LETTER

# A basement-interacted fluid in the N81 deposit, Pine Point Pb-Zn District, Canada: Sr isotopic analyses of single dolomite crystals

**Paulina Gromek · Sarah A. Gleeson · Antonio Simonetti**

**Abstract** The Mississippi Valley-type Pb-Zn deposits of the Pine Point district (Northwest Territories, Canada) are located close to the eastern edge of the present day Western Canadian Sedimentary Basin. The deposits are thought to have formed as the result of basin-wide fluid flow in the Presqu'ile Barrier, the host to the ore deposits. A laser ablation multi-collector inductively coupled plasma mass spectrometric study of $^{87}Sr/^{86}Sr$ ratios of ore-related dolomites from the N81 deposit at Pine Point indicates that at least two sources of Sr were present in the mineralizing system. One fluid has a range in Sr isotopic values from 0.07073 to 0.71200 and is interpreted to be derived from Middle Devonian seawater that interacted with clastic units in the basin. The second fluid has higher Sr isotopic values (up to 0.71520), similar to those found in some Canadian Shield brines, and is interpreted to represent an evaporated seawater-derived brine which has interacted with crystalline basement rocks. Reactivation of old structures in the basement may have provided a pathway for cross-formation fluid flow to the site of mineralization. The data suggest that the stratigraphic location of the Pine Point District, near the interface between the Western Canadian Sedimentary Basin and its basement, may have exerted a fundamental control on the formation of these deposits.

**Keywords** Strontium isotopes · Dolomite · Laser MC-ICP-MS · Basement · Shield brines · Pine Point

P. Gromek · S. A. Gleeson (✉) · A. Simonetti
Department of Earth and Atmospheric Sciences,
University of Alberta,
Edmonton, AB T6G 2E3, Canada
e-mail: sgleeson@ualberta.ca

*Present Address:*
P. Gromek
BP Canada,
240-4 Avenue SW,
Calgary, AB T2N 2H3, Canada

*Present Address:*
A. Simonetti
Department of Civil Engineering and Geological Sciences,
University of Notre Dame,
156 Fitzpatrick Hall,
Notre Dame, IN 46556, USA

## Introduction

The Pine Point ore district comprises over 100 Pb- and Zn-bearing sulphide bodies hosted by Middle Devonian carbonates of the Western Canadian Sedimentary Basin (WCSB). Pine Point is considered to be a classic example of a Mississippi Valley-type (MVT) district (Leach et al. 2005), and studies on the district have been important in the development of genetic models for MVT deposits (Beales and Jackson 1968). Although some early workers suggested that metals for the Pine Point deposits were derived from the basement to the WCSB (Campbell 1966; Krebs and Macqueen 1984), more recently, a model of large-scale movement of topographically driven fluids related to orogenic events in the WCSB has been proposed (Garven 1985; Qing and Mountjoy 1992, 1994). In this model, the fluids and metals for the deposits are derived from within the sedimentary basin, and the foreland basin-related fluid flow model has been widely applied to other carbonate-hosted mineral deposits (Leach et al. 2005). This model is supported by a regional

west to east variation in C- and O-isotope values, as well as fluid inclusion homogenization temperatures in saddle dolomite cements across the basin, along with an associated change in the average $^{87}Sr/^{86}Sr$ compositions of the carbonates (Qing and Mountjoy 1992).

In fluids derived from seawater, variance from the original $^{87}Sr/^{86}Sr$ seawater composition occurs during water–rock interactions or mixing with fluids from different sources (Mountjoy et al. 1992). In the WCSB, Machel and Cavell (1999) defined an $^{87}Sr/^{86}Sr$ value (0.7120) named the "Maximum Sr Isotope Ratio of Basinal Shale" (MASIRBAS) and suggested that fluids with Sr isotopic values greater than Devonian seawater (i.e. 0.7080), but lower than 0.7120, have acquired some radiogenic Sr from interactions with basinal shales. They also suggested that fluids with isotopic compositions higher than 0.7120 have acquired radiogenic Sr by interacting with radiogenic Precambrian rocks. Thus, the Sr isotopic composition of carbonate minerals has been used to identify fluid flow pathways in the WCSB but can also be a powerful tool in elucidating the origin and evolution of mineralizing fluids in MVT deposits.

Here, we report in situ Sr isotope laser ablation–multicollector inductively coupled plasma mass spectrometry (LA-MC-ICP-MS) data from single crystals of ore-related coarse white dolomites (CWD) and saddle dolomites (SD) from the Pine Point N81 deposit. This technique permits Sr isotope characterization to be carried out at high spatial resolution (hundreds of microns scale) and reveals, for the first time, a high degree of variation in the $^{87}Sr/^{86}Sr$ composition that is not elucidated with bulk $^{87}Sr/^{86}Sr$ analytical methods. These data indicate that the Sr in ore-related carbonates at N81 comes from at least two sources strongly support a role for basement-derived Sr in the mineralizing system and suggest that the topographic fluid flow model should be modified to include some fluid flow across the basement–basin interface.

## Background geology, sample selection and petrography

The Pine Point District lies close to the present day erosional edge of the WCSB where the Paleozoic sediments overlie the crystalline basement rocks of the Churchill Province of the Canadian Shield. The exact nature of the Precambrian basement rocks beneath the Pine Point District is unknown, but two diamond drill holes intersected micaceous quartzite and biotite granodiorite beneath the district (Norris 1965). Proximal to the Pine Point District, the crystalline basement contains several important regional structures including Hay River Fault Zone, which comprises the Great Slave Lake Shear Zone and the MacDonald Fault Zone (Fig. 1; Hoffman 1987; Panǎ 2006). The crystalline Precambrian basement is overlain by a package of Middle Ordovician
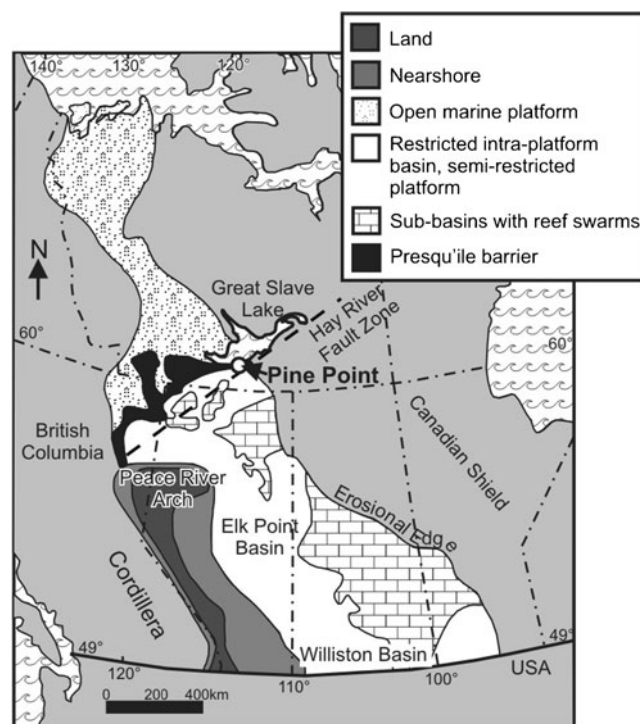


Fig. 1 Simplified geological map of the Western Canadian sedimentary basin after Qing and Mountjoy (1992). Pine Point is located at the present day erosional edge of the basin, close to the surface outcrop of the Precambrian Canadian Shield. The Hay River Fault Zone (HRFZ) comprises the Great Slave Lake Shear Zone and the Macdonald Fault Zone (Panǎ 2006)

or older, siliciclastic, evaporitic and carbonate rocks. The maximum thickness of these units is less than 100 m (Norris 1965). These units are unconformably overlain by Lower to Middle Devonian carbonates and sulphate-bearing evaporite units which underlie the Peace River Arch, which separates the Mackenzie open marine platform sediments to the north and the evaporite-bearing Elk Point restricted platform basin to the south (Fig. 1). The "Presqu'ile" Barrier is an elongate reef complex and hosts the Pine Point ore deposits (Krebs and Macqueen 1984; Rhodes et al. 1984; Qing 1998).

The mineralization and associated alteration is paragenetically complex and has been documented by Skall (1975), Kyle (1981), and Krebs and MacQueen (1984). The limestones of the Presqu'ile Barrier were dolomitized to form a pre-ore white matrix dolomite known as "Presqu'ile dolomite" (Skall 1975; Rhodes et al. 1984). The main stage of mineralization resulted in the precipitation of sulphides including galena, sphalerite, marcasite and pyrite in open spaces and replacing the host rocks within the Presqu'ile dolomites. Associated with these sulphides are ore-stage coarse nonsaddle and saddle dolomites, which also occur as vein and cavity fills and host rock replacements (Krebs and Macqueen 1984). At the N81 deposit, pyrobitumen, calcite and native sulphur post-date the main sulphide mineralization (Gromek

2006). Ore-stage sulphides have strongly positive $\delta^{34}$S values suggesting that most of the sulphur in the deposit was sourced from seawater sulphate (Sasaki and Krouse 1969).

The samples selected for this study include (1) CWD and (2) coarse white SD. The CWD is generally white or cream in hand sample. In thin section, this dolomite is anhedral and has an average grain size of approximately 0.5 mm. In transmitted light, the dolomite is opaque due to the presence of abundant, small (<3 μm) fluid inclusions and it has a dull red cathodoluminescence. Saddle dolomite formed where open space was available for development, and in this study, the CWD and SD are considered to form as a continuum. The SD crystals have cores of dark, inclusion-rich material and an outer rim of transparent, fluid inclusion-free dolomite. These rims are, on average, approximately 0.2 mm in width and have a bright orange cathodoluminescence. Both dolomite types are associated with ore at Pine Point and are equivalent to the "coarsely crystalline" and "saddle dolomite" of Qing and Mountjoy (1994) or the "coarse-grained dolomite" of Krebs and MacQueen (1984).

## Analytical technique

In situ Sr isotope analyses were carried out on six samples from two drill holes from the N81 deposit of the Pine Point District (Supplementary Material). Thin and thick sections (up to ~100 μm thick) were analysed on the basis of a detailed petrographic study (Gromek 2006). Samples chosen for this study were examined by electron microprobe for small mineral inclusions and were found to be mineral inclusion free.

In situ Sr isotope compositions were determined using a Nu plasma (MC-ICP-MS) instrument coupled to a 213-nm Nd: YAG laser system (New Wave Research) at the University of Alberta using an analytical protocol similar to that described by Schmidberger et al. (2003). The dolomite samples were ablated in spot and, more commonly, raster (line) mode using a spot size of 160 μm, a repetition rate of 10 Hz and an approximate 3 mJ laser output (see figure in Supplementary Material Online Resource 2). The laser ablation runs varied depending on the amount of time taken to ablate through the dolomite grains. Ion signals for each Sr isotope were measured using Faraday cups and recorded in volts; the total ion signal is a function of the total amount of Sr analysed and thus is a proxy for the Sr concentrations in the carbonates. Reproducibility and accuracy of the Sr isotope data were verified using a modern day coral (Indian Ocean) internal standard, which yielded an average $^{87}$Sr/$^{86}$Sr value of 0.70910±0.00005 (2$\sigma$, $n$>50; Bizzarro et al. 2003), identical to the TIMS value of 0.709098±0.000020 (Bizzarro et al. 2003). The $^{87}$Rb/$^{86}$Sr ratio of the samples was monitored to assess potential contamination by Rb-bearing detrital material present within the carbonates.

## Results

### Coarse white dolomite

Forty-three CWD $^{87}$Sr/$^{86}$Sr ratios were acquired and range from 0.7073 to 0.7152 (Supplementary Material). Two CWD samples from N81-65 0249′ had $^{87}$Sr/$^{86}$Sr ratios of 0.7088 and 0.7090 with relatively high 1/Sr ion signal (4.04 and 3.2 V, respectively). N81-98 3205 360′ CWD had an average $^{87}$Sr/$^{86}$Sr value of 0.7118 with minimum and maximum values of 0.7073 and 0.7152 and 1/Sr ion signal values ranging from 1.04 to 4.55 V. The $^{87}$Sr/$^{86}$Sr values from sample N81-98 3225 450′ ranged from 0.7086 to 0.7097, with an average value of 0.7090 and low 1/Sr ion signal values (<2.8 V; Supplementary Material). The $^{87}$Sr/$^{86}$Sr ratio of the CWD from N81-98 3208 367′ varied from 0.7129 to 0.7152 and had the most radiogenic average Sr isotope value (0.7142) of all the CWD samples with similarly low 1/Sr ion signal values ranging from 1.28 to 1.53 V (Fig. 2).

### Saddle dolomite

Analyses were carried out on the cloudy cores and three clear SD rims (Supplementary Material; Fig. 2). In total, the SD cores from N81-65 0249 are comparable with the CWD from this sample and range from 0.7075 to 0.7094. In sample N81-98 3205 360′, the cores of the SD crystals define a range in isotopic compositions from 0.7140 to 0.7150 and the SD rims have values that range from 0.7146 to 0.7152 (Supplementary Material).
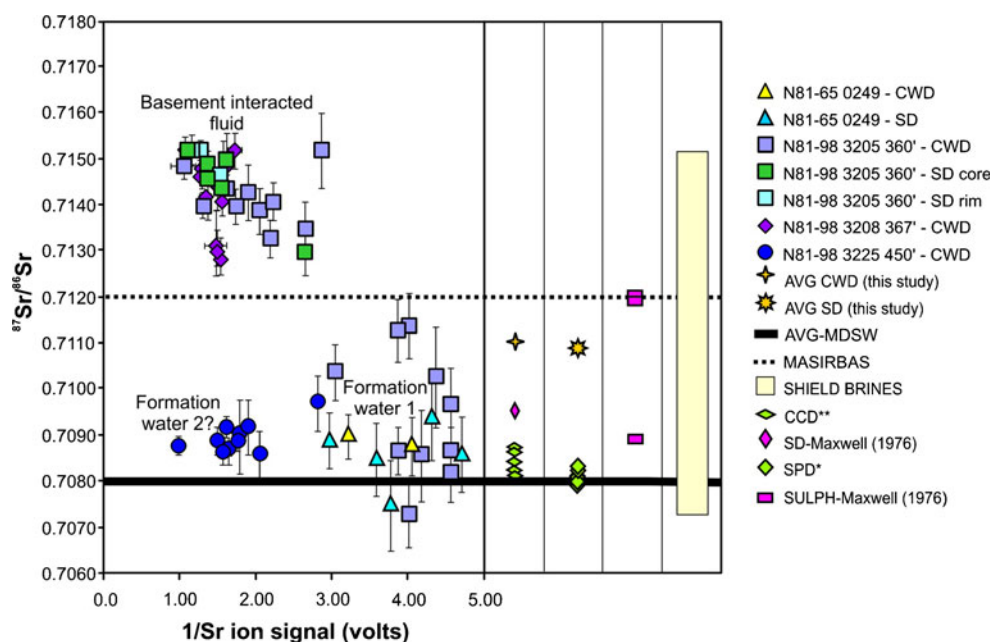
## Discussion

Three previous Sr isotope studies have been carried out at the Pine Point District. Maxwell (1976) and Medford et al. (1983) reported Sr isotopic ratios from sulphides and carbonates similar to that of Devonian seawater but also had some more elevated values (Fig. 2). A subsequent study by Mountjoy et al. (1992) reported Sr isotopic data from saddle dolomites in the range 0.7084 to 0.7083 and concluded that the higher $^{87}$Sr/$^{86}$Sr ratios of Medford et al. (1983) were the result of contamination of the analyses by radiogenic post-ore calcites.

This study has detected significant variations in the $^{87}$Sr/$^{86}$Sr ratios of the CWD and SD samples. In isotopic space, the $^{87}$Sr/$^{86}$Sr data broadly define two clusters (Fig. 2); one that ranges from 0.7065 to the MASIRBAS value of 0.7120 and a second that ranges from 0.7125 to 0.7160, suggesting that the ore-stage CWD and SD dolomites at Pine Point record two distinct Sr isotopic sources in the hydrothermal system.

Some of the samples with $^{87}$Sr/$^{86}$Sr ratios that range from 0.07070 to 0.7120 have a Sr isotopic composition similar to

that of Middle Devonian seawater. The spread of data from seawater compositions to the MASIRBAS value of 0.7120, however, suggests that this fluid may have interacted with clastic units in the basin. This is supported by a fluid inclusion study by Gleeson and Turner (2007), elsewhere in the district, that identified a Ca-Na-Cl-rich brine that was formed from partially evaporated seawater that had inter-acted with subsurface clastic units in the mineralizing sys-tem. It is possible that this fluid supplied the $H_2S$ generated from the thermochemical reduction of seawater at greater depths in the basin (Grasby et al. 2002). It is worth noting that sample N81-98 3225 450′ has a significantly lower 1/Sr ion signal, which suggests that the Sr concentration can be variable in samples with the same isotopic composition, which may suggest a third fluid in the system. Further work is required to confirm if this is the case.

The second group of data have Sr isotopic values ranging from 0.7120 to 0.7152; similar values have been found in saddle dolomites elsewhere in the WCSB (Davis and Smith 2006). These studies have suggested that the source of the radiogenic Sr is likely to be the clastic units or basement rocks below the altered carbonate sequences. The Sr isotopic com-position of the thin sequence of Lower Paleozoic sediments that underlie the Pine Point District is not known. Crystalline basement units in northern Alberta, approximately 100 km south of Pine Point, have $^{87}Sr/^{86}Sr$ ratios of 0.7030 and 0.7100 (Nielsen et al. 1981), and the Sr isotopic compositions of Rae Province gneisses, anorthosites and dykes to the east of Pine Point range from 0.70464 to 0.88318 (Crocker et al. 1993).

There are no experimental data from the WCSB on the extraction of Sr from crystalline basement rocks by basinal fluids. Low temperature brines hosted by the Canadian Shield units, however, may provide a natural analogue.

These shield brines have $^{87}Sr/^{86}Sr$ ratios that range from 0.706 to 0.755 and have a wide range in Sr compositions (Negrel and Casanova 2005). Bottomley et al. (1999) report analyses of shield brines from Yellowknife (130 km north of Pine Point) with Sr isotopic compositions that range from 0.70778 to 0.7148, similar to the range in compositions in the ore-stage dolomites at N81 (Fig. 2). Bottomley et al. (2005) suggested that these brines originated as evaporated Devonian seawater that migrated into the basement sequen-ces and acquired $^{87}Sr$ by interaction with the crystalline rocks. There are no published analyses of the metal content of the shield brines. However, the Pb isotopic signature of the ore at Pine Point (Cumming et al. 1990) is consistent with the derivation of the Pb from the underlying granite–granulite basement (Paradis et al. 2006). Furthermore, mod-elling of the Zn to Pb ratios of the ore led Hitchon (2006) to conclude that the mineralizing fluids had compositions that are significantly different from present day formation waters in the Devonian aquifers and that an external "geothermal" fluid formed the deposits. Our data, therefore, add further support to the hypothesis that there is a basement-derived component in the mineralizing system.

Pană (2006) presented a structural model whereby exten-sional structures in the basement formed subsidiary struc-tures during post-Middle Devonian shearing along the Hay River Fault Zone and propagated into the carbonate sequen-ces. The detailed kinematics of the model are not well con-strained but reactivation of older basement structures during deformation may have produced the fluid flow conduits necessary to transmit basement-interacted fluids through the evaporite sequences to the site of mineralization with minimal interaction, as observed by Davidson (2009) in salt sequences elsewhere. A component of vertical fluid flow at

Pine Point is supported by the patterns of metal zonation in some deposits (Kyle 1981) and may explain why some deposits (e.g. X15 and W17) were found below the Presqu'ile dolomite (Skall 1975; Kyle 1981). Kyle (1981) notes that the Pine Point District was found by exploration geologists who followed the MacDonald Fault (Hay River Fault Zone) from the Precambrian Shield along its projected strike under the Paleozoic cover. Thus, it may be no coincidence that the district developed at the edge of the basin, where the sedimentary sequence is relatively thin and the potential for interaction with basement-derived fluids was maximised along the fault zone. We suggest, therefore, that the N81 deposit is an example of a MVT deposit with some component of fluid flow across the basin–basement interface. Such fluid flow has been observed in other sediment-hosted deposits such as the Zn-Pb deposits of the Irish Midlands (Wilkinson et al. 2005) and some of the Proterozoic U deposits (Boiron et al. 2010).

## Conclusions

Small-scale Sr isotopic analyses of ore-stage dolomite crystals have identified variations in isotopic composition at the N81 deposit in the Pine Point District that are not elucidated with bulk analyses. Coarse white and saddle dolomites at N81 contain Sr that is interpreted to have been derived from at least two fluid sources. One fluid originated as middle Devonian seawater. The second fluid, which has a higher $^{87}Sr/^{86}Sr$ ratio, is interpreted to be a basement-modified fluid and was introduced into the mineralizing system along faults. This fluid could have supplied some of the Pb to the deposit. We suggest that the proximity of the carbonate units to the faulted crystalline basement, at the present day erosional edge of the basin, exerted a fundamental control on the location of the mineralization at the N81 deposit.

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