



Research paper

The origin of Late Devonian (Frasnian) stratiform and stratabound mudstone-hosted barite in the Selwyn Basin, Northwest Territories, Canada



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ABSTRACT

Multiple occurrences of bedded barite (BaSO_4) are preserved in mudstones of the upper-most Canol Formation (Frasnian) of the Selwyn Basin (Canada). Samples of barite were collected from eight sites (seven measured sections), and examined using petrographic and isotopic techniques ($^{87}\text{Sr}/^{86}\text{Sr}$, $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values) to determine whether the barite has a diagenetic or hydrothermal origin.

Barite at all locations occurs in two morphologies: (1) laminated barite composed of thin layers of microcrystalline barite with lesser amounts of pyrite and hyalophane (Ba-K-feldspar) interlaminated with mudstone, and (2) nodular barite consisting of barite, hyalophane, cymrite (hydrated Ba-silicate) and minor pyrite. At the Cowan occurrence, one sample has a mineral assemblage comprising barite and nodular barium carbonates (witherite, norsethite).

The $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values of the laminated barite co-vary, and range between +24.1 and +35.3‰ and +14.8 to +18.3‰ respectively, with the exception of one sample with $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values of +32.9‰ and −0.5‰. The laminated samples have an average $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.7085 ($n = 8$), which is consistent with Frasnian seawater (0.7081) that has undergone some minor water sediment interaction during early diagenesis.

The nodular barite has highly variable $\delta^{34}\text{S}$ (+4.8 to +56.5‰) and $\delta^{18}\text{O}$ (+9.2 to +19.3‰) values, which developed from more evolved diagenetic pore fluids with limited sulphate re-supply (i.e., as a relatively closed system). In these conditions sulphate reduction likely occurred at a slower rate, and the sulphate in the barite records both kinetic ($\delta^{34}\text{S}_{\text{SO}_4}$) and equilibrium ($\delta^{18}\text{O}_{\text{SO}_4}$) isotope effects. One sample of nodular barite has a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7086. The single sample with a barite and barium-carbonate assemblage at Cowan has a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7133, a $\delta^{34}\text{S}$ value of +23.9‰, and a $\delta^{18}\text{O}$ value of +10.7‰ and may represent a local hydrothermal event at this site.

Our data supports the interpretation that the barite horizon is a stratigraphic marker throughout the Canol Formation. Laminated and nodular barite formed as a result of diagenetic processes, which recycled Ba from organic matter in the sediment below, and reprecipitated it above, the sulphate methane transition zone in the sediments.

The results of this study are similar to those obtained for other Late Devonian sediment-hosted barite occurrences in North America (Yukon, Nevada, New York State), meaning these occurrences likely formed by similar processes and diagenetic pore water conditions within oceanic sediments that covered the North American continent during the Late Devonian. We suggest that the combination of high biological productivity and low seawater sulphate at this time contributed to the efficient cycling of Ba from seawater into the sediments on the continental margin.

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1. Introduction

Barite enrichment is a common feature of organic-rich mudstones deposited throughout the Phanerozoic, and has been identified in sedimentary basins from the Cambrian (Wang and Li, 1991), Late Devonian – Mississippian (Cecile et al., 1983; Johnson et al., 2004; Lash, 2015) and Cretaceous (Griffith and Paytan, 2012). The solubility of barite is low in oxidised, sulphate-bearing fluids ($\log K = 10^{-9.96}$; Bowers et al., 1984). As such, most crustal fluids cannot transport barium together with sulphate, and models of barite deposition in sedimentary systems typically require mixing between barium-rich and sulphate-rich fluids, which occurs primarily in either hydrothermal or diagenetic environments (Jewell, 2000; Griffith and Paytan, 2012; Hanor, 2000). In low temperature marine environments, barite precipitation may initially be mediated by biological processes operating in the water column, resulting in the sedimentation of barite associated with planktonic organisms (e.g., Dehairs et al., 1980; Bishop, 1988). Barium is then re-mobilised during anoxic diagenesis when productivity-derived barium is concentrated in sulphate-depleted, methane-bearing pore fluids during breakdown of organic matter; barite precipitation occurs when these fluids circulate and mix with shallower sulphate-bearing pore waters (e.g., Torres et al., 1996). In contrast, hydrothermal barite forms when high-temperature, barium-bearing fluids ascend from deeper crustal levels and are exhaled into sulphate-bearing seawater (e.g., Jamieson et al., 2016).

Petrographic and isotopic techniques can help distinguish between these two models of barite formation (e.g., Paytan et al., 2002). As Sr is readily substituted into the barite crystal structure (<3 mol%; Monnin and Cividini, 2006), the $^{87}\text{Sr}/^{86}\text{Sr}$ composition of barite is particularly useful for distinguishing between hydrothermal and seawater Sr (and therefore, barite) sources (Griffith and Paytan, 2012). The $^{87}\text{Sr}/^{86}\text{Sr}$ composition of seawater is primarily controlled by detrital (weathering) and hydrothermal (mid-ocean ridge) inputs, which may be recorded by barite in the marine sedimentary record (e.g., Paytan et al., 2002).

For the majority of barite deposits, the sulphate is derived from seawater. Therefore $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values for barite ($\delta^{34}\text{S}_{\text{barite}}$, $\delta^{18}\text{O}_{\text{barite}}$) can be compared to available data for coeval seawater (e.g., evaporites, Claypool et al., 1980; carbonate-associated sulphate, John et al., 2010). The nature of $\delta^{34}\text{S} - \delta^{18}\text{O}$ co-variation in barite can provide important information on fluid chemistry during barite precipitation, and in particular, different pathways of sulphate reduction (e.g., Johnson et al., 2009; Antler et al., 2015). Where barite precipitates directly from seawater (or sediment pore fluids in close communication with seawater) it can also provide an additional proxy for seawater $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values (e.g., Paytan et al., 1998). However, as a reliable record for seawater, barite precipitation must occur during the earliest stages of sedimentation, and prior to modification of pore fluids via bacterial sulphate reduction (BSR) (Cecile et al., 1983; Magnall et al., 2016a).

In the Selwyn Basin (Canada, Fig. 1) there are multiple occurrences of sediment-hosted barite (Cecile et al., 1983; Goodfellow and Jonasson, 1984). Barite is a feature of Late Devonian shale-hosted massive sulphide (SHMS) mineralisation at MacMillan Pass District, Yukon (Ansdell et al., 1989; Gardner and Hutcheon, 1985), and in the Gataga District, British Columbia (Paradis et al., 1998). A temporal and spatial association between the stratiform barite and base metal sulphide mineralisation has led to the general acceptance of a hydrothermal model for barite formation, which involves co-transportation of Ba, Pb and Zn in hydrothermal fluids and precipitation of barite and sulphides when these fluids mixed in a stratified water column (Goodfellow and Jonasson, 1984). In this model, occurrences of so-called 'barren barite' (where base

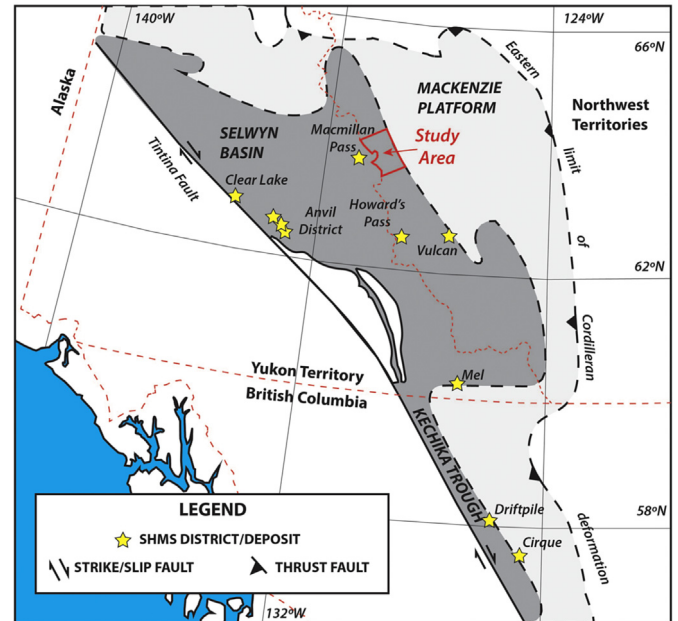


Fig. 1. Location of the Selwyn Basin showing distribution of Devonian-Mississippian strata of basin (dark grey), platform affinities (light grey) and shale-hosted massive sulphide deposits (SHMS, yellow stars), modified from Goodfellow and Lydon, 2007; and Magnall et al., 2015. The study area is within the red outline and is presented in more detail in Fig. 2. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

metal sulphides are absent) are considered to be distal expressions of hydrothermal activity and are thought to precipitate directly as a result of the interaction of hydrothermal fluids with seawater (e.g., Goodfellow, 1987; Goodfellow and Jonasson, 1984; Lydon et al., 1985). This interpretation has led to the assumption that the $\delta^{34}\text{S}_{\text{barite}}$ values are indicative of the composition of coeval seawater. Specifically, previous workers have considered the highly positive $\delta^{34}\text{S}_{\text{barite}}$ values from the Late Devonian deposits to represent periods of basin stagnation, when BSR occurred in a restricted, euxinic ($\text{H}_2\text{S} > \text{SO}_4^{2-}$) water column (e.g., Goodfellow and Jonasson, 1984). However, Magnall et al. (2016a) suggested that at MacMillan Pass, the barite formation *pre-dates* sulphide mineralisation and formed by normal diagenetic processes at a relict sulfate methane transition zone (SMTZ) beneath the sediment water interface (SWI). The hydrothermal event occurred when fluids sourced from a greater depth in the basin migrated along faults and into the shallower, barite-bearing part of the sedimentary column (Magnall et al., 2016b).

There are many other sites with barren stratabound barite in the Selwyn Basin that are not associated with sulphide mineralisation or hydrothermal activity; these units have not been the focus of detailed research and their origin is unknown. Determining whether this barite formed by hydrothermal or diagenetic processes, and the extent to which $\delta^{34}\text{S}_{\text{barite}}$ values record primary seawater, has important implications for accurate paleoenvironmental reconstructions. For example, can Frasnian strata in the Selwyn Basin be compared to modern and ancient continental margin sediments where barium is cycled (e.g., Torres et al., 1996), or did barite form in a restricted basin from a regional hydrothermal source of barium?

In this study, we describe multiple occurrences of barren barite (and associated barium-bearing minerals) in Frasnian strata of the Selwyn basin ≈ 90 – 110 km to the NNE of the SHMS deposits at MacMillan Pass. There is no known hydrothermal activity in the

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