## GEOLOGY AND GEOCHEMISTRY OF HIGH-GRADE IRON-ORE DEPOSITS IN THE KIVIVIC, TIMMINS AND RUTH LAKE AREAS, WESTERN LABRADOR

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#### ABSTRACT

The occurrence of high-grade (> 55 wt. % Fe) iron-ore deposits in the Schefferville and Menihek regions of the Labrador Trough has been known since the 1920s, and these deposits have been mined from 1954 to 1982, and again from 2011. The deposits are hosted by the Sokoman Formation iron formation, and consist predominantly of soft friable iron ore and some lenses and layers of hard ore. There is a strong structural and stratigraphic control on the location of ore bodies, and most deposits occur in synclinal structures or steeply dipping homoclines.

High-grade iron ores are subdivided into three main types: blue, yellow, and red ores. Lesser amounts of high-Mn ores are also recorded, whereas a distinct rubble-ore was previously mined in some deposits. Major-element geochemistry shows that all ore types contain > 55% iron. Blue ore is the most common, and contains mostly hematite and martite, and has average iron contents of  $65.7 \pm 2.4$  wt. % Fe. The yellow ore contains a large proportion of goethite (LOI @ 400° C of  $4.77 \pm 1.1$ wt. %), whereas a single sample of red ore has a large proportion of clay minerals and is relatively enriched in Al (1.9 wt. %  $Al_2O_3$ ). Trace-element contents are generally low, with some samples slightly enriched in light rare-earth elements compared to samples of the least altered Sokoman Formation protolith. Hematite separates, from blue ore samples, are depleted in  $\delta^{18}O_{VSMOW}$  compared to regional samples of magnetite from the Sokoman Formation. This may be due to the influx of isotopically light fluids at low temperatures or hydrothermal fluids at elevated temperatures.

Previous genetic models for iron enrichment in this area proposed a supergene lateritic model, and simple leaching of quartz and other gangue minerals and associated relative enrichment of iron. However, comparison with other high-grade iron-ore deposits worldwide indicates these ores formed more likely due to supergene-mimetic and/or hypogene processes. A new genetic model proposes an early hypogene event (during or soon after the Hudsonian Orogeny) that formed an altered and possibly enriched proto-ore that was later overprinted by supergene mimetic enrichment. This model could explain the large variability between individual ore bodies due to different degrees of supergene overprinting of early hypogene processses. Extensional tectonics (possibly associated with the opening of the North Atlantic) reactivated normal faults that crosscut the ore bodies, resulting in erosion of exposed ore bodies, which led to the formation of detrital iron deposits (rubble ores) in graben structures. All deposits were subsequently affected by late-stage weathering, forming hard duricrusts above some deposits and remobilizing Fe and Mn.

#### **INTRODUCTION**

Numerous high-grade (> 55% Fe) iron-ore deposits occur in the Sokoman Formation of the Labrador Trough (eastern margin of the Superior Province), and form a linear belt close to the provincial border of western Labrador and northeastern Québec (Figure 1). These deposits have been intermittently mined since 1954, and recent exploration has identified, measured, and indicated resources of 110.66 Mt of iron ore at 58.55% Fe (excluding the hard, high-grade hematite deposits of the eastern Labrador Trough, Table 1). Despite the economic importance of these ore bodies, their origin has not been the subject of detailed research since the 1960s. The early studies concluded that the high-grade ironore deposits were formed during the Mesozoic, due to the downward percolation of groundwater and subsequent leaching of silica, forming enriched residual iron-ore deposits (Stubbins *et al.*, 1961; Gross, 1968). This supergene model for the formation of high-grade iron-ore deposits reflected the current state of understanding at the time. However, recent research into the formation of similar ore bodies in Australia, Brazil, South Africa and India has



**Figure 1.** Map showing regional geology and location of high-grade iron-ore deposits (modified from Wardle 1982a, b). Inset map shows location of Marginal, Ore and Eastern zones of Harrison et al. (1972).

shown that the enrichment of low-grade (~30% Fe) iron formation to form high-grade (> 55% Fe) deposits is often complex (*e.g.*, Morris, 1985; Barley *et al.*, 1999; Taylor *et al.*, 2001; Netshiozwi, 2002; Beukes *et al.*, 2003, 2008; Angerer and Hagemann, 2010; Morris and Kneeshaw, 2011; Figueiredo e Silva *et al.*, 2013), with multiple, superimposed hypogene processes (involving ascending hydrothermal fluids) and supergene processes (involving descending groundwater at low temperatures).

This report outlines the main geological features of high-grade iron-ore deposits in the Schefferville and Menihek areas of western Labrador and northeastern Québec, based on fieldwork from 2012 to 2015, data from previously published research, and industry data. New lithogeochemical and oxygen isotope data from select deposits in three main areas of recent mining activity (Kivivic, Timmins and Ruth Lake areas; Figure 2) are also presented. These data are compared with the geological and geochemical characteristics of other major high-grade iron-ore deposits worldwide, and are used to develop a new genetic model for high-grade iron-ore deposits in the Labrador Trough.

## **HISTORY OF EXPLORATION**

The first reference to iron ore in Labrador was by Father Pierre Babel, a Jesuit missionary who travelled the region in the 1860s. In the 1890s, A.P. Low of the Geological Survey of Canada reported on the iron formations near Menihek Lake and Astray Lake, and noted the potential for large deposits. However, the region attracted little interest at the time because it was remote and essentially uncharted. In 1929, high-grade iron ore was first discovered in the Knob Lake area near the Québec–Labrador border by W.F. James and J.E. Gill, working for the New Québec Company. These discoveries sparked the interest in the region, and exploration by Labrador Mining and Exploration (LM&E) in the 1930s and 1940s identified significant resources of highgrade direct shipping ore (DSO) in this area.

Following a period of intense exploration in the late 1940s, a resource of over 400 million tonnes of high-grade DSO was proven. In November 1950, the newly created Iron Ore Company of Canada (IOCC) began development of these deposits, and by 1954, a railway had been completed from the port of Sept-Îles to the new townsite at Schefferville, with production commencing in the Ruth Lake No. 3 deposit. The IOCC conducted mining operations from 1954 to 1982, during which time more than 250 million tonnes (Mt) of ore was produced (Neal, 2000). The iron-ore mines in the Schefferville and Menihek regions closed in 1982, mainly due to low iron-ore prices and increased competition from lower cost producers. However, internal IOCC documents, from 1983, indicated that non NI 43-101 compliant measured, indicated and inferred ore reserves (> 50% Fe, < 10% SiO<sub>2</sub>) of nearly 250 million tonnes remained undeveloped in 65 distinct occurrences in Labrador and Québec.

Increasing iron-ore prices in the 2000s led to renewed exploration activity in the Labrador Trough, including known occurrences of high-grade iron ore in the Schefferville and Menihek regions. This exploration included trenching and drilling of known deposits and former stockpiles, and resulted in the development of updated NI 43-101 compliant mineral reserves for a number of these deposits (Table 1). This exploration identified measured and indicated resources of 110.66 Mt of iron ore (@ 58.55% Fe) in 18, high-grade hematite deposits (excluding the 'hard' highgrade hematite deposits at Houston, Malcolm 1 and Joyce Lake described in Conliffe, 2015), and significant historical resources in other deposits currently under evaluation (e.g., the Howse deposit with non NI 43-101 indicated reserves of 28.23 Mt @ 57.94% Fe). In 2011, Labrador Iron Mines Ltd. (LIM) began production, with 3.6 Mt of iron ore mined from the James and Redmond 2B deposits from 2011 to 2013 (Plate 1A). Tata Steel Minerals Canada Limited (TSMC) began initial mining from the Timmins area in 2013, and recently started production from its new processing facility ~20 km northwest of Schefferville (Plate 1B) aiming to produce 6 Mt of sinter fines and pellet feed annually.

#### **REGIONAL GEOLOGICAL SETTING**

The Labrador Trough consists of Paleoproterozoic (2.17 to 1.87 Ga; Rohon et al., 1993; Findlay et al., 1995; Machado et al., 1997) sedimentary and volcanic rocks, collectively known as the Kaniapiskau Supergroup (Zajac, 1974; Wardle and Bailey, 1981; Le Gallais and Lavoie, 1982). The Kaniapiskau Supergroup is subdivided into three sedimentary and volcanic cycles (Figure 3), rocks of which were deposited in a foreland basin on the eastern margin of the Superior Province (Zajac, 1974; Wardle and Bailey, 1981; Le Gallais and Lavoie, 1982; Clark and Wares, 2005). Rocks of Cycle 1 (the lower cycle) developed during a rifting phase on the eastern margin of the Superior Craton at least 2.17 billion years ago (Rohon et al., 1993). The immature sandstones and siltstones of the Seward Formation grade upward into the passive margin sediments of the Attikamagen Group, which includes Le Fer Formation (siltstone and shale), Denault Formation (dolomite), Fleming Formation (chert breccia) and Dolly Formation (shale and siltstone). Cycle 2 is a transgressive sequence that progresses from shelf-type rocks of the Wishart Formation (sandstone and siltstones) and the Sokoman Formation (iron formation) at the base to deeper water turbidites of the Meni-



**Figure 2.** Simplified geological maps of the main mining areas, showing the distribution of the Sokoman Formation, location of high-grade iron-ore deposits and sample locations. A) Kivivic area; B) Timmins area; C) Ruth Lake area and; D) Redmond area.

Deposit	Province	Status	Measured	+ Indicated]	Inferred	
			Mt	% Fe	Mt	% Fe
Bean Lake	NL	Prospect			0.21	53.21
Denault	QC	Prospect	4.67	54.73		
Ferriman 4	QC	Prospect	9.36	55.37	2.08	53.78
Fleming 7N *	NL	Active Mine	6.57	61.03		
Goodwood	QC	Prospect	36.00	59.05	0.82	53.32
James	NL	Dormant Mine			0.23	52.77
Kivivic 1C *	NL	Active Mine	6.68	59.98		
Kivivic 2 *	NL	Active Mine	13.38	61.06		
Kivivic 3	NL	Prospect	1.75	60.70	1.94	58.97
Kivivic 4	NL	Prospect	7.54	58.26	0.60	59.11
Kivivic 5	NL	Prospect	8.75	60.34		
Knob Lake 1	NL	Prospect	5.08	54.71	0.64	51.78
Redmond 2B	NL	Dormant Mine	0.52	59.07	0.03	57.19
Redmond 5	NL	Prospect	1.58	55.03	0.06	52.33
Sunny 1	QC	Prospect	3.56	58.96	0.75	54.88
Timmins 3	NL	Active Mine	2.15	59.67	0.49	59.70
Timmins 4 *	NL	Dormant Mine	2.13	60.54		
Timmins 7	NL	Prospect	0.94	58.38		
Total			110.66	58.55	7.84	55.19

 Table 1. Summary of the published NI 43-101 measured + indicated, and inferred, resources of high-grade iron-ore deposits in the Ore Zone (data from New Millennium Iron Corp. and Labrador Iron Mines Ltd., reports available at <a href="http://www.sedar.com">www.sedar.com</a>)

\* Resource estimate compiled prior to recent mining

hek Formation at the top. The intermediate to mafic volcanic and volcanoclastic rocks of the Nimish Formation are interbedded with the Sokoman Formation in the Dyke Lake area (Evans, 1978). A syenite cobble, from a polymictic conglomerate in the Nimish Formation, yielded a U–Pb zircon age of 1877.8  $\pm$  1.3 Ma, which was interpreted as an approximate age for the coeval Sokoman Formation (Findlay *et al.*, 1995). In places, Cycle 2 is unconformably overlain by the Tamarack River Formation arkosic sandstones and siltstones (Cycle 3), which are interpreted as a synorogenic molasse.

The Labrador Trough forms the western part of the larger New Québec Orogenic belt, which records the oblique convergence and collision of the Archean Superior Craton to the west and the Archean to Paleoproterozoic Core Zone to the east at between 1.82 to 1.77 Ga, during the Hudsonian Orogeny (Wardle *et al.*, 1990, 2002). Harrison *et al.* (1972) subdivided the rocks of the Kaniapiskau Supergroup in the Labrador Trough into three zones, based on their structural geology (inset, Figure 1). The westernmost allochthonous Marginal Zone consists of relatively undeformed rocks of Cycle 2 that lie discordantly on the Superior Province basement rocks; its eastern boundary is marked by the Stakit Lake Fault. East of this fault, Cycles 1 and 2 rocks of the Kaniapiskau Supergroup are repeated by a series of highangle reverse faults and tight folds. This zone is termed the Ore Zone (Harrison *et al.*, 1972) and hosts the high-grade iron-ore deposits described in this study. Farther to the east, the Eastern Zone of Harrison *et al.* (1972) is characterized by open to tight folds and more widely spaced faults. The stratigraphy of the Eastern Zone is broadly similar to the Ore Zone, although the Fleming Formation has not been recorded in the Eastern Zone, and the Denault Formation is restricted to a narrow area on the eastern margin of the this zone. The high-grade iron-ore deposits in the Eastern Zone are described in Conliffe (2015).

The iron-ore deposits in the Labrador Trough are hosted by the Sokoman Formation, a 30- to 350-m-thick sequence of cherty iron-rich sedimentary rocks that can be correlated throughout the Labrador Trough. Stratigraphic and sedimentological studies in the Schefferville area have shown that the Sokoman Formation was deposited in a shallow to moderately deep shelf environment (Zajac, 1974; Pufahl *et al.*, 2014), and is characterized by significant lateral and vertical facies variations that represent changes in basin architecture and relative sea levels. The base of the Sokoman Formation is commonly marked by a dark-green to black ferruginous shale containing minor tuffs, referred to as the Ruth Formation (Zajac, 1974). Above this, the Sokoman Formation has traditionally been subdivided into three broadly defined units, termed the Upper, Middle and Lower



**Plate 1.** *A) Mining operations in the James Mine (summer 2012); B) Tata Steel Minerals Canada processing plant, with train loading high-grade ore in foreground; C) cross-section of high-grade iron-ore deposit exposed in wall of Gagnon A deposit showing synclinal structure with blue ore (B), yellow ore (Y) and red ore (R); D) cross-section through Timmins 4 deposit, showing well-preserved bedding and synclinal structure (during 2014 mining operations, bench height 3 m).* 

Iron formations. The Lower Iron Formation is predominantly composed of a magnetite and hematite-poor carbonatesilicate facies, referred to as the SCIF (Silicate Carbonate Iron Formation) in the Ore Zone. This grades upward into the Middle Iron Formation (MIF), dominated by an oxide facies and with abundant coarse-grained hematite and/or magnetite (commonly > 50% hematite and magnetite) and sugar-textured quartz. The Upper Iron Formation is another iron oxide-poor, carbonate-silicate facies rock.

## **HIGH-GRADE IRON-ORE DEPOSITS**

## **OVERVIEW**

In total, more than eighty high-grade iron-ore deposits have been recorded in the Ore Zone, and numerous other zones of altered and enriched iron formation also noted. The size and grade of iron-ore occurrences varies greatly, ranging from small pockets of altered iron formation to large deposits with > 30 Mt of iron ore at > 55% Fe. They are concentrated in a broadly linear belt up to 12 km wide, which stretches for over 100 km from the Red Dragon deposits ~20 km south of Schefferville, to the Eclipse deposit ~85 km northwest of Schefferville. This study concentrates on iron-ore deposits in the Kivivic, Timmins and Ruth Lake areas (Figure 2). All of these deposits share some important characteristics as described below, although there are significant variations in the mineralogy, stratigraphy and structural settings of individual ore bodies.

In contrast to the hard, hematite-rich iron-ore deposits in the Eastern Zone (*see* Conliffe, 2015), iron-ore deposits in



**Figure 3.** Simplified stratigraphy of the Kaniapiskau Supergroup.

the Kivivic, Timmins and Ruth Lake areas are mainly soft friable ore, along with minor hard ore in some deposits (Stubbins et al., 1961). Three main types of high-grade iron ore have been described in these deposits; these are commonly referred to as blue, yellow and red ores (Stubbins et al., 1961; Gross, 1968), and some authors (e.g., Dupéré, 2014) describing a Mn-rich ore type in some deposits. These ores are commonly stratigraphically controlled (Plate 1C) and original bedding and sedimentary structures are well preserved (Plate 1D), indicating that those three ore types formed in-situ as alteration products due to the leaching of gangue minerals and enrichment in iron. A detrital iron deposit, referred to as rubble ore (Stubbins et al., 1961; Gross, 1968), was previously mined in five deposits in the Ruth Lake and Redmond areas, and represents surficial detrital deposits that formed proximal to exposed and eroding high-grade iron-ore deposits.

#### **Blue Ores**

Blue ores are the most common type in the Ore Zone, making up > 60% of the ore-grade material (Stubbins *et al.*, 1961). They often grade into low-grade (< 45 wt. % Fe) altered oxide-facies of the Middle Iron Formation along strike (Gross, 1968), although the contact between blue ore and the Sokoman Formation protolith is occasionally sharp, and hard blocky ore has been recorded near the contact (Stubbins *et al.*, 1961). Blue ore is generally fine- to medium-grained, friable (Plate 2A), and highly porous (average 31% porosity). Iron-ore deposits outcropping at the current exposure level commonly have a hard cap of goethite-rich iron ore known as a duricrust, which is a product of recent weathering (Stubbins *et al.*, 1961).

The blue ore is predominantly composed of finegrained friable blue hematite, and variable amounts of goethite, martite, red hematite, Mn-oxides and minor quartz and magnetite (Stubbins et al., 1961). Occasionally, granules retain the oolithic nature of the proto-ore, and hematite and martite grains are commonly overgrown by secondary hematite and goethite (Gross, 1968). Large vugs and pores are often filled by vitreous goethite and Mn-oxides. Lenses and beds of hard, porous, blue ore are observed in some deposits, and consist either of hematite fragments cemented by goethite or beds of massive hematite separated by goethite and silica-rich bands (Plate 2B). Massive hematite consists of fine-grained, microplaty hematite and is generally limited to thin bands (< 10 cm), although significant thicknesses (< 2 m) of hard hematite-rich ore have been locally recorded (e.g., Red Dragon deposit; Plate 2C). Friable and altered quartz veinlets crosscut blue ore in some deposits (Stubbins et al., 1961; Plate 2A).

#### **Yellow Ores**

Yellow ores are most common in the Lower Iron Formation, and form due to the leaching and enrichment of the Fe-silicate and carbonate-rich iron formations, in particular the SCIF. Some silicate and carbonate-rich units in the Middle Iron Formation are also altered to yellow ore. The yellow ores display transitional contacts with iron formation protolith, with contact zones defined by leaching of carbonates and silica and transformation of Fe-silicates, such as minnesotaite, to goethite (Stubbins et al., 1961). The characteristic yellow colour of this ore type (Plate 2D) is due to the high content of ochreous goethite (limonite), with lesser amounts of red earthy hematite and martite and minor quartz. Yellow ores are generally very soft and sandy, and the high phosphorous and moisture content, along with lower Fe grade (~50% Fe) of the ores, means that they require beneficiation to produce a saleable product (Journeaux et al., 2010; Dupéré, 2014).



**Plate 2.** *A)* Soft, friable blue ore in the Timmins 4 deposit, crosscut by altered and friable quartz veins; B) high-grade blue ore in the James deposit, with alternating bands of hard blue hematite and soft friable hematite, goethite and quartz; C) hard blue ore from the Red Dragon deposit; D) soft, goethite-rich yellow ore exposed in trench at the Barney deposit; E) red ore in the Fleming 7 deposit (during 2014 mining operations); F) sample of rubble ore from the Redmond 1 stockpiles.

#### **Red Ores**

Red ores are mainly composed of red earthy hematite and goethite, and contain variable amounts of clay minerals and fine-grained quartz. The amount of red ore in individual deposits varies widely, and in some deposits it makes up most of the ore-grade material (*e.g.*, Fleming 7 deposit; Plate 2E). Red ores are common at the base of the iron formation, and grade along strike into Ruth Formation protolith (Stubbins *et al.*, 1961; Gross, 1968). The high aluminium content of the red ores, coupled with complex intergrowth of hematite and aluminous clay minerals, make beneficiation difficult (Gross, 1968).

#### **Rubble Ores**

Rubble ores have been recorded in five iron-ore deposits in the Ruth Lake and Redmond areas, but no examples of rubble ore have been identified in the Kivivic or Timmins areas. All *in-situ* examples of rubble ore were mined out during the IOCC's mining operations, therefore, descriptions of the setting of this ore type are based on Stubbins *et al.* (1961) and samples collected from stockpiles. The ore consists of angular to subrounded fragments ranging in size from < 1 cm to tens of metres, and are cemented by goethite, manganese oxides, quartz and ferruginous clays (Plate 2F). In the Redmond 1 deposit, the rubble ores are interbedded with a lacustrine clay unit that contains numerous fossil leaves and insects thought to be Cretaceous (Blais, 1959; Dorf, 1967).

#### **Mn-rich Ores**

Pockets of Mn-rich (> 5% Mn) ores are common in some deposits as well as in the stratigraphically lower Wishart and Fleming formations. This ore type is rich in Mn-oxides and hydroxides such as pyrolusite and hollandite (Stubbins *et al.*, 1961), and all known enrichments of Mnoxides are associated with structural discontinuities (*e.g.*, fault, well-developed cleavage, fracture-zones; Dupéré, 2014). Although NI 43-101 resource estimates have been developed for two Mn-rich deposits (Dupéré, 2014), none have been mined in the Labrador Trough, and the presence of high contents of Mn in some iron ores requires beneficiation and blending for processing.

#### STRUCTURAL SETTING OF HIGH-GRADE IRON-ORE DEPOSITS

Although there are variations in the individual structural settings of high-grade iron-ore occurrences in the Ore Zone, it is clear that there is a strong structural control on the distribution of all known ore bodies (Gross, 1968; Krishnan, 1976). Most of the structural deformation in the area is associated with the Hudsonian Orogeny (1.82 to 1.77 Ga), with compression occurring along a northeast–southwest axis (Harrison *et al.*, 1972; Krishnan, 1976). This compression initially folded the sediments into a series of tight to open synclines and anticlines, with second-order drag folds locally common (Krishnan, 1976). This was followed by the development of a series of high-angle reverse faults, generally dipping to the northeast (Harrison *et al.*, 1972), which strike subparallel to the general strike of bedding and repeating the stratigraphic sequence.

Most of the high-grade deposits occur in synclines or homoclines that are usually terminated on the northeastern side by high-angle reverse faults known as boundary faults (Figure 4; Krishnan, 1976). The synclines vary in style, from broad, open, gently plunging synclines (e.g., Goodwood, Kivivic 4, Wishart 1), to tight, steeply dipping synclines (e.g., Kivivic 3), and to complexly folded synclines that record at least two generations of folding (e.g., Timmins 4). However, these synclinal structures extend beyond the ore bodies, and with grade and alteration decreasing along strike in the same structure. Recent drilling by TSMC in the Timmins area shows that the iron formation between the Timmins 4 and Timmins 1 deposits (along strike in the same synclinal structure) consists of strongly oxidized and altered hematite and quartz with little secondary enrichment. In addition, Gross (1968) reported that the iron formation along strike from the Wishart 1 deposit grades from highgrade ore to partially leached and altered iron formation to iron formation protolith within the same synclinal structure. This indicates that although these structures may have been important in focussing later fluid flow associated with secondary enrichment, they are not the primary control on the distribution of ore bodies.

Krishnan (1976) recorded swarms of near vertical fractures in some deposits, which run almost perpendicular to the strike of bedding. In the Timmins area, ore bodies are located where these fractures crosscut synclinal structures, with the lateral extent of the deposits locally dictated by the width of the fracture system. These fracture swarms postdate the main stage of Hudsonian deformation, and Krishnan (1976) speculated that they may be related to block faulting in the underlying Archean basement rocks.

A number of deposits have also been affected by a later extensional deformation event. This extensional event has only been recorded in the Ruth Lake and Redmond areas, where normal faults have been identified in five deposits (Gross, 1968). These normal faults occur at high angles to the strike of the iron formation, and they roughly parallel the fracture swarms noted by Krishnan (1976). During this late extensional event, grabens and half-grabens formed, into which eroded fractions of the exposed iron formation in the



**Figure 4.** Cross-sections through selected high-grade iron-ore deposits, showing the stratigraphic and structural controls on the ore bodies.

footwall blocks were deposited, forming detrital iron deposits (*i.e.*, rubble ores) that include large blocks (up to 10 m) of iron ore (Stubbins *et al.*, 1961). Therefore, this extensional event must have postdated the main period of iron-ore

formation and the fossil record gives some minimum age constraints. The rubble ores contain fragments of fossilized wood thought to be Mesozoic (Blais, 1959). Further, the presence of insect and plant detritus in the lacustrine clay in Redmond 1 indicates that small ponds or lakes formed in these grabens during the Early Cretaceous. Although the exact timing of this extensional event is unknown, Krishnan (1976) argued that it may be related to the opening of the North Atlantic during the Mesozoic. The study area is located well away from the axis of this rift basin, but similar riftrelated extension and fault reactivitation has been recorded from the St. Lawrence rift system during the Mesozoic. Tremblay et al. (2013) reported apatite fission-track data for the St. Lawrence rift system indicating that normal faulting associated with the breakup of Pangea occurred from ca. 250-200 Ma, followed by tectonic inversion and compression from 160-140 Ma. In the absence of any other viable mechanism to account for the observed extensional normal faulting in the Labrador Trough postdating Hudsonian deformation, it is assumed here that this extensional event is of a similar age as in the St. Lawrence rift system.

#### WHOLE-ROCK GEOCHEMISTRY

#### **METHODS**

Twenty-five samples of high-grade ore were collected from twelve high-grade iron-ore deposits; ten from the Kivivic area, eight from the Timmins area and seven from the Ruth Lake area (Figure 2). Eighteen of these samples are classified as blue ore, three samples represent yellow ore, red ore and high-Mn ore are represented by one sample each, and two samples represent duricrust from the undeveloped Denault deposit. With the exception of the duricrust samples, all samples were collected from active mines, drillcore and trenches to minimize the effect of recent surface weathering. In addition, fourteen samples of MIF and five samples of Ruth Formation shales were collected from away from the ore bodies. These samples were selected due to the low degree of visible alteration and are referred to below as the least-altered MIF and Ruth Formation. They are considered to represent the protolith to the iron-ore deposits and were collected for comparison with high-grade ore samples.

All samples selected for geochemical analysis were prepared at the GSNL geochemistry laboratory in St. John's. Samples were milled using ceramic and tungsten carbide mills. Due to possible contamination from the tungsten carbide mill, tungsten and cobalt values are not reported. To determine precision and accuracy, reference standards and analytical duplicates were inserted at a frequency of one in twenty samples. Major-element compositions were analyzed by ICP-OES methods, following lithium tetraborate and metaborate fusion. Rare-earth elements and selected trace elements were determined by ICP-MS analysis following an identical sample digestion procedure, whereas the remaining trace elements (Be, Cu, Li, Mn, Ni, Pb, Rb, Sc, Ti, Zn) were analyzed by ICP-MS after total acid digestion. The Loss-on-Ignition (LOI) was calculated after heating the sample to 400° and 1000°C. The LOI at 400°C results in the liberation of water from goethite but not from carbonates and clay minerals, while LOI at 1000°C results in the breakdown of all minerals. This allows the percentage of goethite in the sample to be calculated, which is approximately 10 times the LOI at 400°C (calculating for the H<sub>2</sub>O content of pure hydrated goethite based on stoichiometric ratio to Fe<sub>2</sub>O<sub>3</sub>).

#### RESULTS

The major-element and trace-element compositions of high-grade ore types as well as least-altered MIF and Ruth Formation samples are summarized in Table 2. The majorelement contents are plotted in Figure 5. The blue ores have the highest grade, containing on average  $65.7 \pm 2.4$  wt. % Fe and silica contents of  $3 \pm 2.4$  wt. % SiO<sub>2</sub>. The Al<sub>2</sub>O<sub>3</sub>, P<sub>2</sub>O<sub>5</sub> and TiO<sub>2</sub> contents are generally low (< 0.5 wt. %, < 0.05 wt. %, and < 0.05 wt. %, respectively). MnO contents are 0.18  $\pm$  0.1 wt. %, and are lower than the MnO content of average MIF (Figure 5). All other major-element contents are low (generally less than 0.1 wt. %), with MgO and CaO values moderately to strongly depleted compared to the average MgO and CaO content of MIF samples (Table 2). The LOI at 400°C for blue ore samples ranges from 0.1 to 2.9 wt. %, which corresponds to goethite contents of approximately 1 to 29%.

The iron content of the yellow ores is lower than that of the blue ores ( $61.1 \pm 2.6$  wt. % Fe), coupled with higher LOI  $(6.5 \pm 1.2 \text{ wt. }\% @ 1000^{\circ}\text{C} \text{ and } 4.77 \pm 1.1 \text{ wt. }\% @ 400^{\circ}\text{C})$ and  $Al_2O_3$  (0.8 ± 0.2 wt. %). This reflects the large proportion of goethite and clay minerals in the yellow ore. The single red ore sample is characterized by high Al<sub>2</sub>O<sub>3</sub> content (1.9 wt. %), which is significantly higher than the  $Al_2O_3$ content of MIF samples but relatively depleted compared to the least-altered Ruth Formation protolith (Figure 5). The high LOI at 1000°C (5.8 wt. %) and lower LOI at 400°C (1.4%) indicate that this sample of red ore also contains a high proportion of clay minerals. The high-Mn ore has a relatively low Fe content (57.8 wt. % Fe) and MnO of 11.3 wt. %. The two duricrust samples have very high LOI (7.6% LOI @ 400°C), indicating that nearly all the iron in these samples occurs as secondary goethite.

The trace-element content of blue and yellow ore samples are generally low (< 50 ppm, most values < 10 ppm) and are similar to the average MIF, although some samples are relatively enriched in high mobile elements such as Sr and Ba (Figure 6; Table 2). The high-Mn ore sample is also enriched in Ba (672 ppm). The red ore sample is characterized by higher trace-element contents that are usually considered immobile such as V, Zr and Nb (Table 2).

Table 2. Summary statistics for major-element, trace-eleme	nt and REE content of high-grade iron-ore samples, and of aver-
age least-altered MIF and Ruth Formation protolith	

	]	BlueYellowOreOre		Red Ore	Red High Ore Mn Ore		Duricrust		Average MIF protolith		Average Ruth protolith	
n	1	17		3	1	1		2	1	4	5	;
	Av	StDev	Av	StDev			Av	StDev	Av	StDev	Av	StDev
SiO <sub>2</sub> (wt %)	3.0	2.9	5.0	5.0	2.6	0.6	4.9	3.8	47.1	14.7	50.3	7.4
$Al_2O_3$	0.4	0.5	0.8	0.2	1.9	0.3	0.2	0.1	0.1	0.0	8.9	5.0
Fe <sub>2</sub> O <sub>3</sub>	93.9	3.4	87.3	3.6	88.0	82.6	84.6	1.3	49.2	15.4	22.0	8.0
Fe	65.7	2.4	61.1	2.6	61.6	57.8	59.1	0.9	34.4	10.7	15.4	5.6
MgO	0.03	0.02	0.05	0.03	0.01	0.03	0.02	-	1.30	1.34	1.52	0.65
CaO	0.03	0.02	0.05	0.02	0.03	0.02	0.02	0.00	0.11	0.16	0.10	0.07
Na <sub>2</sub> O	0.05	0.03	0.07	0.03	0.02	0.04	0.02	0.01	0.06	0.03	0.09	0.06
K <sub>2</sub> O	0.05	0.04	0.08	0.05	0.02	0.35	0.04	-	0.09	0.04	4.14	4.11
TiO <sub>2</sub>	0.04	0.05	0.03	0.01	0.22	0.01	-	-	0.01	0.00	0.98	0.74
MnÔ	0.2	0.1	0.2	0.1	0.1	11.3	0.1	0.0	0.4	0.5	0.6	0.7
$P_2O_5$	0.07	0.04	0.10	0.09	0.20	0.01	0.16	0.17	0.02	0.01	0.20	0.15
LOI @ 1000°C	1.7	1.0	6.5	1.2	5.8	3.4	9.7	1.7	2.2	2.1	10.5	2.4
LOI @ 400°C	1.0	0.8	4.8	1.1	1.4	1.4	7.6	1.4	-	-	-	-
Rh (nnm)	45	2.5	44	17	2.7	12.2	15	_	49	17	64 7	557
Sr	10.5	18.4	49	3.6	182.5	39.2	0.9	-	2.8	1.7	15.3	11.6
Ba	37.1	55.5	23.1	20.7	39.5	672.1	1.8	0.5	17.0	19.3	314.6	298.5
V	22.8	12.1	18.2	8.2	139.6	10.9	14.3	9.5	15.8	5.2	224.7	91.5
Cu	5.9	2.6	12.0	3.1	5.5	26.5	12.0	7.0	6.1	2.0	22.8	13.1
Zn	36.3	5.7	41.9	5.0	53.2	36.5	55.0	3.9	24.9	7.9	48.6	26.2
Y	7.3	4.4	7.0	4.7	13.8	15.0	8.3	6.4	8.7	11.1	34.7	22.5
Zr	35.5	22.2	33.6	1.7	67.9	24.2	13.7	0.2	14.0	7.0	289.2	256.1
Nb	6.7	5.5	4.7	2.9	18.1	3.7	2.6	0.9	3.0	1.2	69.6	61.1
La	6.3	7.2	2.5	2.9	43.7	11.1	4.5	3.2	4.0	3.3	62.9	41.2
Ce	13.8	10.3	12.1	16.0	78.7	54.3	21.6	13.0	5.3	2.7	118.1	88.0
Pr	1.2	1.3	0.5	0.5	8.3	2.9	1.2	0.6	0.8	0.8	14.4	10.0
Nd	4.5	5.0	2.2	1.7	30.6	13.2	5.6	3.7	3.6	4.3	53.6	37.0
Sm	0.9	0.9	0.5	0.4	5.0	2.6	1.3	0.7	0.8	0.9	9.1	6.2
Eu	0.3	0.2	0.2	0.1	1.2	0.7	0.3	0.2	0.3	0.3	1.9	0.8
Tb	0.9	0.7	0.7	0.3	4.3	3.5	1.3	0.7	1.1	1.3	7.6	5.3
Gd	0.2	0.1	0.1	0.0	0.4	0.5	0.2	0.1	0.3	0.3	1.1	0.8
Dy	1.0	0.6	0.9	0.5	2.7	3.1	1.2	0.8	1.1	1.2	6.3	4.5
Но	0.3	0.1	0.2	0.1	0.5	0.5	0.2	0.1	0.3	0.3	1.2	0.8
Er	0.7	0.4	0.7	0.3	1.3	1.7	0.7	0.4	0.7	0.8	3.7	2.4
Tm	0.1	0.1	0.1	0.1	0.2	0.2	0.1	-	0.1	0.1	0.5	0.3
Yb	0.6	0.3	0.6	0.3	1.3	1.2	0.6	0.3	0.6	0.5	3.5	2.4
Lu	0.1	0.1	0.1	0.0	0.2	0.1	0.1	-	0.2	0.1	0.5	0.3
$\Sigma$ REE	29.8	24.6	21.4	21.3	178.4	95.7	38.8	2.1	18.6	15.3	284.4	199.7
Eu/Eu*SN	1.32	0.19	1.63	0.52	1.26	1.02	1.15	0.00	1.11	0.43	1.17	0.24

The total REE content ( $\Sigma$  REE) of all blue and yellow ore samples ranges from 4.2 to 87.8 ppm, whereas the single red ore sample has  $\Sigma$  REE of 178.4 ppm. When normalized to PAAS (Post-Archean average Australian Shale; McLennan, 1989), the least-altered Ruth Formation protolith shows a very typical flat REE pattern close to 1, whereas the MIF protolith has slightly lower values, especially in the LREE. The blue ore samples can be subdivided into two main populations: most of the samples show a pattern very similar to that of the MIF, somewhat lower than average shales and with a positive slope (higher HREE with respect to the LREE). In contrast, six of the blue ore samples



**Figure 5.** Bivariate plots of  $Fe_2O_3$  vs. select major elements and LOI @ 400°C for high-grade iron-ore samples. Also included are values for average least-altered MIF and Ruth Formation protolith.



**Figure 6.** Bivariate plots of  $La/Yb_{PAASN}$  ratios as proxy for LREE enrichment and concentrations of: (a) Ba; (b) Sr and; (c) total REE content.

have fairly flat REE profiles and are enriched in LREE (Figure 7B) relative to most of the blue ore samples shown in Figure 7A. These samples plot in between the patterns of MIF and Ruth Formation. The REE profiles from yellow ore samples can also be subdivided into two main populations (Figure 7C). Two samples of yellow ore, which come from lenses of yellow ore near the middle of the Sokoman Formation (MIF protolith), display a positive sloped pattern with similar HREE concentrations, but slightly lower LREE concentrations compared to the least-altered MIF. The third sample, from the base of the Sokoman Formation in the Goodwood deposit, has a flat REE profile, with slightly increased LREE concentrations compared to the least-altered MIF. The red ore sample has a REE pattern very similar to that of the Ruth Formation, with the exception of slightly lower HREE.

All samples have a minor positive Eu anomaly, which likely reflects positive Eu anomalies in the Sokoman Formation protolith (Lachance, 2015; Raye *et al.*, 2015). The blue ore samples also have a wide range of Ce anomalies (Ce/Ce\*), with weak to strong positive Ce anomalies in most samples and some samples that show minor negative Ce anomalies (Figure 8A).

#### **OXYGEN ISOTOPE ANALYSIS**

#### **METHODS**

Five samples of high-grade blue ore from the Sunny 1, Timmins 4 (two samples), James, and Ruth 8 deposits (Figure 2) were selected for oxygen isotope analysis, which represent splits of whole-rock geochemical samples that have high Fe contents (> 64% Fe) and low goethite contents (< 1% LOI @ 400°C). Hematite was separated by hand picking and heavy liquid separation (bromoform, specific gravity 2.89 g/cm<sup>3</sup>) at the GSNL geochemistry laboratory in St. John's, followed by fine crushing, hand magnet separation and reaction with dilute acetic acid to remove as many impurities as possible. All analyses were carried out at the Laboratory for Stable Isotope Science (LSIS) at the University of Western Ontario, London, Ontario, Canada, under the supervision of Dr. F. Longstaffe, who provided the following summary of the protocols applied to the XRD procedure and the methodology for determination of the oxygen isotope composition.

Prior to oxygen isotope analyses, sample purity was checked by powder X-ray diffraction (pXRD). The samples were analyzed using a high-brilliance Rigaku X-ray diffractometer, equipped with a rotating anode (CoK $\alpha$  source operated at 160 mA and 45 kV) and a graphite monochromator. Scans were performed at 2° 20/min with a step size of 0.02° 20. The results confirmed that the samples consisted of > 97% hematite.



**Figure 7.** *REE distribution patterns normalized to post-Archean average Australian Shale (PAAS) for high-grade iron-ore samples. Shaded areas show the range of values for least-altered MIF (grey) and Ruth Formation (red) protoliths.* 

For oxygen isotope analyses, approximately 8 mg of sample powder were weighed into spring-loaded sample holders, evacuated overnight at *ca.* 150°C, and then placed into nickel reaction vessels and heated in vacuo at 300°C for a further 3 hours to ensure removal of surface water. The samples were then reacted overnight at *ca.* 580°C with ClF<sub>3</sub> to release oxygen (Borthwick and Harmon, 1982 following Clayton and Mayeda, 1963). The oxygen was converted to  $CO_2$  over red-hot graphite for isotopic measurement using a

Prism II dual-inlet, stable-isotope-ratio mass-spectrometer. The oxygen isotopic analyses are reported using the normal  $\delta$ -notation in parts per thousand (‰) relative to Vienna Standard Mean Ocean Water (VSMOW). The  $\delta^{18}$ O values for standards analyzed at the same time (accepted values in parentheses) were: LSIS-Quartz, +11.4 ± 0.2‰, n = 7 (+11.5‰) and LSIS-Carbon Dioxide, +10.26 ± 0.06‰, n = 8, (+10.3‰).



**Figure 8.** Bivariate plots of (a)  $Ce/Ce^*$  vs  $Pr/Pr^*$  and (b)  $Ce/Ce^*$  vs  $La/Yb_{PAASN}$   $Ce/Ce^* = Ce_{PAASN}/((La_{PAASN}+Pr_{PAASN})/2)$ .  $Pr/Pr^* = Pr_{PAASN}/((Ce_{PAASN}+Nd_{PAASN})/2)$ .

#### RESULTS

The results of oxygen isotope analysis for hematite are provided in Table 3 and Figure 9. The  $\delta^{18}O_{VSMOW}$  values of hematite ranges from -3.99 to +1.08‰ (Table 3, Figure 9). These values are generally higher than the  $\delta^{18}O_{VSMOW}$  values

Deposit	Sample ID	δ <sup>18</sup> O <sub>VSMOW</sub> (‰)
Sunny 1	13JC064A01	1.08
Timmins 4	Tl4a	-2.61
	Tl4b	-2.69
James	JC12-070	-2.19
Ruth 8	13JC070A01	-3.99

of hematite from high-grade iron-ore deposits in the Eastern Zone (Figure 9, Conliffe, 2015), but lower than magnetite from taconite in the Eastern and Marginal zones. The sample from Sunny 1 represents the highest value, close to the magnetite values, and the two samples from Timmins 4 and James closely overlap. Ruth 8 has the lowest  $\delta^{18}O_{VSMOW}$  values of this study and is similar to some deposits of the Eastern Zone.

#### DISCUSSION

#### GEOCHEMICAL CHARACTERISTICS OF HIGH-GRADE ORE SAMPLES

All high-grade iron-ore samples are strongly enriched in Fe and depleted in SiO<sub>2</sub>, MgO and CaO compared to their least-altered MIF, which reflects the leaching of guartz, Fesilicates and carbonates during enrichment and the possible precipitation of secondary iron oxides and iron oxyhydroxides. Previous studies of other sedimentary-hosted highgrade iron-ore deposits have shown that comparisons between the whole-rock geochemical and oxygen isotope signatures of the least-altered iron formation protolith and enriched iron ore can provide information on ore-forming processes (e.g., Gutzmer et al., 2006; Beukes et al., 2008; Gutzmer et al., 2008; Thorne et al., 2009; Figueiredo e Silva et al., 2013; Hensler et al., 2014). The following section compares the geochemical characteristics of high-grade ore samples with the least-altered MIF and Ruth Formation protolith, as well as with other high-grade iron-ore deposits of supposed supergene and hypogene origin, which will aid in developing a new genetic model for the formation of these deposits.

#### Whole-rock Geochemistry

The blue ore samples are characterized by high Fe contents (65.7  $\pm$  2.4 wt. % Fe) and the LOI @ 400°C shows that hematite ( $\pm$  martite) is the predominant iron-bearing mineral in these ores. With the exception of iron, the blue ore samples generally have low major-element and trace-element contents, which are similar to or relatively depleted when compared to the least-altered MIF protolith (Table 2). Ele-



**Figure 9.**  $\delta^{is}O_{VSMOW}$  isotope values of hematite in blue ore samples from selected high-grade hematite ore bodies. Also included is oxygen isotope data from hematite separates from highgrade iron-ore deposits in the eastern Labrador Trough (Conliffe, 2015) and magnetite separates from Sokoman Formation in the Eastern Zone (Conliffe, 2015) and Marginal Zone (author, unpublished data, 2016).

vated  $Al_2O_3$  (up to 2.2 wt. %) and  $P_2O_5$  (up to 0.2 wt. %) contents in some blue ore samples may be related to the presence of shale bands in the iron formation protolith. Based on their REE profiles, blue ore samples have been subdivided into two main populations, most having similar profiles to the least-altered MIF protolith (Figure 7A), and some samples being relatively enriched in LREE (Figure 7B). The samples with similar REE profiles to the leastaltered MIF protolith suggest that REE were essentially immobile during enrichment, and Gutzmer et al. (2008) suggested that this may be a common feature of high-grade iron-ore deposits that formed due to hypogene processes. In contrast, Gutzmer et al. (2008) showed that high-grade ironore deposits that formed due to supergene processes have negative sloped REE patterns (i.e., relatively enriched in LREE) when compared to the least-altered iron formation protolith, similar to the observation in some blue ore samples in this study. Further, they suggested that this may be a possible way to distinguish between ore deposits that formed due to hypogene vs supergene enrichment, with HREE mobilized in a low temperature environment during supergene weathering resulting in the relative enrichment in LREE in the weathered residue.

In this study, the blue ore samples enriched in LREE are also relatively enriched in mobile elements such as Ba and Sr, and have higher  $\Sigma$  REE contents (Figure 6); with the Mnrich ore sample showing a similar LREE-enriched profile (Figure 7D). This indicates that the enrichment is due to LREE mobility, rather than relative enrichment of LREE in a HREE-depleted weathered residue as suggested for supergene deposits by Gutzmer et al. (2008). Evidence for the mobilization of LREE is supported by correlations with Ce anomalies, and the samples with the strongest LREE enrichment displaying negative Ce anomalies (Figure 8B). Under oxidizing conditions, soluble Ce3+ is oxidized to insoluble  $Ce^{4+}$ , resulting in a positive Ce anomaly in the weathered residue, and it is therefore unlikely that these samples represent weathered residue. This implies that the observed enrichment in LREE, Sr and Ba  $(\pm Mn)$  is associated with remobilization of these elements,

either during a phase of supergene enrichment or due to recent groundwater flow. The observation that most blue ore samples do not show any relative enrichment in REE may indicate that hypogene, rather than supergene processes were important in ore formation, although the complex behaviour of REE in hydrothermal systems make any definitive conclusions on the hypogene origin of the ore deposits difficult. Given the small sample size and low concentrations of REE in these samples, further research is required to determine the mobility of REE during alteration and enrichment.

The yellow ore samples are characterized by higher goethite contents than the blue ores (up to 5.8 wt. % LOI @ 400°C) but otherwise have broadly similar geochemical characteristics. Previous studies have shown that the yellow ores commonly form because of enrichment and alteration of the SCIF (Stubbins *et al.*, 1961), although pockets of yellow ore do occur within the MIF. These pockets of yellow ore may represent enrichment of silicate and carbonate facies recorded in the MIF (Zajac, 1974) and suggests that goethite preferentially replaces silicates and carbonates, which is consistent with petrographic observations showing that goethite pseudomorphs Fe-silicates in the SCIF close to the ore bodies (Stubbins *et al.*, 1961).

The single red ore sample has a relatively high  $Al_2O_3$  content (1.9 wt. %), which is consistent with data reported by Stubbins *et al.* (1961) who showed that red ore samples have an average  $Al_2O_3$  content of 2.1 wt. %. The values are

higher than the Al<sub>2</sub>O<sub>3</sub> content of least-altered MIF protolith (0.1 wt. %), but are significantly lower than that of the leastaltered Ruth Formation protolith shales into which they grade along strike (8.9  $\pm$  5 wt. %; Figure 5). This relative depletion of Al<sub>2</sub>O<sub>3</sub> is not matched by depletion in other elements considered immobile (e.g., V, Zr, Nb), and is difficult to understand if these red ores formed because of simple supergene enrichment and the relative enrichment of iron from ~15 wt. % Fe, in the least-altered Ruth Formation protolith, to ~60 wt. % Fe in the ores. Aluminium is generally considered to be an immobile element, but studies of weathered soils have shown that it can be extensively leached and redistributed under low pH fluid conditions (pH < 5; Kurtz et al., 2000). Therefore, the fluids responsible for the formation of the red ores are interpreted to have been acidic and capable of leaching significant amounts of aluminium.

#### **Oxygen Isotopes**

Previous studies have shown that the oxygen isotope composition of high-grade ores can provide important information on the nature and temperature of the fluids associated with alteration and enrichment (Gutzmer et al., 2006; Beukes et al., 2008; Thorne et al., 2009; Hensler et al., 2014). The leaching of quartz, iron carbonates and iron silicates associated with iron enrichment requires significant volumes of fluid (Evans et al., 2013) and therefore it is likely that the  $\delta^{18}$ O composition of secondary hematite would be in equilibrium with the infiltrating fluid. Temperate dependant oxygen isotope fractionation curves for hematite and water show that below ~100°C, hematite precipitating in isotopic equilibrium with the parent fluid should be enriched in <sup>18</sup>O, and above this temperature it would be depleted in <sup>18</sup>O (Clayton and Epstein, 1961; Yapp, 1990). Therefore, the <sup>18</sup>O content of hematite in high-grade ore bodies reflects the temperature that these ore deposits formed at, as well as the <sup>18</sup>O content of fluids responsible for the alteration and enrichment.

The  $\delta^{18}$ O values of hematite from the blue ores are lower than  $\delta^{18}$ O<sub>VSMOW</sub> values of magnetite reported from the Sokoman Formation in the Labrador Trough. Conliffe (2015) reported  $\delta^{18}$ O values of 1.57 to 3.23‰ from magnetite in taconites in the Snelgrove Lake area, whereas magnetite separates from MIF in the Howells River area yielded  $\delta^{18}$ O<sub>VSMOW</sub> values of 1 to 1.6‰ (author's unpublished data). This is consistent with data from Raye *et al.* (2015), who reported  $\delta^{18}$ O values of -1 to 2.6 for magnetite separates from the Sokoman Formation close to the western margin of the Labrador Trough. This relative depletion of <sup>18</sup>O in highgrade ores has been observed in other studies (Gutzmer *et al.*, 2006; Beukes *et al.*, 2008; Thorne *et al.*, 2009; Hensler *et al.*, 2014) and is commonly related to the influx of hydrothermal fluids at high temperatures (magmatic, metamorphic or meteoric fluids, or basinal brines; Angerer *et al.*, 2014). These studies have used the calibration of Yapp (1990) and extrapolated it to 300°C (*e.g.*, Gutzmer *et al.*, 2006; Beukes *et al.*, 2008; Thorne *et al.*, 2009; Hensler *et al.*, 2014), which shows that at increasing temperatures, the secondary hematite should be more depleted in  $\delta^{18}$ O compared to the infiltrating fluid. Therefore, the  $\delta^{18}$ O values in this study may indicate the infiltration of hydrothermal fluids similar to those thought to have formed the high-grade iron-ore deposits in the eastern Labrador Trough (Conliffe, 2015), albeit at lower temperatures. Alternatively, the  $\delta^{18}$ O signature of the blue ores may instead be related to influxes of isotopically light (< -10‰ <sup>18</sup>O) meteoric fluids at low temperatures, with secondary hematite enriched in <sup>18</sup>O compared to the infiltrating fluid.

#### **GENETIC MODELS FOR HIGH-GRADE IRON ORE**

Early research into the origin of high-grade iron-ore deposits in the Labrador Trough suggest they formed during the Mesozoic as a result of supergene enrichment (Stubbins et al., 1961; Gross, 1968). In this model, the downward percolation of groundwater into the fractured iron formation is responsible for the leaching of gangue minerals and associated oxidization and, therefore, a relative Fe enrichment. Similar models were proposed for the formation of other high-grade iron-ore deposits elsewhere in North America and Australia during that time (e.g., MacLeod, 1966). However, recent research into the origin of the sedimentary-hosted high-grade iron-ore deposits in Australia, Brazil, South Africa and India have shown that these deposits are commonly more complex, having multiple stages of hypogene and/or supergene enrichment processes (Morris, 1985; Barley et al., 1999; Taylor et al., 2001; Netshiozwi, 2002; Beukes et al., 2003, 2008; Angerer and Hagemann, 2010; Morris and Kneeshaw, 2011; Figueiredo e Silva et al., 2013). Ramanaidou and Wells (2014) subdivided enriched, bedded, high-grade iron-ore deposits into three main types: 1) residual iron ore, 2) martite-goethite ores, and 3) martite microplaty hematite ores. These ore bodies are commonly influenced by later weathering, and detrital iron deposits (DID), such as rubble ore, can form due to the weathering of exposed ore bodies. In the following sections each of these ore-deposit types are described, and the features of each type are compared with the high-grade iron-ore deposits in the Ore Zone of the Labrador Trough.

#### **Residual Iron-ore Deposits**

Residual iron ores form from supergene lateritic processes, with iron enrichment due to dissolution of silica, carbonates and Fe-silicates, and some precipitation of secondary iron oxides (Ramanaidou and Wells, 2014). This process is similar to the previously proposed models for formation of high-grade iron-ore deposits in the Labrador Trough (Stubbins et al., 1961; Gross, 1968), but several features of this model are not found in the deposits of this study. For instance, residual iron-ore deposits are characterized by near complete destruction of original textures (Ramanaidou, 2009; Ramanaidou and Wells, 2014). However, all iron-ore deposits in the Labrador Trough retain primary features such as bedding and oolitic textures. In addition, residual iron ores form mainly due to simple subtraction of select major elements, and iron enrichment is relative. Although the ores in the Labrador Trough are more porous than the iron formation protolith, simple leaching of gangue minerals is not enough to account for the observed iron enrichment (Stubbins et al., 1961). Further evidence supporting processes of addition of iron is apparent in the red ores of the Ruth Formation, where enrichment of iron is not accompanied by enrichment of other immobile elements such as Al (Stubbins et al., 1961; Table 2). Therefore, it is considered unlikely that high-grade iron-ore deposits in the Labrador Trough represent residual iron-ore deposits as once thought.

#### Martite-Goethite Ore Deposits

Martite–goethite ores have been described in detail by Morris and Kneeshaw (2011) and are dominantly composed of soft friable ore, abundant goethite and martite (generally > 50% goethite). The yellow ores and some of the blue ores can be potentially classified as martite-goethite ores due to their high goethite content. Martite–goethite ores are thought to have formed by supergene mimetic processes, where gangue minerals are replaced by iron oxyhydroxides at depth (Morris, 1985). Groundwater is circulated in largescale electrochemical cells, driven by seasonal atmospheric effects, and facilitated by the excellent conductive properties of magnetite layers in the iron formation. The groundwater penetrates the iron formation well below the surface in an open-ended aquifer, and flow is facilitated by pre-existing structures (folds, faults and fractures).

Iron is leached from the iron formation close to the surface (possibly involving biogenic reactions in the vadose zone) and transported deep into the systems as soluble  $Fe^{2+}$ ions in the groundwater (Morris and Kneeshaw, 2011). These  $Fe^{2+}$ -rich supergene fluids leach gangue minerals such as chert, Fe-silicates and carbonates, with silica solubility at depth greatly increased due to ferrous-ferric reactions with quartz (Morris and Fletcher, 1987). During this supergene mimetic replacement, the original textures of the iron formation are preserved, where leached gangue minerals are pseudomorphed by  $Fe^{3+}$  iron oxyhydroxides, such as goethite (Ramanaidou and Wells, 2014). Primary magnetite is oxidized to martite and hematite is unchanged. The detailed preservation of primary textures is common in high-grade ore deposits in the Labrador Trough, where goethite is seen pseudomorphing Fe-silicates (Stubbins *et al.*, 1961). The leaching of iron close to the surface results in the formation of a residual, silica-rich cap above the ore bodies (Morris and Kneeshaw, 2011). Although this silica-rich cap would likely be eroded away in most ore bodies, it could account for the quartz-rich units located above the Knob Lake and Red Dragon ore bodies, which are difficult to explain if these deposits formed due to simple leaching of gangue minerals close to the surface due to supergene lateritic processes.

## Martite-Microplaty Hematite Ores

Hematite ores consist predominantly of fine-grained martite and microplaty hematite, and minor goethite and silica. These ores can be soft, friable, or hard, and commonly preserve the original textures of the iron formation protolith (Morris and Kneeshaw, 2011; Ramanaidou and Wells, 2014). Martite-microplaty hematite ores are similar to the blue ores in the Labrador Trough, which consist predominantly of fine-grained hematite and martite and minor goethite and silica (Stubbins *et al.*, 1961; Gross, 1968).

The origin of martite-microplaty hematite ores remains contentious, and three main models for their formation are proposed (see Ramanaidou and Wells, 2014). A syngenetic or syndepositional model has been proposed for some ironore deposits where no evidence of original chert bands is preserved (Lascelles, 2007). This model is not applicable to the blue ores as there is abundant evidence of original ironrich and chert-rich layers (Plate 2B). The supergene mimetic-metamorphism model proposes a multistage process, with the formation of a martite-goethite ore body, which was subsequently buried and subjected to low-grade metamorphism (80-100°C), that transformed goethite into microplaty hematite (Morris, 1985; Morris and Kneeshaw, 2011). In the Labrador Trough, there is no evidence of burial, or lowgrade metamorphism, after the formation of these ore bodies, and petrographic evidence indicates that goethite precipitation in the blue ores generally postdates the oxidation of magnetite from the iron formation protolith into martite and fine-grained hematite (Stubbins et al., 1961). In addition, it seems unlikely that significant dehydration of goethite could occur in the blue ores, but not affect the underlying yellow ores that contain abundant goethite pseudomorphing fine textures in the SCIF.

A hypogene model for the formation of martitemicroplaty hematite ores involving hypogene alteration and the circulation of hydrothermal fluids after diagenesis has been proposed by a number of studies (Barley *et al.*, 1999; Taylor *et al.*, 2001; Netshiozwi, 2002; Angerer and Hagemann, 2010; Figueiredo e Silva *et al.*, 2013). These studies define hydrothermal fluids as warm (*i.e.*, higher temperature than the ambient rock temperature) water-rich fluids that react with the rocks, independent of fluid source. Hypogene alteration involves changes in the chemical or mineralogical composition of the host rock due to these ascending hydrothermal fluids (Hagemann *et al.*, 2008). Hydrothermal fluids are capable of leaching Fe-silicates, carbonates and chert (Angerer *et al.*, 2014), oxidizing magnetite to martite hematite (liberating Fe<sup>2+</sup>; Ohmoto, 2003), precipitating secondary microplaty hematite and creating secondary porosity. Ohmoto (2003) also showed that the transformation of magnetite to hematite and transport of Fe<sup>2+</sup> in solution is most efficient at low pH conditions, and the influx of acidic hydrothermal fluids may also explain the relative depletion in Al observed in the Ruth Formation shales.

# EVIDENCE FOR EARLY HYPOGENE ALTERATION IN THE LABRADOR TROUGH

A hypogene or hydrothermal origin for high-grade ironore bodies in the Labrador Trough was considered by Stubbins et al. (1961), who stated that "hydrothermal contributions might explain better some of the mineralogical and chemical transformations noted" (Stubbins et al., 1961). However, a hydrothermal origin for these fluids was dismissed, due to the lack of intrusions close to the ore bodies and the association of ore bodies with the present erosional surface (Stubbins et al., 1961; Gross, 1968). Recent studies from ancient and active fold thrust belts have shown that significant amounts of formation waters, meteoric fluids and/or basinal brines can flow through fold hinges (Sibson, 2005) or along faults (Lacroix et al., 2014), and intrusive igneous activity is not required to account for the flow of hydrothermal fluids through these rocks. In addition, Conliffe (2015) proposed that structurally controlled high-grade iron-ore bodies in the Eastern Zone of the Labrador Trough formed because of hydrothermal fluid flow, and these deposits are located in similar structural settings as iron-ore deposits in the Ore Zone (e.g., synclines, homoclines associated with high-angle reverse faults). Combining observations from the two zones, it is likely that significant volumes of hydrothermal fluids circulated through the Sokoman Formation during orogenesis, and this fluid flow would have been focussed along structures such as fold hinges and highangle reverse faults. Fluid flow would also have been enhanced where the iron formation was crosscut by fracture swarms, which would account for the distribution of ore bodies noted by Krishnan (1976). Although the timing of this brittle deformation is unknown, it is likely that the intersection of these brittle fracture zones with the high-angle reverse faults provided a conduit for fluid flow and they were an important factor in the development of ore bodies.

Although hydrothermal fluid flow does not account for all of the characteristics of high-grade iron-ore deposits in the Ore Zone, it may have led to the development of an enriched proto-ore, which was later modified and further enriched by supergene processes. Evidence for this early alteration and enrichment is preserved along the margins of the ore bodies, with the MIF grading into low-grade altered iron formation along strike (similar to high-grade ore bodies in the Eastern Zone; Conliffe, 2015) and the local occurrence of hard and blocky blue ore near sharp contacts with MIF protolith (Stubbins et al., 1961). The presence of friable and altered quartz veinlets crosscutting blue ore (Stubbins et al., 1961; Plate 2A) is consistent with episodes of hydrothermal activity predating supergene leaching and alteration. In addition, lenses of hard, hematite-rich lenses and layers in some deposits supports a hypogene model for at least some of the iron enrichment. However, the present data are unable to distinguish a distinct geochemical signature of this event, and more detailed *in-situ* geochemical and isotopic analysis is required to confirm the nature and extent of any hypogene enrichment event.

#### MODEL FOR FORMATION OF HIGH-GRADE IRON-ORE DEPOSITS IN THE ORE ZONE, LABRADOR TROUGH

The geological and geochemical characteristics of highgrade iron-ore deposits in the Ore Zone indicate that rather than a simple process of leaching silica by supergene fluids, iron enrichment was likely a complex and multistage process of superimposed hypogene and supergene events (Figure 10). A new model for iron enrichment is presented below, which outlines the nature and relative timing of these events. However, there is considerable variation in the metallurgy, Fe-grade, structural setting and relative proportion of different ore types (including martite–goethite and martite microplaty hematite ores) between individual ore bodies, and further detailed research is required to determine the relative importance of these events in the formation of individual deposits.

During the Hudsonian Orogeny, the rocks of the Kaniapiskau Supergroup were extensively deformed in an active fold and thrust belt, forming a series of high-angle reverse faults and tight folds, which are crosscut by later swarms of vertical fractures (Harrison *et al.*, 1972; Krishnan, 1976). The initial deformation created fluid pathways, which could have facilitated the flow of hydrothermal fluids. Conliffe (2015) showed that similar hypogene events in the eastern Labrador Trough were capable of creating significant ore bodies with little or no late supergene overprint. In the Ore Zone, this alteration may have formed a moderately enriched proto-ore, consisting predominantly of microplaty hematite, martite and quartz and lenses and layers of hard, high-grade ore. In the ore bodies, any evidence for an early, hypogene alteration stage in the Ore Zone is



**Figure 10.** *Relative timing of possible alteration and enrichment events associated with the genesis of high-grade iron-ore deposits in the ore zone. See text for details.* 

largely overprinted by later events. However, little attention has been paid to the iron formation along the margins of known ore bodies, and recent drilling in the Timmins area has revealed the presence of strongly altered and partially leached, but low-grade (< 45 wt. % Fe), iron formation (consisting of hard, fine-grained hematite and quartz) between the Timmins 1 and Timmins 4 ore bodies. This may represent proto-ore, which was unaffected by later events, and further research is required to determine the relative importance of early hypogene alteration in iron enrichment.

The second stage of iron enrichment is the downward percolation of groundwater and associated supergene enrichment, which postdates Hudsonian deformation and possible hypogene alteration, but predates extensional tectonics associated with the opening of the North Atlantic. The exact timing of any supergene event(s) is unknown. Recent  $(U-Th)^{/21}Ne$  and  $^{4}He^{/3}He$  ages from goethite in similar deposits in the Gogebic iron range, Michagan, USA, yielded ages of  $772 \pm 41$  Ma and  $453 \pm 14$  Ma (Farley and McKeon, 2015). These ages are difficult to reconcile with any major tectonic events in the Gogebic iron range (Farley and

McKeon, 2015), and they show that the supergene weathering can occur at multiple times over geological time.

The supergene event in the Labrador Trough is mimetic and preserves the original features of the iron formation protolith, unlike the supergene lateritic model proposed by previous authors (Stubbins et al., 1961; Gross, 1968). The iron formation must have been exposed to the surface to facilitate leaching of iron in the vadose zone (Morris and Kneeshaw, 2011), and groundwater would have penetrated to deep levels along pre-existing fractures and faults. These fluids would have leached any remaining gangue minerals and replaced them with secondary goethite, resulting in the friable, soft ore found in most deposits. The relative amounts of goethite, martite and hematite in the various ore types would have been strongly controlled by the original mineralogy of the protolith. Units rich in Fe-silicates and carbonates formed goethite-rich yellow ores (e.g., SCIF), whereas units originally consisting mainly of iron oxides and chert formed hematite-rich blue ores (e.g., MIF and proto-ore after hypogene alteration). This groundwater penetration may also have been enhanced by increased porosity associated with early hypogene events. Leaching of iron in the upper iron formation would have formed a cap of leached and friable silica that has been eroded from above most deposits but is still recorded in the Knob Lake and Red Dragon deposits.

The main stages of iron enrichment, as described above, must have occurred prior to extensional tectonism, as evidenced by large (> 15 m) blocks of ore-grade material found in detrital iron deposits that fill graben structures (Stubbins et al., 1961). The exact timing of this extensional event is unknown, but it is likely to be associated with the opening of the North Atlantic, with similar extensional faulting in the St. Lawrence rift system thought to have occurred between 250 and 200 Ma (Tremblay et al., 2013). Following this tectonic event, exposed iron-ore deposits were eroded, and shallow blankets of outwash scree containing detritus from these ore bodies accumulating in nearby depressions such as grabens, thereby forming detrital iron deposits above five of the iron-ore deposits in the Ore Zone. Although the exact timing of the onset of this type of deposition is unknown, the presence of a lacustrine clay deposit containing fossilized Cretaceous insects and wood close to the top of the rubble ores in the Redmond 1 deposit indicates that it continued until at least 100 Ma (Blais, 1959; Dorf, 1967).

All iron-ore deposits in the Ore Zone are affected by late-stage, secondary weathering, which is a common feature of most iron-ore deposits worldwide (Ramanaidou and Wells, 2014). Although this lateritic weathering does not seem to be postdates the main stages of iron enrichment, it is responsible for the effective redistribution of iron and other mobile elements (e.g., Mn), precipitating vitreous goethite and Mn-oxides in vugs and pore spaces (cementing previously friable ore) and forming pockets of Mn-rich ores. Unlike the other enrichment events, this is not restricted to the Sokoman Formation, with ferruginous manganese deposits also recorded in the stratigraphically lower Wishart and Fleming formations (Dupéré, 2014). Lateritic weathering also formed the hard, goethite-rich duricrust above some of the ore bodies, which protected the underlying soft friable ore from erosion.

#### **CONCLUSIONS**

Numerous high-grade iron-ore deposits occur in the Schefferville and Menihek areas of the Labrador Trough, in a structural zone known as the Ore Zone (Harrison *et al.*, 1972). These deposits have a wide range of geological characteristics, which is consistent with a complex and multistage enrichment of both hypogene and supergene processes.

Whole-rock geochemical data show important differences between different ore types. Blue ore samples have the highest Fe grades, and are generally low in undesirable elements (e.g., Al, P, Mn). Yellow ore samples have a much higher LOI at 400°C, reflecting their high goethite content. The one red ore sample in this study has a high clay content (LOI @ 1000°C >> LOI @ 400°C), and is relatively enriched in Al compared to other ore samples but strongly depleted in Al compared to the least-altered Ruth Formation protolith. Rare-earth-element (REE) data show that some ore samples are enriched in light REE, which may reflect the mobility of these elements during enrichment. Oxygen isotope analysis of hematite from blue ore samples reveals that they are depleted in  $\delta^{\scriptscriptstyle 18}O_{\rm VSMOW}$  compared to magnetite in the Sokoman Formation protolith. This depletion may be due to the influx of isotopically light meteoric water at low temperatures, or hydrothermal fluids with  $\delta^{18}$ O around 0‰ at higher temperatures (> 150°C). These two contrasting interpretations show the difficulty in deciphering the nature of the fluids responsible for enrichment without an independent geothermometer.

Comparisons between high-grade iron-ore bodies in the Ore Zone, and other high-grade iron-ore deposits worldwide, show that their characteristics are incompatible with enrichment due to simple supergene lateritic processes as proposed by previous researchers. Instead, these deposits share similarities with martite-goethite and martitemicroplaty hematite ore types. A new genetic model proposes that they formed by early hypogene alteration (forming an enriched proto-ore), which was later overprinted by supergene mimetic enrichment. The timing of these processes is unknown, but hypogene alteration likely occurred during or soon after the Hudsonian Orogeny (1.82 to 1.77 Ga). Supergene mimetic enrichment postdated the hypogene alteration, but occurred prior to reactivation of normal faults during the opening of the North Atlantic (250 to 200 Ma). Following a period of extensional faulting, some ore bodies were exposed and eroded (forming detrital iron deposits in grabens above 5 deposits), and all ore bodies were affected by late-stage weathering and remobilization of iron and manganese. Further research is required to determine the relative importance of hypogene and supergene alteration and the timing of these events.

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