investigations of the sources of metals reported systematic depletions of Au and associated elements (As, Ag, Hg, Mo, Sb and W) from the

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high metamorphic grade metasedimentary rocks in the Otago and Alpine Schists, South Island, New Zealand (Pitcairn et al., 2006a, 2010a, 2014a). The suite of elements depleted in the high-grade rocks is identical to those enriched in the orogenic Au deposits of Otago, and the depletions are interpreted to represent the source of metal enrichments in the deposits at shallower levels.

The metamorphic dehydration model successfully explains many aspects of the formation of orogenic Au deposits but we do not yet fully understand why some metamorphic belts are heavily mineralised whereas others contain few Au deposits. Potential explanations include the Au content of the protolith source rock, the relative timing of heat input, prograde dehydration and deformation, the efficiency of fluid focussing, and the efficiency of trapping mechanisms that cause Au precipitation. An important question is whether Au is always mobilised during prograde metamorphism. Will Au always be mobilised from a sequence of sedimentary and/or volcanic rocks that are metamorphosed to at least amphibolite facies conditions, or are certain lithochemical or hydrothermal conditions required for Au to be released into the prograde metamorphic fluid?

In this study we investigate the mobility of Au and related elements As and Sb during metamorphism of the Dalradian Supergroup metasedimentary terrane in Scotland (Fig. 1). This terrane is arguably the most extensively studied metamorphic terrane in the world, with much of our understanding of Barrovian metamorphism and the role of fluids in metamorphic processes coming from studies of these rocks (Ague, 1997; Ague and Baxter, 2007; Barrow, 1893; Chinner, 1966; Graham et al., 1983; Masters and Ague, 2005; Skelton et al., 1995; Tilley, 1925; Vorhies and Ague, 2011). Gold mineralisation has occurred in the Dalradian such as at Cononish in the Central Highlands of Scotland, and also more extensively in the Irish Dalradian (Parnell et al., 2000; Hill et al., 2013). However, there is uncertainty surrounding the deposits that do occur and their genetic relationship to Dalradian metamorphism (Hill et al., 2013). Comparisons between the mobility of Au in a poorly mineralised metamorphic belt such as the Dalradian with more heavily mineralised equivalents can provide useful insights into the formation of orogenic Au deposits, and help identify the factors that control the formation of abundant Au deposits.

We use major and trace element analyses of a large suite of rocks representative of the lithological and metamorphic variability to quantify the mobility of Au, As, and Sb during metamorphism of Dalradian metasedimentary rocks from Scotland. We also investigate the occurrence and composition of sulphide minerals in these rocks to further understand the mineral reactions that mobilise these metals. We focus specifically on Au, As and Sb as these elements have given the strongest signals of metamorphic mobility in previous studies (Pitcairn et al., 2006a, 2010a, 2014a). Specifically developed analytical methods with ultra low detection limits have been used for this study (Pitcairn et al., 2006b) as these methods provide the analytical power to observe depletions in low abundance elements. Our results provide further understanding of the controls on development of abundant orogenic Au mineralisation during metamorphism.

2. Geological setting

The Dalradian Supergroup in Scotland is bound to the north by the Great Glen Fault, and to the south by the Highland Boundary Fault

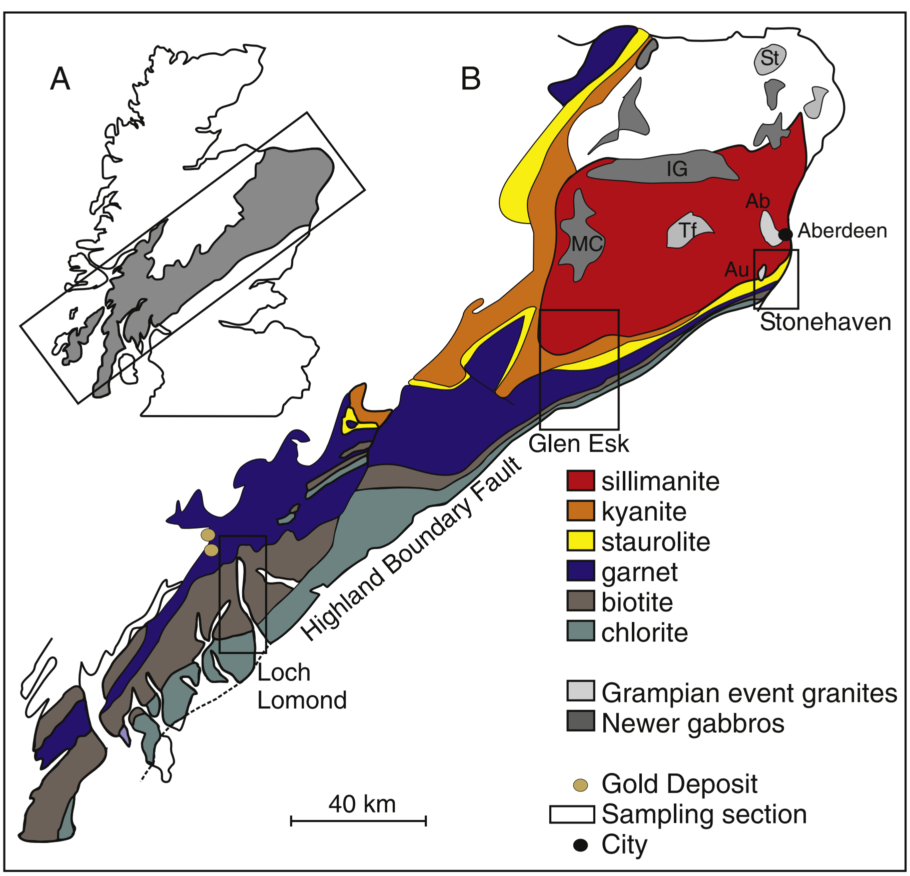

Fig. 1. A — The distribution of Dalradian rocks in Scotland and the position of map B. B — Geological map of central Scotland showing the distribution of the Dalradian Supergroup rocks, the locations of different metamorphic zones, and the locations of sampling sections. Abbreviations: Au — Auchlee granite, Ab — Aberdeen granite, IG — Insch gabbro, MC — Morven–Cabrach gabbro, St — Strichen granite, Tf — Tillyfourie granite.
Modified from Vorhies and Ague (2011).

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