Provenance of metalliferous mudstones associated with the Lemarchant volcanogenic massive sulphide (VMS) deposit, central Newfoundland, Canada: Insights from Nd isotopes and lithogeochemistry

Stefanie Lode¹*, Stephen J. Piercey¹, Jonathan Cloutier²

¹Department of Earth Sciences, Memorial University, 300 Prince Philip Drive, St. John’s, NL, Canada, A1B 3X5

²Department of Earth Sciences, University of St. Andrews, St. Andrews, Scotland, UK, KY16 9AL

*Correspondence (slode@mun.ca)

Abstract: Neodymium isotope data on metalliferous mudstones (exhalites) and tuffs from the Cambrian Lemarchant VMS deposit provide insights into the tectonic environment of the Tally Pond volcanic belt, Canada. The Lemarchant samples have εNd513 = −6.0 to −1.8, whereas associated volcanic rocks have εNd513 of +0.4 to +1.4. The more evolved values in the exhalites have not been reported yet; however, they overlap the εNd of the underlying Ganderian Neoproterozoic Sandy Brook Group (εNd = −6.5 to −1.9), and Crippleback Intrusive Suite (εNd = −5.9 to −5.2). The evolved Nd isotopic signatures suggest that the Tally Pond volcanic belt was formed upon Ganderian arc basement, which itself was possibly built upon, or proximal to, the Gondwanan
Amazonian margin. Erosion of older crustal material and Tally Pond volcanic belt volcanic rocks, together with coeval eruption of the volcanic rocks, released Nd-rich detritus into the water column. Uptake of eroded detrital and scavenged Nd resulted in mixed Nd sources (juvenile and evolved), archived in the exhalites. The results of this study are of significance not only for occurrences of exhalites within the Tally Pond volcanic belt, but also have exploration implications for VMS districts globally.

Metalliferous mudstones (exhalites) are abundant in the Tally Pond volcanic belt, central Newfoundland Appalachians, Canada, and are locally genetically associated with volcanogenic massive sulphide (VMS) deposits (Swinden 1991; Squires & Moore 2004). The Tally Pond volcanic belt, which is part of the Dunnage Zone, Newfoundland, Canada, belongs to the Cambrian (~515 Ma) to Permian (~275 Ma) Appalachian-Caledonide mountain belt that hosts numerous VMS deposits, including the past-producing Duck Pond and Boundary mines, and the precious metal-bearing Lemarchant deposit (Fig. 1A-B; Williams 1979; Swinden 1988, 1991; Evans & Kean 2002; Grenne & Slack 2003; Rogers et al. 2007; van Staal &Barr 2011; Piercey et al. 2014; Hollis et al. 2015). The Tally Pond group (~513-509 Ma) volcanic rocks and related massive sulphide mineralization formed during arc rifting during the construction of the Cambrian to Early Ordovician Penobscot Arc, which is known to be built upon Ganderian Neoproterozoic (~563 Ma) arc basement of the Crippleback Intrusive Suite and the coeval Sandy Brook Group (Pollock et al. 2002; Zagorevski et al. 2007; Zagorevski et al. 2010; Piercey et al. 2014).
In the Neoproterozoic and Early Cambrian Ganderia was located north-west of the Gondwanan Amazonian margin (Fyffe et al. 2009; van Staal et al. 2012; Murphy et al. 2014). The Penobscot arc represented the leading edge of Ganderia in a supra-subduction zone setting and arc rifting was initiated due to slab roll-back along this margin (Jenner & Swinden 1993; Schulz et al. 2008; Murphy et al. 2014). The basement to the Ganderian arc is not exposed; however, detrital zircon and Nd isotopic studies indicate the presence of older crustal rocks that were derived from the Gondwanan Amazonian craton (Nance et al. 2008; Schulz et al. 2008). Rifting of the Penobscot Arc led to the formation of volcanogenic massive sulphide (VMS) mineralization and associated hydrothermal sedimentary rocks of the Tally Pond group (Rogers et al. 2006; Copeland et al. 2009; Zagorevski et al. 2010; Piercey et al. 2014; Lode et al. 2016).

During rifting of the Penobscot arc there was extension, massive sulfide formation, and the genesis of hydrothermal sedimentary rocks that formed from the deposition from buoyant hydrothermal plumes from black smokers (Hekinian et al. 1993; Hannington et al. 1995; German & Von Damm 2003). These black smokers and associated hydrothermal sedimentary rocks occur where hydrothermal fluids are focused along deep synvolcanic faults in extensional settings (e.g., ocean ridges, rifted arcs, or backarc basin spreading centres) (Fig. 2; Lydon 1984; Hannington et al. 2005; Gibson et al. 2007). The hydrothermal fluids consist of modified seawater, which is entrained through oceanic or rift-related continental crust, and are variably metal bearing with Fe, Mn, Cu, Pb, and Zn, as well as reduced sulphur and Si (Von Damm 1990; German & Von Damm 2003; Galley et al. 2007; Tivey 2007; Huston et al. 2010). The metals and other ligands are generally derived from seawater (S) and leached from host rocks (e.g., metals, Si±S) (Fig.
Hydrothermal plume-derived Fe-oxyhydroxides are efficient scavengers of trace metals (e.g., oxyanions such as HPO$_4^{2-}$, HVO$_4^{2-}$, CrO$_4^{2-}$, HAsO$_4^{2-}$) and rare earth elements (REE) plus Y from seawater (Mills & Elderfield 1995; Rudnicki 1995). A rifted arc environment exposes rock units of different ages, hence varying Nd isotopic signatures, which contribute detrital material to the hydrothermal matter in the exhalative sedimentary rocks due to erosional and weathering processes (Keto & Jacobson 1988; Mills & Elderfield 1995). Therefore, exhalites not only record seawater REE (including Nd) but also the provenance components of the detrital sources at the time of formation (Mills & Elderfield 1995; Peter 2003; Lode et al. 2015).

By using various isotopic tracers, such as Nd isotopes, it is possible to decipher the potential sources of various components in hydrothermal sedimentary rocks. The Nd isotopic system is specifically useful for understanding the relative roles of evolved versus juvenile crust, and provides further insight into the tectonic environment and provenance of the metalliferous mudstones, as it is robust and not significantly modified by diagenetic, hydrothermal, and metamorphic processes (McCulloch & Wasserburg 1978; McLennan et al. 2003). In addition, the separation of Sm-Nd in Earth’s reservoirs is particularly useful in delineating juvenile versus evolved crust and the time-integrated sources of materials in Earth materials (McCulloch & Wasserburg 1978; Rollinson 1993; McLennan et al. 2003). The Tally Pond belt volcanic rocks have εNd signatures that are typically positive, whereas their basement rocks, i.e., the rifted arc rocks of the Neoproterozoic Crippleback Intrusive Suite and the Sandy Brook Group show more evolved εNd values (McLennan et al. 1993; Rogers et al. 2006; Nance et al. 2008;
McNicoll et al. 2010; Piercey et al. 2014). Given the level of preservation of stratigraphy of the lithofacies in the Lemarchant deposit, including the metalliferous mudstones, this deposit is an excellent location to understand the provenance of hydrothermal mudstones in ancient rifted arcs. Correspondingly, the Nd isotopic signatures in the mudstones may be useful in outlining their provenance and the potential contributions of local versus basement versus distal sources in their genesis.

The purpose of this study is to: 1) determine the sources of Nd in the metalliferous mudstones and massive sulphides of the Lemarchant deposit; and 2) because the Tally Pond volcanic belt is formed upon Ganderian and possibly older basement rocks, to evaluate the relative roles of mantle and evolved crustal inputs that contributed to the Lemarchant hydrothermal sedimentary rocks using the Nd isotope compositions of metalliferous mudstones.

**Regional Geology**

The Tally Pond volcanic belt is located within the Central Mobile Belt, Newfoundland, Canada, which is part of the Cambrian (~515 Ma) to Permian (~275 Ma) Appalachian mountain belt (Williams 1979; Swinden 1988; Rogers et al. 2007; van Staal & Barr 2011). The Newfoundland Appalachians are divided into four tectonostratigraphical zones (from west to east): Humber, Dunnage, Gander and Avalon zones (Fig. 1A; Williams 1979; Swinden 1988, 1991). The Dunnage Zone represents the Central Mobile Belt (Williams et al. 1988; Swinden 1991; Rogers et al. 2007). These zones result from and were affected by the successive accretion of three micro-continental blocks during the Early Palaeozoic to Middle Palaeozoic (i.e., Dashwoods, Taconic orogenesis;
Ganderia, Salinic orogenesis; and Avalonia, Acadian orogenesis) and related interoceanic arcs and backarcs (Swinden 1991; Zagorevski et al. 2010). In the Palaeozoic (Middle Cambrian to Ordovician), these ribbon-shaped micro-continental blocks separated from Gondwana and Laurentia, forming peri-Gondwanan and peri-Laurentian terranes and subsequently accreted to Laurentia creating the composite Laurentian margin (Rogers et al. 2007; Zagorevski et al. 2010; van Staal & Barr 2011). The Exploits Subzone represents two phases of arc-backarc formation, the Cambrian to Early Ordovician Penobscot Arc and the Early to Middle Ordovician Victoria Arc (Zagorevski et al. 2010). The Tally Pond volcanic belt and its VMS deposits (Duck Pond and Boundary mines; Lemarchant deposit; Fig. 1B) are hosted in the lower Victoria Lake supergroup within the Exploits Subzone, which is comprised of Cambrian to Ordovician volcanic and sedimentary rocks (Dunning et al. 1991; Rogers et al. 2007; McNicoll et al. 2010; van Staal & Barr 2011). The Victoria Lake supergroup is further subdivided into six assemblages (Zagorevski et al. 2010; Piercey et al. 2014), which are bounded by faults, and are from east to west: 1) the Tally Pond group; 2) the Long Lake group; 3) the Tulks group; 4) the Sutherlands Pond group; 5) the Pats Pond group; and 6) the Wigwam Pond group; the Tulks, Long Lake, and Tally Pond groups are known to host VMS deposits. These six tectonic assemblages yield U-Pb zircon ages ranging from ~513 to 453 Ma (Dunning et al. 1987; Evans et al. 1990; Dunning et al. 1991; Evans & Kean 2002; Zagorevski et al. 2007; McNicoll et al. 2010). Furthermore, the Tally Pond group is informally subdivided into the felsic volcanic rock dominated Bindons Pond formation (also referred to as Boundary Brook formation) and the mafic volcanic rock dominated Lake Ambrose formation (Rogers et al. 2006). The latter contains island arc tholeiitic
basalts to andesites with $\varepsilon$Nd$_{511}$ of +3.1 (Dunning et al. 1991; Evans & Kean 2002; Rogers et al. 2006), whereas the former contains predominantly transitional to calc-alkalic rhyolitic to dacitic rocks with $\varepsilon$Nd$_{511}$ of +1.8 to +2.6 (Rogers et al. 2006; Piercey et al. 2014). The Cambrian felsic volcanic rocks of the Bindons Pond formation contain inherited zircons with Neoproterozoic U-Pb ages of 563 Ma (McNicoll et al. 2010).

**Deposit Geology and Lithofacies**

The Lemarchant VMS deposit is hosted within the Bindons Pond formation and is capped by a <1 to 20 m thick layer of metalliferous mudstones occurring at the contact between the Bindons Pond and Lake Ambrose formations (Fig. 3A; Copeland et al. 2009; Fraser et al. 2012; Lode et al. 2015). These sulphide-rich metalliferous mudstones extend discontinuously around the massive sulphides for one to four kilometres (Copeland et al. 2009; Fraser et al. 2012; Lode et al. 2015). Three main types of exhalative mudstones occur at the Lemarchant deposit: 1) mudstones immediately on top of massive sulphide mineralization between the felsic and mafic volcanic rocks (exhalative mudstone-massive sulphide (EMS)-type; Fig. 3A-C, G-H); 2) mudstones extending laterally outwards from mineralization, but at the same stratigraphical level and without immediate association with mineralization (felsic-exhalative mudstone-mafic (FEM)-type; Fig. 3D); or 3) interflow mudstones within the hanging wall basaltic rocks (interflow exhalative mudstone (IFE)-type; Fig. 3E). Interflow mudstones occur commonly within 15 meters above the massive sulphide mineralization, but are present up to 70 meters above the ore horizon. Independent of their stratigraphical positions, the mudstones are brown to black, graphite-rich, finely laminated, and contain fine carbonaceous/organic-rich laminae that
are intercalated with siliciclastic, volcaniclastic and/or amorphous kidney-shaped chert±apatite layers (Fig. 3C, F). The main sulphide phases are pyrite (framboidal, massive and euhedral) and pyrrhotite, with minor marcasite, chalcopyrite, sphalerite, arsenopyrite and galena. Contents of chalcopyrite, sphalerite, and galena increase proximal to mineralization. The sulphides occur both parallel to bedding, and in later stage, stringer-like veins (Fig. 3A, D-E).

Methodology

Sampling, methods, and quality control and quality assurance (QA/QC)

Samples were collected during stratigraphical mapping and drill core logging of the Lemarchant deposit from drill holes that have metalliferous mudstones and include the Lemarchant Main Zone, the Northwest and 24 zones, as well as the North and South targets (Fig. 4A). Samples were taken from representative exhalative mudstone types (EMS, FEM, and IFE), tuff, and surrounding lithological mafic and felsic volcanic units. The whole rock lithogeochemical data were previously evaluated and presented in Lode et al. (2015), including analytical methods and QA/QC for lithogeochemical data. Lithogeochemical data are reproduced here only to compare to Nd isotope results.

Neodymium isotopes

Twelve representative samples of the Lemarchant mudstones were selected for Nd isotopic determinations from the three mudstone types and tuffs that are intercalated with the mudstones. These samples were chosen to cover both the horizontal and vertical distributions of all mudstones types and tuff occurring in the Lemarchant area. Additionally, one least altered sample of the felsic and mafic volcanic rocks (Fig. 3G-H)
were selected for analyses, and for comparison to mudstone samples. Samarium and Nd isotopic compositions were measured at Memorial University using a multicollector Finnigan MAT 262 thermal ionization mass spectrometer (TIMS) in static and dynamic acquiring modes. Samples for Nd analyses were prepared using the methods of Fisher et al. (2011) from whole-rock powders using a multi-acid (HF, HNO₃, and HCl) dissolution-evaporation process following methods. Separation of Sm and Nd was obtained using conventional two-step column chemical methods (Fisher et al. 2011).

Accuracy and precision for the Nd analyses were determined using the standards JNdi-1 and BCR-2 as reference materials following methods described in Fisher et al. (2011).

The JNdi-1 and BCR-2 standards have following reported values: ¹⁴³Nd/¹⁴⁴Nd = 0.512115 and ¹⁴³Nd/¹⁴⁴Nd = 0.512633, respectively (Tanaka et al. 2000; Raczek et al. 2003). Standards were run every 11 samples with each analytical batch. Additionally, blanks were utilized during each analytical run to test contamination; none was detected.

Precision was determined using the percent relative standard deviation (%RSD) on the replicate analyses of the reference materials, and accuracy was determined using percent relative difference (%RD) from accepted values. Analyses for the Lemarchant samples have an average 0.0013 %RSD for ¹⁴³Nd/¹⁴⁴Nd and 0.00055 %RD for ¹⁴³Nd/¹⁴⁴Nd.

The results herein are presented using the epsilon notation (εNd) and calculated for a formation age of 513 Ma, the U-Pb age of the host stratigraphy as reported by Dunning et al. (1991); data are presented in Table 1 and Figures 4B, 5A-B, and Figure 8. εNd₅₁₃ was calculated by εNdₜ = (¹⁴³Nd/¹⁴⁴Nd rock,t/¹⁴³Nd/¹⁴⁴Nd CHUR,t) × 10⁴ after Rollinson (1993) and fSm/Nd = [(¹⁴⁷Sm/¹⁴⁴Nd sample,t)/(¹⁴⁷Sm/¹⁴⁴Nd CHUR,t) - 1] after McLennan et al. (1990).

Chondrite uniform reservoir (CHUR) values utilized in this study are ¹⁴³Nd/¹⁴⁴Nd of
0.512638 and a $^{147}\text{Sm}/^{144}\text{Nd}$ of 0.1967 (Hamilton et al. 1983; Rollinson 1993). Depleted mantel model ages ($T_{DM}$) were calculated using depleted mantle values of $^{144}\text{Nd}/^{144}\text{Nd} = 0.513163$ and $^{147}\text{Sm}/^{144}\text{Nd} = 0.2137$, and a decay constant of $\lambda = 6.54 \times 10^{-12}$ (DePaolo 1981).

**Results**

*Neodymium isotopic systematics.* The Lemarchant mudstones ($n = 10$), have $\varepsilon_{Nd} = -6.0$ to $-1.8$ and $T_{DM} = 1.63$ to 3.05 Ga (Table 1). Overall, the three types of Lemarchant mudstones (EMS = proximal; FEM = distal; IFE = interflow) have similar $\varepsilon_{Nd}$ values; however, the EMS-type have slightly lower $\varepsilon_{Nd}$ values and range from $-5.6$ to $-4.1$ with an average of $-4.8$, the FEM-type are less evolved and range from $\varepsilon_{Nd} = -4.0$ to $-3.2$ with an average of $-3.7$, and the IFE-type has the widest range of $\varepsilon_{Nd} = -6.0$ to $-1.8$ and average of $-3.9$ (Table 1; Fig. 4B, 5A-B). The Lemarchant tuff samples ($n = 2$) have $\varepsilon_{Nd} = -5.7$ to $-4.7$ with an average of $-5.2$ and $T_{DM} = 1.75$ to 1.81 Ga. In $\varepsilon_{Nd}$ versus Th/Sc space the Lemarchant mudstones and tuff have Th/Sc ratios of 0.06 to 1.93 and fall between the arc andesite fields, with samples that have greater Th/Sc containing lower $\varepsilon_{Nd}$ values like the upper crust (Fig. 5A). These more evolved samples also trend towards the field of the 563 Ma Crippleback Intrusive Suite and Sandy Brook Group crustal basement rocks (recalculated here at 513 Ma for comparison; Fig. 5A). The Lemarchant felsic and mafic volcanic rock measured in this study (one each) have $\varepsilon_{Nd} = +0.4$ and a $T_{DM} = 1.47$ Ga, and $\varepsilon_{Nd} = +1.4$ and a $T_{DM} = 1.74$ Ga, respectively, and plot in the field for arc rocks (Table 1; Fig. 5B). These values for the Lemarchant volcanic rocks are similar to values reported by Rogers et al. (2006) and McNicoll et al. (2010) for felsic and mafic volcanic rocks of the Tally Pond volcanic belt,
including samples from the ‘Upper Block’ and the ‘Mineralized Block’ of the Duck Pond deposit (Fig. 5B).

The $f^{\text{Sm/Nd}}$ reflects the fractional deviation of $^{147}\text{Sm}/^{144}\text{Nd}$ from CHUR in parts per $10^4$ because of light rare earth element enrichment (i.e., lower Sm/Nd) during igneous differentiation processes (McLennan et al. 2003). Accordingly, in $f^{\text{Sm/Nd}}$-$\varepsilon\text{Nd}$ space (Fig. 5B) the Lemarchant mudstone and tuff samples have more evolved $\varepsilon\text{Nd}_{513}$ values than the Lemarchant volcanic rocks, and are overall comparable in $\varepsilon\text{Nd}$ to values reported by Rogers et al. (2006) for the Neoproterozoic Crippleback quartz monzonite and Sandy Brook Group rhyolite. However, the Lemarchant mudstone and tuff samples have $f^{\text{Sm/Nd}}$ higher than the Neoproterozoic Crippleback quartz-monzonite and Sandy Brook Group rhyolite and trend towards those of the Tally Pond group volcanic rocks (Fig. 5B; McLennan et al. 2003). The $\varepsilon\text{Nd}$ values of the Lemarchant mudstone and tuff samples do not show any spatial variations throughout the zones of the deposit and/or with depth in the stratigraphy in the Lemarchant area (Fig. 4A-B). The $T_{DM} = 1.63$ to 3.05 Ga of the Lemarchant mudstones are older than reported values for the coeval felsic volcanic rocks of the ‘Upper Block’ and ‘Mineralized Block’ at Duck Pond of 1.06 and 1.35 Ga, and 0.95 Ga, respectively (McNicoll et al. 2010), and those of the Crippleback Intrusive Suite (1.26 and 1.35 Ga) and the Sandy Brook Group (1.15 to 1.34 Ga) (Rogers et al. 2006; this study).

**Immobile element systematics**: Volcanic rocks of the Tally Pond group that are associated with the hydrothermal sedimentary rocks and volcanic and igneous rocks of the Sandy Brook Group and Crippleback Intrusive Suites are shown on the immobile element Zr/Ti-Nb/Y classification diagram by Winchester and Floyd (1977) and Pearce...
The volcanic rocks from the Lemarchant deposit are subalkaline basaltic andesites, with the more felsic rocks trending towards dacite boundary, and the more mafic rocks trending towards the basalt boundary (Fig. 6A). Because of the limited sample number for volcanic rocks from this study, fields from Cloutier et al. (in press) were added for felsic, intermediate, and mafic volcanic rocks from the Lemarchant deposit (Fig. 6A). Additionally, samples for Tally Pond belt felsic and mafic volcanic rocks, the Sandy Brook Group rhyolite and basalt and Crippleback quartz monzonite of Rogers (2004) and Rogers et al (2006) were also added for comparison. Chemically, the volcanic rocks of Lemarchant show a wide distribution, with felsic-dominated rhyolite-dacites of the Bindons Pond formation as well as intermediate andesite-basaltic andesites and mafic rocks of the Lake Ambrose formation (Cloutier et al. in press). The alteration-independent Nb-Y diagram after Pearce et al. (1984), which discriminates the general tectonic settings, shows that the Tally Pond belt volcanic rocks plot predominantly in the volcanic arc field (Fig. 6B), which is consistent with potential source rocks for the detrital constituent in the hydrothermal sedimentary rocks and regional models for the Tally Pond group (e.g., Rogers et al. 2007; Piercey et al. 2014).

**Discussion**

*Provenance, tectonic setting, and the role of crustal input*

The Tally Pond volcanic belt represents the oldest magmatism of the Penobscot Arc and was developed during phases of arc rifting at the leading edge of the Ganderian margin.
Penecontemporaneously, further rifting on the trailing edge of Ganderia, led to the formation of the Ellsworth belt (~509-505 Ma) of coastal Maine and New Brunswick representing the separation of Ganderia from the Gondwanan Amazonian margin (Fyffe et al. 2009; van Staal et al. 2012). The volcanic rocks of the Ellsworth terrane comprise tholeiitic basalts and rhyolites with $\varepsilon_{Nd}^{500}$ values ranging from +5.6 to +8.6, but also calc-alkaline rhyolite (R-1 Rhyolite) that yielded $\varepsilon_{Nd}^{500}$ values near zero (Schulz et al. 2008). The latter are similar to $\varepsilon_{Nd}$ values of felsic and mafic volcanic rock samples from the Tally Pond belt (Bindons Pond and Lake Ambrose formations) of this study ($\varepsilon_{Nd} = +1.4$ and +0.4, respectively), which are comparable with values that were previously reported for the Tally Pond volcanic rocks (Fig. 5B; Rogers et al. 2006; Zagorevski et al. 2010). This illustrates that the Lake Ambrose formation basalts have predominantly juvenile signatures ($\varepsilon_{Nd}^{511} = +3$; Rogers et al. 2006 and this study), whereas Bindons Pond formation rhyolites and dacites have less juvenile values ($\varepsilon_{Nd}^{511} = +1.8$ and +2.6) (Rogers et al. 2006; Zagorevski et al. 2010). There is a noticeable difference in $\varepsilon_{Nd}^{513}$ values between the sedimentary and volcanic rocks of the Lemarchant deposit, however. In general, the mudstones and tuffs have lower $\varepsilon_{Nd}^{513}$ values ranging from −6.0 to −1.8 (Fig. 5A-B), similar to the Sandy Brook Group rhyolite $\varepsilon_{Nd}^{513} = −6.5$ to −1.9, as well as those of the Crippleback Intrusive Suite $\varepsilon_{Nd}^{513} = −5.9$ to −5.2 (Rogers et al. 2006). Moreover, the Lemarchant mudstones have similar $\varepsilon_{Nd}^{513}$ values throughout the sections of the Lemarchant Main Zone, the Northwest and 24 zones, and the North Target (Fig. 4A-B), albeit proximal Lemarchant mudstones (EMS-type) have more evolved $\varepsilon_{Nd}^{513}$ values than the more distal mudstones (FEM-type; Figs. 4A-B).
There are a number of potential Nd sources in hydrothermal mudstones, including seawater-derived/scavenged, detrital, and hydrothermally-derived components (Goldstein et al. 1984; Mills et al. 1993; Mills & Elderfield 1995). Scavenging of REE from seawater occurs during mixing of the hydrothermal fluids with seawater, where oxyanions (HPO$_4^{2-}$, HVO$_4^{2-}$, CrO$_4^{2-}$, HAsO$_4^{2-}$), trace elements, and rare earth elements (REE, including Nd) are scavenged from seawater onto Fe-oxyhydroxides, and subsequently deposited on the mount flanks and in topographic depressions around the hydrothermal vent site in a rift-graben or caldera basin (de Baar et al. 1988; Rudnicki 1995; German & Von Damm 2003; Peter 2003). Nd isotopic signatures measured from modern seawater show a wide range that indicate that continental Nd is the predominant source of REE in modern seawater resulting in different Nd values within the main water masses/oceans (Goldstein et al. 1984; Bertram & Elderfield 1993; Tachikawa et al. 2003). Thus, exposure of crustal basement during arc rifting would bring crustal-derived evolved Nd into the ambient seawater, together with Nd derived from the continuously erupting and erosion of the more juvenile Cambrian Tally Pond volcanic rocks. The Nd is contributed to the Nd budget of hydrothermal sediment either dissolved or as detrital particles, and via adsorption onto hydrothermally-derived particles, such as Fe-oxyhydroxides (Wood & Williams-Jones 1994; Mills & Elderfield 1995; Rudnicki 1995; Chavagnac et al. 2005). In contrast, hydrothermal Nd is a minimal component in hydrothermal sediment, mostly because REE are in extremely low concentrations in seafloor hydrothermal fluids and initial hydrothermal Nd signatures in the hydrothermal sediment are often rapidly overprinted by Nd scavenged from seawater (Elderfield 1988; Mills et al. 1993; Mills & Elderfield 1995). Most hydrothermal sediment contains up to
~80% hydrothermally-derived matter (i.e., derived directly from the fluid and scavenged from seawater), whereas the remainder consists of sedimentary detritus, including volcaniclastic and epiclastic material and windblown dust, which are also possible Nd sources in metalliferous sediments (Boström et al. 1972; Boström 1973; Cavanagh et al. 2005; Goldstein et al. 1984).

Considering these processes and potential Nd sources, it is noticeable that even though the Lemarchant hydrothermal sediments predominantly consist of hydrothermally-derived matter (e.g., Zn-Pb-Cu-Fe-S), their Nd budget contains only minor hydrothermally-derived Nd. The dilution of hydrothermal fluids by seawater, scavenging processes, and contributions of detrital matter generally annihilates the initial hydrothermal Nd signatures in hydrothermal sediments (Mills & Elderfield 1995).

Because of the restricted nature of a rifted arc basin, where predominantly locally occurring provenance rocks (i.e., Tally Pond volcanic rocks, Crippleback Intrusive Suite and Sandy Brook Group basement rocks) are eroded, this locally-derived detrital Nd is subsequently archived in the hydrothermal sedimentary rocks within the basin (Figs. 7A-B, 8). The Nd in these hydrothermal sediments was derived predominantly from scavenging and detrital matter, which explains their evolved Nd signatures; signatures that are not present in the more juvenile Tally Pond volcanic rocks. Furthermore, it is suggested that the more evolved Nd signatures of the proximal mudstones represent early stages of arc-rifting, which were dominated by erosion of the rifted Neoproterozoic Ganderian and possibly older crustal basement, whereas the more distal mudstones reflect greater contributions from the continuously erupting and erosion of the more juvenile Cambrian Tally Pond group volcanic rocks (Fig. 7A-B).
The presence of Neoproterozoic Ganderian arc rocks (Crippleback Intrusive Suite and coeval Sandy Brook Group) beneath the Tally Pond volcanic belt is indicated by inherited zircons (563 Ma) in the Cambrian felsic volcanic rocks of the Tally Pond belt (Rogers et al. 2006; Rogers et al. 2007; McNicoll et al. 2010; Zagorevski et al. 2010).

Neoproterozoic (~553 Ma) inherited zircon ages are also known from the Pats Pond group (~487 Ma), which also have Mesoproterozoic (0.9-1.2 Ga) xenocrystic zircons (Zagorevski et al. 2007, Zagorevski et al. 2010). Plutonic and gneissic boulders, as well as sedimentary rocks from the Ellsworth Formation of coastal Maine and New Brunswick contained small populations of Mesoproterozoic, Palaeoproterozoic, and Archean zircons up to 3.23 Ga, but with a dominant population between 1.07 to 1.61 Ga (Hibbard et al. 2007; Schulz et al. 2008; Fyffe et al. 2009; van Staal et al. 2012). These inherited zircon patterns are consistent for a location of Ganderia along the Gondwanan Amazonian margin (Fyffe et al. 2009; van Staal et al. 2012). The Mesoproterozoic to Archean $T_{DM}$ model ages and Nd isotopic data of the Lemarchant mudstones (1.63 to 3.05 Ga) together with the detrital zircon populations and the Nd signatures of the Tally Pond volcanic rocks, as well as of the Crippleback Intrusive Suite and Sandy Brook Group, indicate an Amazonian provenance, which suggests that the Tally Pond belt also evolved along this margin (Fig. 8; Zagorevski et al. 2007; Pollock et al. 2011; van Staal & Barr 2011; van Staal et al. 2012).

Significant input from crustal material is further supported by the Pb isotopic data of the Lemarchant deposit and other massive sulphide occurrences in the Tally Pond belt (Swinden & Thorpe 1984; Pollock & Wilton 2001; Gill 2015; Lode et al. 2017).

Volcanogenic massive sulphides and associated hydrothermal sediments have Pb sources
that derive their Pb predominantly from leaching of basement rocks, which may include
different reservoirs (Franklin et al. 1981; Swinden & Thorpe 1984; Tosdal et al. 1999;
Ayuso et al. 2003). Lead isotopic data measured in-situ on galena hosted within sulphides
in the hydrothermal sediments using secondary ion mass spectrometry (SIMS), suggested
hydrothermally- and detritally-derived Pb sources (Lode et al. 2017). Especially more
vent distal mudstones showed more radiogenic detritally Pb contributions, which were
characterised by more radiogenic $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios (Mills & Elderfield

Altogether, the Nd and Pb isotopic data support that older crustal basement plays a role in
hydrothermal activity, either through direct leaching (Pb), detrital (Pb+Nd), or via
adsorption/deposition from the water column (Nd). Furthermore, trace element signatures
of the Tally Pond volcanic rocks and provenance-related immobile element systematics
of the metalliferous mudstones are consistent with a formation in a volcanic arc
environment, such as a graben/caldera in a rifted continental arc, or an arc proximal to
continental crust (Rogers et al. 2006; Zagorevski et al. 2010; Piercey et al. 2014).

Therefore, the metalliferous mudstones that precipitate in a rifted arc basin/caldera setting
record the provenance components of the lithologies present in the basin and may be
useful for palaeogeographic reconstructions and additionally provide means to determine
the source of metals that contributed to the formation of the genetically associated
massive sulphides.

**Conclusions**
It is proposed that the volcanogenic massive sulphides of the Lemarchant deposit and related exhalative metalliferous mudstones that are associated with felsic, intermediate, and mafic volcanic rocks are formed from fluids that ascend along deep synvolcanic faults in a rifted arc basin. Eruption and erosion of the Tally Pond belt volcanic rocks that were formed in this rift-related graben/caldera setting, added juvenile Nd to the system. The hydrothermal sediments precipitate in an environment, where Ganderian arc rocks of the Crippleback Intrusive Suite and the coeval Sandy Brook Group was exposed and eroded, which contributed evolved crustal Nd to the ambient seawater. Based on detrital zircon and Nd isotopic studies it is further suggested that unexposed older crustal basement of the Gondwanan Amazonian margin beneath the Ganderian arc rocks additionally contributed evolved Nd to the Nd budget in the metalliferous mudstones. The precipitating hydrothermal metalliferous mudstones record the mixed, i.e., evolved and juvenile, εNd signatures. Furthermore, the hydrothermal sediments have more evolved εNd systematics than the spatially associated Tally Pond belt volcanic rocks indicating the presence of eroded older crustal material that did not significantly contribute to the volcanic rocks. Overall, the Nd isotopic compositions, as well as the lithogeochemical data, of the Lemarchant metalliferous mudstones and suggests that the Lemarchant deposit hydrothermal mudstones record formation within a rifted arc environment built upon Ganderian (exposed) and Gondwanan Amazonian (unexposed) crustal basement, consistent with existing models for the Tally Pond group.

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**References**


**Figure Captions**

**Fig. 1.** (a) Tectonostratigraphical assemblages with the main zones of the Newfoundland Appalachians (Avalon, Gander, Dunnage, and Humber zones) and VMS occurrences within the Notre Dame and Exploits subzones.


Exploits Subzone VMS: 30 – 37 - Tulks Belt Deposits; Tally Pond Belt Deposits: 39 –

Lemarchant; 40 – Duck Pond; 41 – Boundary; 42 – 45 – Point Leamington Belt Deposits.

Modified after (Swinden, 1991) and Piercey (2007). (b) Geological map of the Tally Pond volcanic belt. The Tally Pond group comprises the Lemarchant deposit and the Duck Pond and Boundary mines. Figure after Copeland (2009) and Map 2006-01 from Squires and Hinchey (2006) and Lode et al. (2017).

**Fig. 2.** Schematic illustration of the main aspects of hydrothermal circulation in extensional tectonic environments. In the recharge zone seawater is entrained through crustal and progressively heated during downward migration. Water-rock interactions lead to loss of Mg$^{2+}$, SO$_4^{2-}$, and OH$^-$ and H$_2$S is generated. These reactions produce H$^+$ and create acidic fluids that leach metals out of rocks. In the reaction zone the highest temperatures are reached and the hydrothermal fluids gain their geochemical signatures. The hot fluids rise buoyantly up along synvolcanic faults and are expelled via hydrothermal plume into the ambient seawater. Figure modified after German and Von Damm (2003) and Gibson et al. (2007).
Fig. 3. Core photographs of the main Lemarchant mudstone types, tuff, and associated felsic and mafic volcanic rocks of the Bindons Pond and Lake Ambrose formations, respectively. (a) Finely laminated sulphide-rich EMS-type metalliferous mudstone with cross-cutting stringer type veins and overlying massive sulphide mineralization. Section 101N, LM11-65, mudstone sample CNF30983, 160.7 m. (b) Proximal EMS-type metalliferous mudstone associated with the Lemarchant Main Zone. Section 102+50N, LM10-43, CNF20976, 202.3 m. (c) Proximal EMS-type metalliferous mudstone with intercalated chert-apatite layers. Section 101N, LM07-13, CNF30954, 164.7 m. (d) FEM-type mudstone associated with the Northwest Zone. Section 106N, LM08-28, CNF20986, 240.6 m. (e) Sulphide-rich exhalative interflow mudstone. Section 101+25N, LM13-79, CNF25072, 169.0 m. (f) Lithic crystal vitric tuff that is intercalated with FEM-type mudstone. Section 100+50N, LM13-77, CNF25065, 140.0 m. (g) Felsic to intermediate volcanic rock of the Bindons Pond formation located in the North target. Section 108N, LM11-49, 144.6 m. (h) Mafic to intermediate volcanic rock of the Lake Ambrose formation located in the North target. Section 108N, LM11-49, 422.9 m.

Fig. 4. (a) Spatial distribution of $\varepsilon$Nd for the EMS-, FEM-, and IFE-type mudstones and tuff, as well as the Lemarchant felsic and mafic volcanic rock from this study. Sample data do not show any spatial variations throughout the sections and/or with depth in the stratigraphy in the Lemarchant area. 2σ error bars calculated after algorithm from Ickert (2013). (b) Resource map of the massive sulphides of the Lemarchant Main, 24 Zone, and Northwest Zone. Massive sulphides are projected to the surface. Modified from the resource map of Canadian Zinc Corporation.
Fig. 5. (a) Diagram of $\varepsilon$Nd versus Th/Sc ratio for the three main types of Lemarchant mudstones (EMS, FEM, and IFE) and tuff. Also plotted are data from Rogers et al. (2006) for felsic and mafic volcanic rocks of the Tally Pond belt and the Crippleback/Sandy Brook Group crustal basement rocks. Mid Ocean Ridge Basalt (MORB) field from data from Gale et al. (2014). Arc andesite field from data from Hawkeswoth et al. (1979). All data re-calculated for $\varepsilon$Nd$_{513}$. Diagram modified after McLennan et al. (1993). (b) Plot of $f^{\text{Sm}/\text{Nd}}$ versus $\varepsilon$Nd for the EMS-, FEM-, and IFE-type mudstones and tuff, as well as the Lemarchant felsic and mafic volcanic rock from this study. Also plotted are data from Rogers (2004) and Rogers et al. (2006) for felsic and mafic volcanic rocks of the Tally Pond belt, a felsic volcanic rock samples from the unmineralized Upper Block at Duck Pond and a sample from the Mineralized Block at Duck Pond from data from McNicoll et al. (2010), and the Crippleback/Sandy Brook Group crustal basement rocks. Diagram modified after McLennan et al. (1993).

Fig. 6. (a) and (b) Zr/Ti versus Nb/Y and Nb versus Y discrimination diagrams for volcanic rocks after Winchester and Floyd (1977) and Pearce (1996) for the Lemarchant felsic and mafic volcanic rocks from this study and from data from Rogers (2004) and Rogers et al. (2006). Additionally, data fields for felsic, intermediate, and mafic volcanic rocks was added (Cloutier, in press). Data from Rogers (2004) and Rogers et al. (2006) was also used to plot the Crippleback Lake/Sandy Brook Group crustal basement rocks.
Fig. 7. Model displaying the Cambrian Tally Pond belt with juvenile Nd signatures that is built upon the Ganderian and Gondwanan Amazonian rifted crustal basement with evolved Nd signatures. (a) Early stages of arc rifting with felsic volcanism and formation of massive sulphides and genetically associated metalliferous mudstones. Scavenged and detrital juvenile and evolved Nd is archived in archiving in the metalliferous mudstones resulting in mixed signatures. (b) Final stages of arc rifting and emplacement of mafic volcanic rocks that form the hanging wall to the Lemarchant VMS deposit.

Fig. 8. Diagram of εNd versus age for Tally Pond belt mudstone and volcanic rock samples from this study and from Rogers (2004), Rogers et al. (2006), and McNicoll et al. (2010). The field for Ganderian Neoproterozoic rocks is from Rogers et al. (2006). Fields for the Mesoproterozoic Amazonian crust, the Transamazonian crust, and the West African Craton are from Satkoski et al. (2010) and references therein. Depleted mantle evolution curve is from dePaolo (1981). CHUR = Chondrite uniform reservoir.
Table 1. Sm-Nd isotope data for Lemarchant exhalites and bimodal volcanic rocks

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<th>Sample #</th>
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<th>Rock type</th>
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<th>$^{147}$Sm/$^{144}$Nd Rock $t_0$</th>
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<th>$^{2}$σ</th>
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EMS = Exhalative mudstone immediately associated with massive sulphides (within 5m) and felsic and mafic volcanic rocks; FEM = Exhalative mudstone associated with felsic and mafic volcanic rocks; IFE = Interflow mudstone within mafic volcanic rocks.

*Calculated using $^{143}$Nd/$^{144}$Nd of chondrite uniform reservoir (CHUR) = 0.512638 and $^{147}$Sm/$^{144}$Nd CHUR today = 0.1967 from Rollinson (1993)

**Calculated using the values of $^{143}$Nd/$^{144}$Nd = 0.513163 and $^{147}$Sm/$^{144}$Nd = 0.2137 for the depleted mantle reservoir (Goldstein et al.1984)

$^2$σ calculated using the algorithm after Ickert (2013)
GEOLOGY OF THE TALLY POND VOLCANOGENIC BELT AND ADJACENT AREAS (parts of NTS 12A/09 & 12A/10)


NEOPROTEROZOIC (BURNT POND AND SPENCER’S POND AREAS)
Crippleback Intrusive Suite (~584 Ma) / Sandy Brook Group (~564 Ma)
- Quartz monzonite
- Felsic lithic-vitric-crystal tuff and tuffaceous sediments
- Felsic volcanic rocks (altered and unaltered)
- Mafic volcanic rocks (altered and unaltered)

SYMBOLS
- Geological contact (inferred to defined)
- Inferred block fault
- Inferred thrust fault
- Unconformity
- Anticline (define, approximate) with plunge
- Syncline (define, approximate) with plunge

EXPLOITS SUBZONE
SILURIAN
- Battwood Group
  - Harpoon Gabbro (~465-460 Ma)
  - Composite intrusive rocks - Lemarchant granite (Cambrian or older)
  - Wigwam Brook group (~462-455 Ma)
  - Noel Pauls Brook group (~465-455 Ma)
  - Graphitic argillite (Caradoc or older)
  - Siliciclastic sediments (Caradoc or older)
  - Tally Pond group (~513-509 Ma)

CAMBRIAN AND/OR ORDOVICIAN
- Victoria Lake supergroup
  - Harpoon Gabbro (~465-460 Ma)
  - Composite intrusive rocks - Lemarchant granite (Cambrian or older)
  - Wigwam Brook group (~462-455 Ma)
  - Noel Pauls Brook group (~465-455 Ma)
  - Graphitic argillite (Caradoc or older)
  - Siliciclastic sediments (Caradoc or older)
  - Tally Pond group (~513-509 Ma)

VMS (Cambrian). Massive sulphide locations projected to surface
- Bindons Pond formation
- Felsic volcanic rocks, Bindons Pond formation
- Lake Ambrose formation
- Mafic to andesitic volcanic rocks,
- Lake Ambrose formation (altered and unaltered, Cambrian)
Heat source/rift
Synvolcanic intrusion (Lemarchant microgranite)
Magmatic volatiles/uids
REACTION ZONE
RECHARGE ZONE
EGRAHCSIDENOZRECHARGE ZONE

Fig. 2
Fig. 4

LEMARCHANT MAIN ZONE

SOUTH TARGET

NORTH TARGET

NORTHWEST ZONE

N 24 ZONE

Error bars: 2σ

Lemarchant Main Zone Deposit

Indicated Resource (7.5% ZnEq)
(surface projection)

Inferred Resource (7.5% ZnEq)
(surface projection)

Drillholes (containing hydrothermal exhalites)

Drillholes (with reported hydrothermal exhalites)

Drillholes (part of the listed holes containing hydrothermal exhalites)

Drillholes (without hydrothermal exhalites)

Fault

Roads

Mafic volcanic rocks
Felsic volcanic rocks
Felsic volcanic rocks (altered, mineralized)
Lemarchant granite (synvolcanic)
Massive sulphide (surface projection)

500 100 200 metres

Lode, Stefanie
Figure 5

(a) Diagram showing the concentrations of Th/Sc and εNd in various rock types. The diagram includes points for Lemarchant proximal mudstone, distal mudstone, and interflow mudstone, as well as data from Rogers (2004) and Rogers et al. (2006). The points are color-coded to represent different rock types and data sources.

(b) Graph showing the distribution of εNd values for different rock types. The graph includes data from McNicoll et al. (2010) and Rogers (2004) and Rogers et al. (2006). The data points are color-coded to represent different rock types and data sources.
Fig. 6

(a) Zr/Ti vs Nb/Y diagram showing the distribution of various rock types, including rhyolite, phonolite, trachyte, andesite, basalt, and foidite. Data from this study and data fields from Cloutier, unpublished data.

(b) Nb vs Y diagram highlighting the different volcanic rock types such as syn-collisional (S-type), volcanic arc (I-type), and ocean ridge (OR-type) within plate (A-type) and anomalous ocean ridge. Data from Rogers (2004) and Rogers et al. (2006).
Felsic volcanism with hydrothermal activity and formation of massive sulphides and associated metalliferous mudstones.

Erosion of Neoproterozoic and older crustal basement rocks releases evolved Nd into seawater.

Erosion of Cambrian Tally Pond belt volcanic rocks releases juvenile Nd into seawater.

Detritally derived and scavenged Nd from seawater results in metalliferous mudstones with mixed Nd isotopic signatures.

Cambrian bimodal Tally Pond volcanic belt built upon Ganderian rifted arc and older Gondwanan/Amazonian crustal basement.

Detritally derived and scavenged Nd from seawater results in metalliferous mudstones with mixed Nd isotopic signatures.
Data from this study (εNd$_{313}$)
- Lemarchant proximal mudstone and Lemarchant felsic to intermediate volcanic rock, Lemarchant mafic to intermediate volcanic rock, Lemarchant
g - Lemarchant interflow mudstone

Data from Rogers (2004) and Rogers et al. (2006)
- Tally Pond belt Lake Ambrose basalt (εNd$_{311}$)
- Tally Pond belt Bindons Pond Fm rhyolite (εNd$_{311}$)
- Sandy Brook Group rhyolite (εNd)
- Crippleback Qz-monzonite (εNd)

Data from McNicoll et al. (2010)
- Duck Pond Upper Block rhyolite and dacite (εNd$_{311}$)
- Duck Pond Mineralized Block ore horizon (felsic volcanic rocks) (εNd$_{309}$)