Paper 11

An Overview of the Use of Petrochemistry in Regional Exploration for Volcanogenic Massive Sulfide (VMS) Deposits

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ABSTRACT

Volcanogenic massive sulfide (VMS) deposits are important global sources of base and precious metals. Igneous geochemistry (petrochemistry) of mafic and felsic rocks associated with VMS deposits is extremely useful in delineating potentially fertile ground for VMS mineralization. In mafic-dominated, juvenile environments (e.g., mafic, bimodal mafic, and mafic-siliciclastic VMS-types) VMS deposits are associated with boninite and low-Ti island arc tholeiite, mid-ocean ridge basalt, and back-arc basin basalt. In evolved environments, those associated with continental crust and typically dominated by felsic magmatism (e.g., bimodal felsic and felsic-siliciclastic VMS-types), VMS-associated mafic rocks have alkalic (ocean island basalt-like), and/or mid-ocean ridge/back-arc basin basalt-like signatures. In these environments alkalic basalts and mid-ocean ridge/back-arc basin basalt -like mafic rocks overlie felsic rocks and mineralization and represent melts derived from lithospheric and asthenospheric mantle sources, respectively. Felsic rocks in Archean sequences are typically tholeiitic, have elevated high field strength elements and rare earth elements, and FIII affinities (low Zr/Y and La/Yb, flat chondrite-normalized rare earth element profiles). In post-Archean evolved environments, felsic rocks associated with VMS deposits have high field strength element- and rare earth element-enrichment and within-plate signatures on discrimination diagrams, like their Archean counterparts, but are more calc-alkalic in composition and commonly have FII affinities. Felsic rocks associated with VMS deposits in post-Archean mafic-dominated, juvenile substrates are associated with trace element depleted rhyolites with tholeiitic to boninitic signatures, and M-type and FIV affinities on discrimination plots. Using mafic or felsic rocks in isolation may lead to erroneous assignments of prospectivity for terrains; however, when mafic and felsic rocks are used in tandem they are powerful tools in outlining potentially prospective regions. Within VMS-hosting environments there are specific petrochemical assemblages of mafic and felsic rocks. Petrochemical assemblages are specific lithogeochemical associations between mafic and felsic rocks that are common to VMS forming environments and are useful in identifying two key ingredients required to form prospective VMS belts: 1) rifting; and 2) high temperature magmatism

INTRODUCTION

Volcanogenic massive sulphide (VMS) deposits have been, and continue to be, important contributors to the global economy. These deposits are important global sources of the base metals Zn, Pb, and Cu, and many deposits (e.g., Eskay Creek, Bousquet-LaRonde) are important sources of precious metals. VMS deposits are one of the best understood mineral deposit types due to numerous studies of ancient deposits, as well as those that are currently forming on the modern seafloor.

Lithogeochemistry and petrology have been an important tool in our understanding of and exploration for VMS deposits since the 1970s, and continue to be an important tool in our arsenal in exploring for these deposits. Most of the early lithogeochemical work in the in the 1970s and 1980s focused on alteration systems (e.g., Ishikawa et al. 1976; Spitz and Darling 1978; Saeki and Date 1980; Date, Watanabe, and Saeki 1983; Gibson, Watkinson, and Comba 1983; Campbell et al. 1984; Lesher, Gibson, and Campbell 1986; Kranidiotis and MacLean 1987; MacLean and Kranidiotis 1987; MacLean 1988) with few studies aimed at using lithogeochemistry to outline prospective belts on a regional scale (e.g., Lesher et al. 1986; Paradis, Ludden, and Gelinas 1988; Swinden et al. 1989).

With the developments of new analytical technology in the late 1980s and 1990s, however, this situation has changed and the application of high precision lithogeochemical data is becoming commonplace in VMS deposit exploration. One of the key advances was the development of inductively coupled plasma mass spectrometer (ICP-MS), once primarily a research tool in universities and government laboratories, which has become commonplace in most commercial lab facilities. The ICP-MS system has provided a means of acquiring high quality analytical data for over forty trace elements with rapid turn-around times on a variety of matrices (e.g., rocks, soils, waters, and biological materials)(Jenner et al. 1990; Eggins et al. 1997; Sylvester 2001, and references therein; Günther and Hattendorf 2005). The ICP-MS system has revolutionized the application

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of lithogeochemistry to VMS exploration and has led to major advancements in understanding volcanic and intrusion lithogeochemistry and its application to delineating prospective regions for VMS exploration (e.g., Swinden 1991; Barrie, Ludden, and Green 1993; Syme and Bailes 1993; Barrett and Sherlock 1996; Swinden 1996; Kerrich and Wyman 1997; Lentz 1998; Syme 1998; Syme et al. 1999; Wyman, Bleeker, and Kerrich 1999; Piercey, Murphy et al. 2001; Piercey, Paradis et al. 2001; Galley 2003; Hart, Gibson, and Lesher 2004). Furthermore, the ICP-MS system has allowed the utilization of a wider range of trace elements in lithogeochemical exploration, including the high field strength elements (HFSE) and rare-earth elements (REE), elements that were unavailable to explorationists in the 1970s and 1980s, except at prohibitively high costs and with low turn-around times.

In this paper the current state of knowledge in lithogeochemistry of mafic and felsic igneous rocks associated with VMS deposits and their application to exploration for VMS systems on a regional scale will be reviewed. Regional scale, as defined in this paper, is >1:50,000 scale and the methods put forth are aimed at defining prospective belts and stratigraphic sequences. The paper will first define the attributes of the target and outline key geological and tectonic settings that these deposit occur in. This will be then followed by reviews of the key advances and current state of knowledge in volcanic and intrusion geochemistry and petrochemical assemblages - key chemostratigraphic relationships useful for delineating prospective environments at the belt scale. The paper will conclude with are the unresolved questions and anticipated advances in the next ten years.

THE TARGET: VOLCANOGENIC MASSIVE SULFIDE DEPOSITS AND THEIR CLASSIFICATION

Volcanogenic massive sulfide (VMS) deposits have formed throughout Earth history from early Archean to the present. They form within extensional geodynamic regimes, in particular rift environments (Figure 1). These rift environments include mid-ocean ridges, back-arc basins, intra-oceanic arc rifts, and continental arc rifts (e.g., Swinden 1991; Hannington et al. 1995; Scott 1997; Syme et al. 1999; Barrett, MacLean, and Tennant 2001; Piercey, Paradis et al. 2001; Dusel-Bacon, Wooden, and Hopkins 2004). On a belt scale, deposits within these rifts are associated with extensional and trans-tensional grabens, synvolcanic and synsedimentary faults (e.g., Gibson 1989; Allen 1992; McPhie and Allen 1992; Setterfield et al. 1995; Allen et al. 1996; Gibson, Morton, and Hudak 1999; Stix et al. 2003; Gibson 2005). These synvolcanic and synsedimentary structures are commonly associated with felsic and mafic dyke swarms that parallel the axis of the rift corridor (e.g., Gibson and Watkinson 1990; Setterfield et al. 1995; Gibson, Morton, and Hudak 1999) and are commonly above a coeval synvolcanic intrusive complex (Figure 1)(e.g., Campbell et al. 1981; Galley 1996, 2003). The dyke swarms and subvolcanic intrusive complexes typically have geochemical signatures identical to the VMS-hosting volcanic sequences; however, many cross-cut and post-date the formation of VMS mineralization (e.g., Galley 1996; Barrett and MacLean 1999; Galley 2003). The subvolcanic intrusive complexes are interpreted to be the manifestation of a geodynamic environment with an elevated geothermal gradient (high heat flow) - a key ingredient for driving hydrothermal circulation required to form VMS mineralization (e.g., Campbell et al. 1981; Galley 1996; Large et al. 1996; Galley 2003). Some workers have also suggested that these intrusive complexes also contribute metals to the VMS hydrothermal system (Large et al. 1996; Galley 2003).

A major challenge in VMS exploration is the identification of high heat flow rift environments. Petrochemistry, in conjunction with geological methods, is particularly useful in identifying rift sequences as rift-related mafic and felsic rocks have have specific petrochemical signatures (see below). Furthermore, petrochemistry can also provide an indicator of the heat flow of a specific geodynamic environment. Many VMS environments are associated with rocks that have signatures indicative of generation and emplacement at high temperatures; hence, their petrochemical signatures can provide an indicator of the geothermal gradient and heat flow of the environment that these rocks were emplaced into.

Figure 1 illustrates the generalized setting of VMS deposits, but there are significant differences in the style and setting of these deposits (Barrie and Hannington 1999; Franklin et al. 2005). These variations may extend to the lithogeochemical signatures of the VMS-associated rocks, hence, a brief note on VMS classification is required. In recent years VMS deposits have been classified into five groups based on their host-rock assemblages (Barrie and Hannington 1999; Franklin et al. 2005):

1) Mafic: these are deposits associated with mafic-dominated assemblages, commonly ophiolitic. The Cyprus, Oman, and ophiolite-hosted deposits in the Newfoundland Appalachians represent classic districts of this group;
2) Bimodal-mafic: these are deposits associated with mafic-dominated settings, but with up to 25% felsic rocks, the latter often hosting the deposits. The Noranda, Flin Flon-Snow Lake and Kidd Creek camps would be classic districts of this group;
3) Mafic-siliciclastic (or pelitic-mafic): these are deposits associated with subequal proportions of mafic and siliciclastic rocks; felsic rocks can be a minor component; and mafic (and ultramafic) intrusive rocks are common. The Besshi deposits in Japan, Outokumpu deposits in Finland, and Windy Craggy in Canada represent classic districts of this group;
4) Felsic-siliciclastic (or siliciclastic-felsic or bimodal siliciclastic): these are deposits associated with siliciclastic dominated settings with abundant felsic rocks and less than 10% mafic rocks. These settings are often shale-rich and the Bathurst camp, Iberian Pyrite Belt, and Finlayson Lake areas are classic districts of this group; and
5) Bimodal-felsic: these are deposits associated with bimodal sequences where felsic rocks are in greater abundance than mafic rocks with only minor sedimentary rocks. The Kuroko, Buchans, and Skellefte camps would be classic districts of this group.

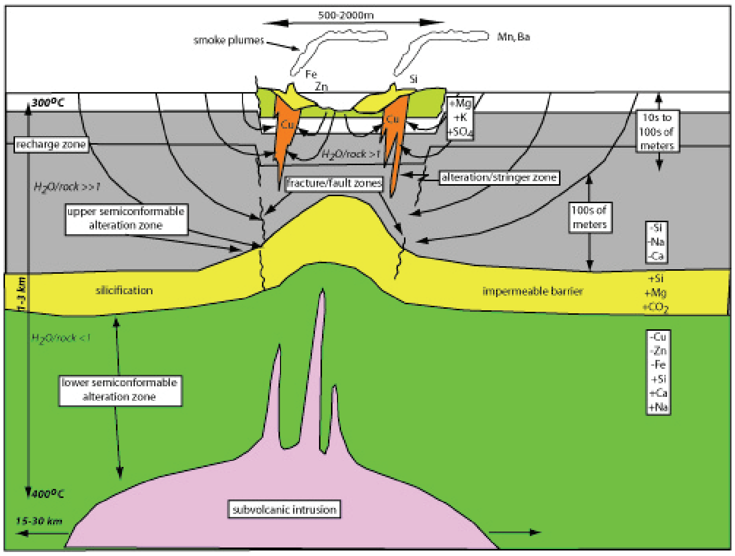

Figure 1: Model for the setting and genesis of volcanogenic massive sulphide (VMS) deposits (from Franklin et al., 2005)

The first three groups above are dominated by mafic material and juvenile environments with very little continental crustal influence. Felsic rocks are derived primarily from melting of hydrated mafic crust, and mafic rocks are predominantly sourced from asthenospheric mantle. The first three groups are also dominated by deposits enriched in Cu-Zn with very little Pb. The last two groups are associated with evolved environments dominated by continental crust or continental crust-derived sedimentary rocks. Felsic rocks in these environments are derived from melting of continental crust or continental crust-derived rocks, and mafic rocks often are derived from both lithospheric and asthenospheric sources. The deposits of the last two groups are notably Zn-Pb-Cu dominated. As will see later, whether one is in a juvenile versus evolved environment will ultimately control the felsic and mafic volcanic geochemistry associated with the VMS deposits in a given district. Further information on this deposit type, classification, genesis, and exploration, is provided in a companion paper by Gibson et al. (this volume).

REVIEW: VOLCANIC AND INTRUSION GEOCHEMISTRY

Volcanic and intrusion geochemical attributes will be discussed together as the composition and textures of both are used primarily to identify regional targets and to identify key regions to undertake detailed exploration (i.e., area selection). In this section, the primary petrological and geochemical signatures in volcanic and intrusive rocks are examined. The primary petrological signatures of rocks are critical to understand because they provide key information on the thermal, tectonic, and petrological history of the mafic and felsic rocks. Thus, it is critical that the freshest, least altered samples (e.g., those having preserved textures, free of veins and secondary minerals) are taken as the geochemical signatures in these rocks will reflect primary tectonic and petrological processes, rather than those associated with secondary alteration (Jenner 1996; Kerrich and Wyman 1997). It is also important that proper analytical techniques and quality control/quality assurance procedures are taken. This is beyond the scope of this paper but readers are referred to the reviews by Jenner (1996) and Kerrich and Wyman (1997).

In the following paragraphs, mafic compositions will be dealt with initially, and will be followed by felsic volcanic compositions. In all the diagrams immobile major and trace elements are utilized including Al_2O_3 and TiO_2, the high field strength elements (HFSE: Zr, Hf, Nb, Ta, Y, Sc, Ti, V), and rare earth elements (REE). These elements remain immobile in most altered and metamorphosed rocks, except under very intense alteration (e.g., chlorite alteration; Campbell et al. 1984; Whitford et al. 1988; Bau 1991; Valsami and Cann 1992), and thus provide us information on the primary petrogenetic signatures for magmatic rocks, even if they are moderately altered. A table of acronyms for geochemistry-related terms and signatures are provided in Table 1.

Mafic Geochemistry

The composition of mafic volcanic and intrusive rocks associated with VMS deposits is, in part, determined by whether they formed in a juvenile or evolved environment. In juvenile environments, deposits are preferentially associated with boninites and low-Ti tholeiites (LOTI) or mid-ocean ridge basalts (MORB) of both the normal- (N-MORB) and enriched- (E-MORB) types (e.g., Figure 2). Boninitic rocks are associated with many ophiolite-hosted (mafic) VMS deposits (e.g., Cyprus, Turner-Albright, Oman, Betts Cove) and bimodal mafic systems (e.g., Kidd Creek, Snow Lake, Rambler), and more rarely in mafic-siliciclastic systems (e.g., Fyre Lake). Boninites are characterized by high MgO (Mg#>0.60), Ni and Cr contents, low TiO_2 (<0.6%), high Al_2O_3/TiO_2 (often >40), low Ti/Sc and Ti/V ratios, and have distinctive U-shaped REE and primitive mantle normalized patterns (Figures 2-6 (e.g., Crawford, Falloon, and Green 1989; Pearce et al. 1992). Low-Ti tholeiites

Table 1: Definitions for acronyms presented within this paper.

Acronym	Definition
VMS	volcanogenic massive sulfide
ICP-MS	inductively coupled plasma mass spectrometer
HFSE	high field strength elements
REE	rare earth elements
LREE	light rare earth elements
LOTI	low Ti island arc tholeiites (or low Ti tholeiites)
MORB	mid-ocean ridge basalt
N-MORB	normal mid-ocean ridge basalt
E-MORB	enriched mid-ocean ridge basalt
OIB	ocean island basalt
BABB	back-arc basin basalt

have somewhat similar geochemical signatures to boninite but can have higher TiO_2 contents and lower Al_2O_3/TiO_2 ratios (Figure 3), and their REE and primitive mantle normalized patterns less U-shaped than boninites (Figures 2 and 6) (Brown and Jenner 1989; Kerrich et al. 1998). Boninitic rocks are interpreted to have formed from mantle sources that are ultra-depleted in incompatible trace elements and require high temperatures to melt (~1200^{o}C-1500^{o}C) (Crawford, Falloon, and Green 1989; Pearce et al. 1992; van der Laan et al. 1992; Falloon and Danyushevsky 2000). Hence, boninitic melts are typically higher temperature melts than those producing normal arc rocks. Furthermore, most boninites and LOTI are associated with forearc extension and the initiation of subduction (Stern and Bloomer 1992; Bedard et al. 1999), or with the initiation of back-arc basins (Crawford, Beccaluva, and Serri 1981; Piercey, Murphy et al. 2001); although some boninite occur in intracratonic settings (e.g., Kemp 2003).

Mid-ocean ridge basalts (MORB) are associated with many mafic-hosted VMS deposits in ophiolites and modern mid-ocean ridges (e.g., TAG, East Pacific Rise). MORB-type rocks are characterized by smooth REE and trace element patterns that are either depleted in light-REE (LREE) in the case of N-MORB, or are flat to weakly enriched in LREE in the case of E-MORB (Figure 2). MORB and MORB-like rocks with weak negative Nb anomalies on primitive mantle normalized plots are called back-arc basin basalts (BABB) and are present in many mafic-type VMS environments in modern and ancient back-arc basins (e.g., Lau Basin, Manus Basin, Semail)(Figures 3-7). In mafic and bimodal-mafic systems (e.g., forearc or back-arc settings) the MORB-type rocks often show an intimate relationship with boninitic and arc-tholeiitic rocks, with MORB either underlying boninite (e.g., Semail), or overlying and/or cross-cutting the boninite (e.g., Troodos, Rambler, Turner-Albright). In some modern back-arcs island arc tholeiites (IAT) are interlayered with BABB and MORB (Figure 2 and 7).

MORB-type rocks are also associated with mafic-siliciclastic deposits in the ancient record (e.g., Greens Creek) and at modern sedimented ridges (e.g., Middle Valley, Guaymas, and Escanaba Trough)(Figures 3-7). MORB-type rocks are interpreted to have formed from incompatible element-depleted mantle with liquidus temperatures ~1200^{o}C (e.g., McKenzie and Bickle 1988; McKenzie and O'Nions 1991; Langmuir, Klein, and Plank 1992), and represent extension either at mid-ocean ridges or within back-arc basins (e.g., Langmuir, Klein, and Plank 1992; Hawkins 1995).

In evolved environments, deposits are preferentially associated with mafic rocks that have MORB and alkalic signatures (or within-plate or ocean island basalt (OIB)) signatures (Figures 2-5 and 8). The MORB present in the evolved environments is often of E-MORB affinity and in some areas there is a complete spectrum of mafic rocks from incompatible element-depleted MORB, to weakly incompatible element-enriched E-MORB, to incompatible element-enriched OIB (e.g. Figure 8). MORB-type rocks in evolved environments are interpreted to represent depleted asthenospheric mantle that upwells beneath a rift and likely reflects the onset of seafloor spreading within a new ocean basin or back-arc basin (e.g., Barrett and Sherlock 1996; Almodóvar et al. 1997; Piercey, Paradis et al. 2002; Rogers and van Staal 2003). Commonly, the MORB-type rocks occur as sills and dykes that cross-cut mineralization, or as flows that overlie felsic rocks and the associated mineralization (i.e., they typically post-date the main mineralization event). Alkalic (OIB-like) mafic rocks are characterized by high HFSE contents (e.g., Nb, Zr), Nb/Y>0.7, elevated TiO_2 (usually >1%), low Al_2O_3/TiO_2, high Ti/V, and LREE-enriched primitive mantle normalized plots that have a positive Nb anomaly relative to Th and La (Figures 2, 3, 5, and 8). Theses types of rocks are often associated with mantle plumes but are also common of continental lithospheric mantle-derived magmas associated with continental- and continental arc-rifting (e.g., van Staal, Winchester, and Bédard 1991; Goodfellow, Cecile, and Leybourne 1995; Shinjo et al. 1999; Colpron, Logan, and Mortensen 2002; Piercey, Murphy et al. 2002). In these evolved environments, alkalic basalts typically cross-cut and overlie the main VMS hosting horizon, and typically show a stratigraphically upward progression above the VMS hosting horizon from alkalic basalts to MORB; this progression is often interpreted to reflect a shift from rifting (alkalic basalts) to true spreading (MORB)(e.g., Rogers and van Staal 2003; Piercey et al. 2004). Alkalic and MORB-type basalts are associated with many bimodal-felsic and felsic-siliciclastic settings from both the modern (e.g., Bransfield Strait, Okinawa Trough) and ancient (e.g., Bathurst, Iberian Pyrite Belt, Finlayson Lake, Eskay Creek) records (Figure 8).

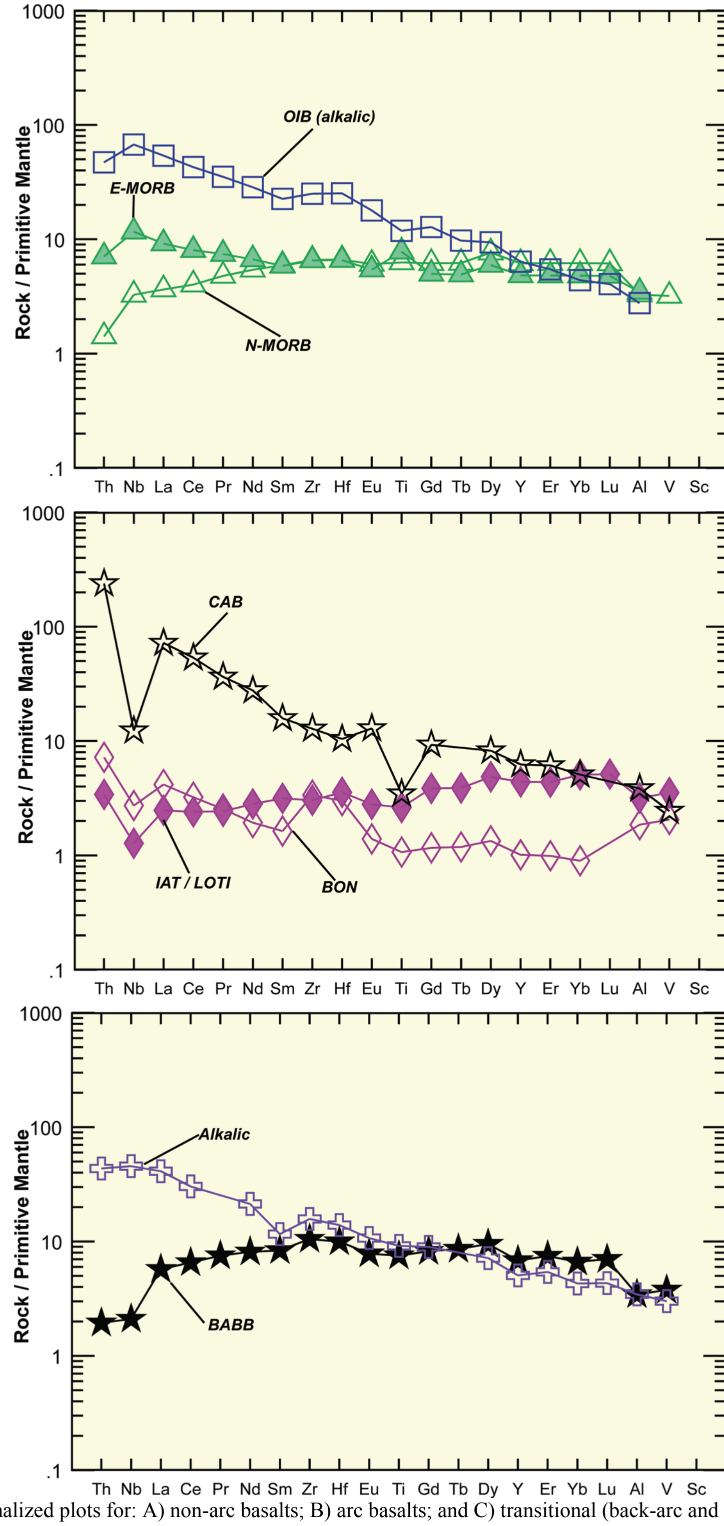

Figure 2: Primitive mantle normalized plots for: A) non-arc basalts; B) arc basalts; and C) transitional (back-arc and arc rift-related basalts). Data from Sun and McDonough (1989). Stolz (1990), Jenner (1981), Piercey et al. (2004), Ewart et al. (1994), and Kepezhinskas et al. (1997). Abbreviations: OIB = ocean island basalt; N-MORB = normal mid-ocean ridge basalt; E-MORB (enriched mid-ocean ridge basalt); CAB = calc-alkaline basalt; IAT = island ard tholeiite; LOTI = low-Ti island arc tholeiite; BON = boninite; and BABB = back-arc basin basalt. Primitive mantle values for this diagram and all others in this paper from Sun and McDonough (1989).

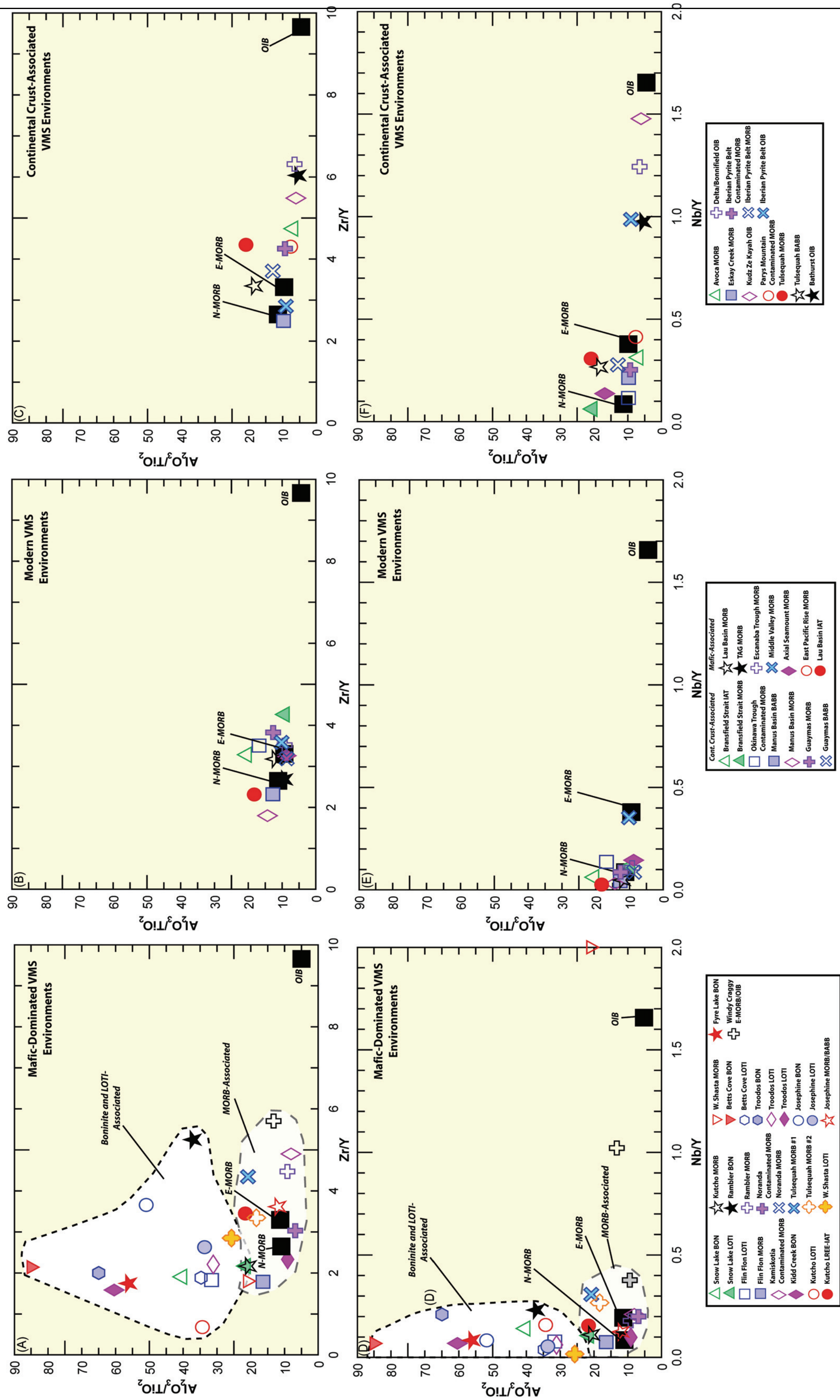

Figure 3: Al_2O_3/TiO_2 versus Zr/Y and Nb/Y for VMS-associated mafic rocks from mafic-dominated environments (a,d), modern environments (b, e), and continental-crust associated settings (c, f). Data sources are listed in Appendix 1.

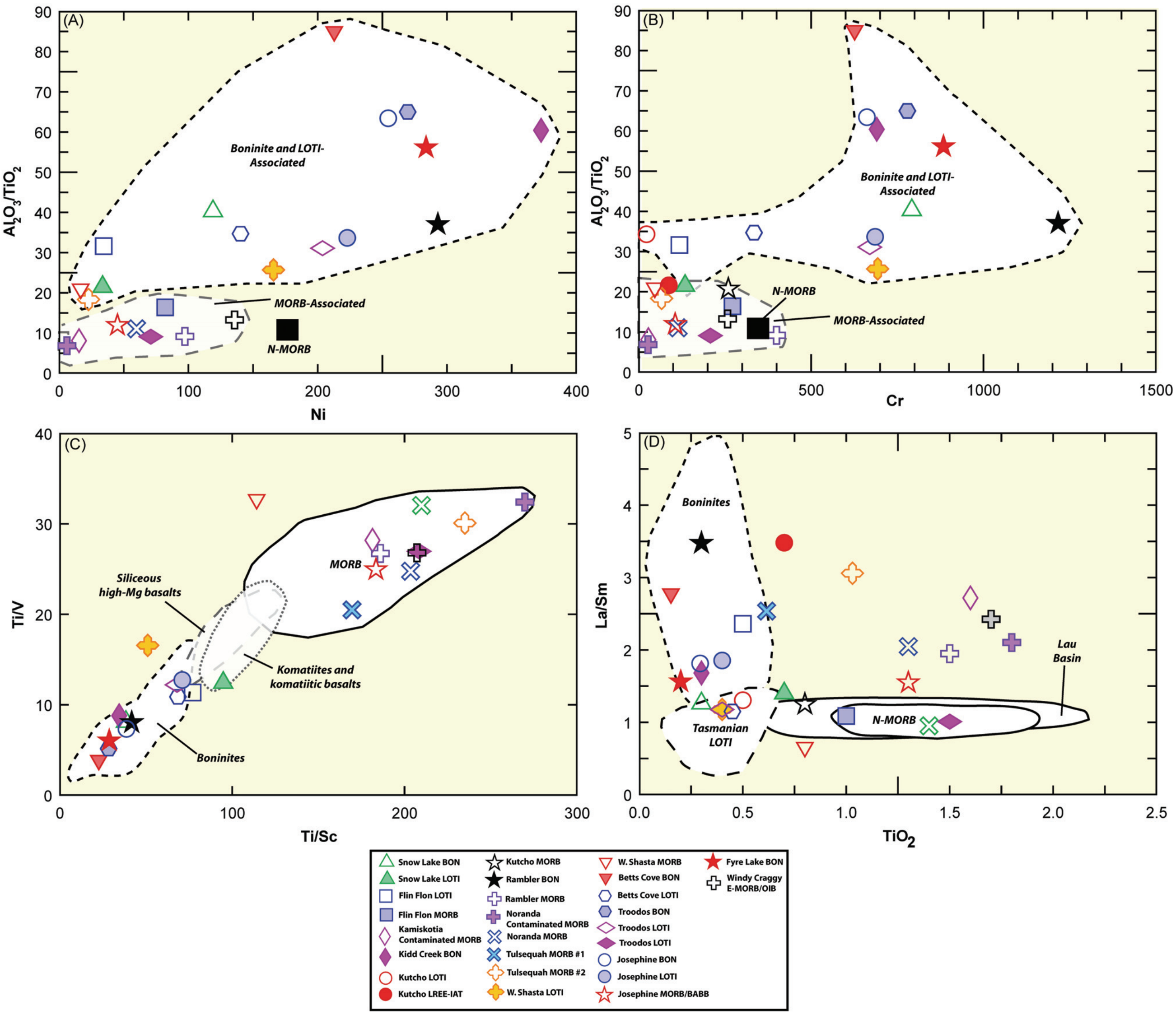

Figure 4: Diagrams for mafic rocks from mafic dominated VMS environments. A) Al_2O_3/TiO_2 versus Ni (A) and Cr (B), Ti/V-Ti/Sc (from Hickey and Frey, 1982), and (D) La/Sm-TiO_2 (from Wyman, 1999; LOTI data from Brown and Jenner (1989) and Meffre et al. (1995)). Data sources can be found in Appendix 1.

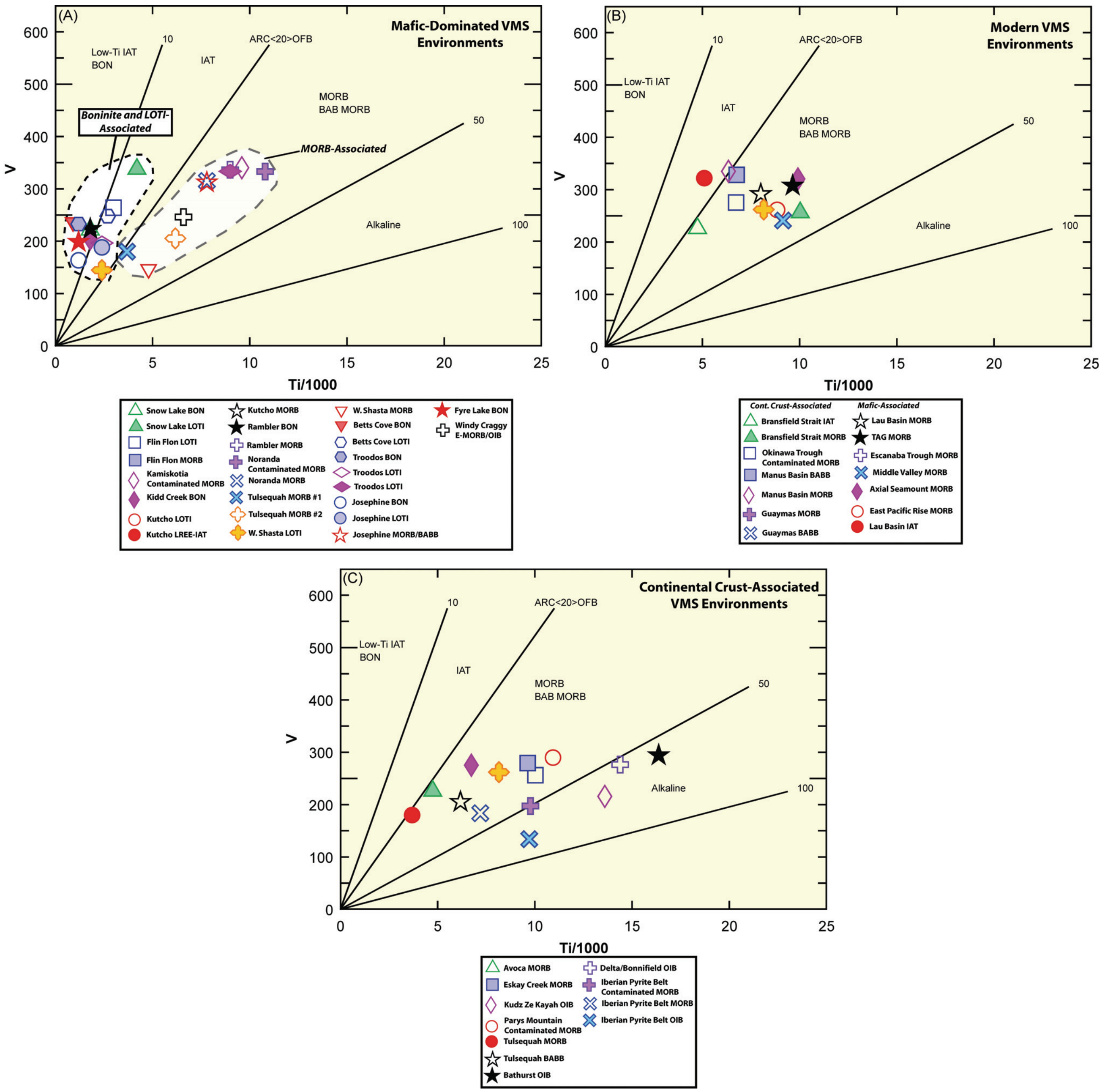

Figure 5: Ti-V discrimination diagram for mafic-dominated VMS environments (a), modern environments (b), and continental-crust associated settings (c). Data sources are listed in Appendix 1. Diagram from Shervais (1982).

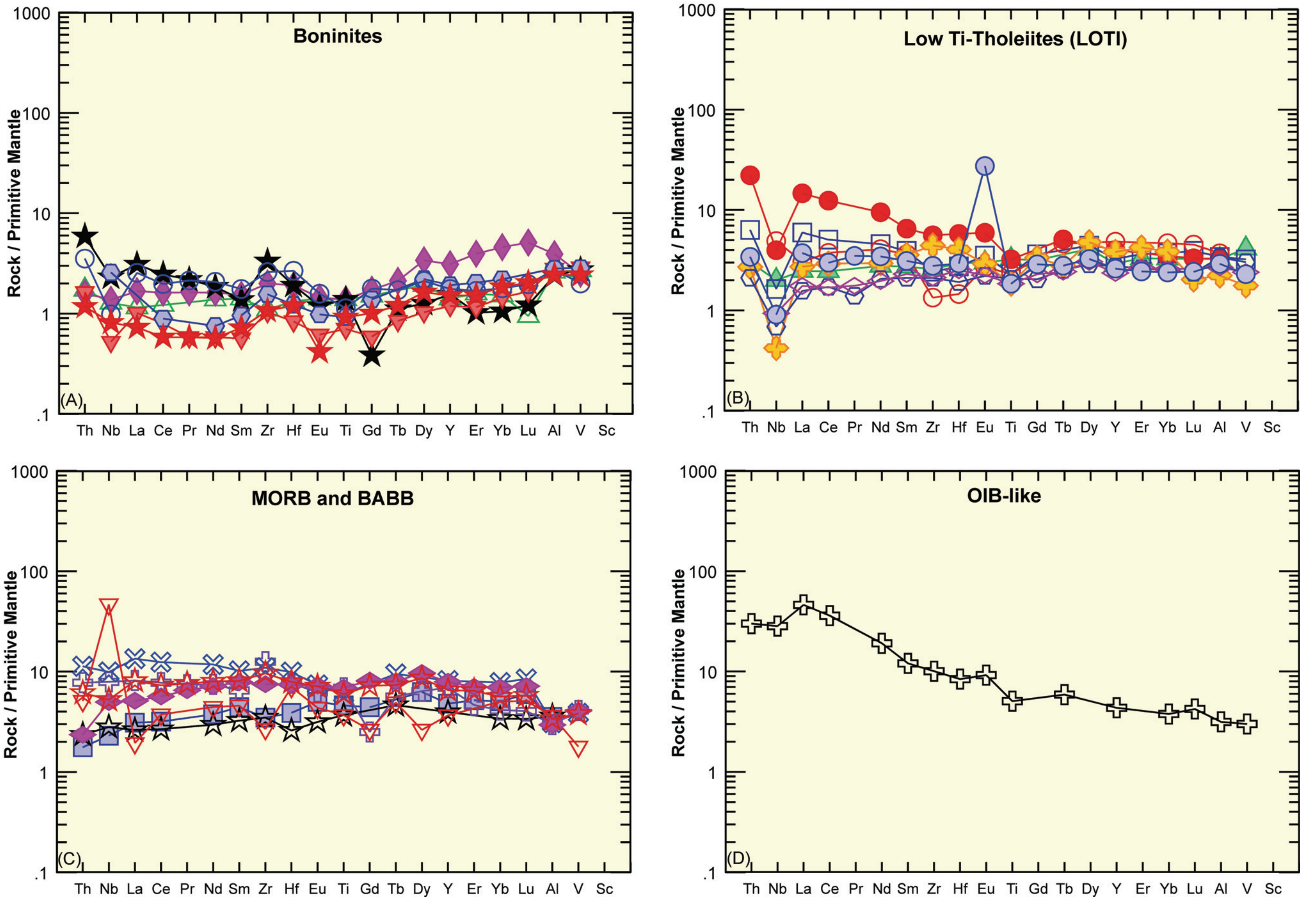

Figure 6: Primitive mantle normalized plots for mafic rocks associated with VMS deposits in mafic dominated VMS environments, including: A) boninites; B) low-Ti island arc tholeiites; C) mid-ocean ridge basalt (MORB) and back-arc basin basalts (BABB); and D) ocean island basalt (OIB)-like. Data sources in Appendix 1.

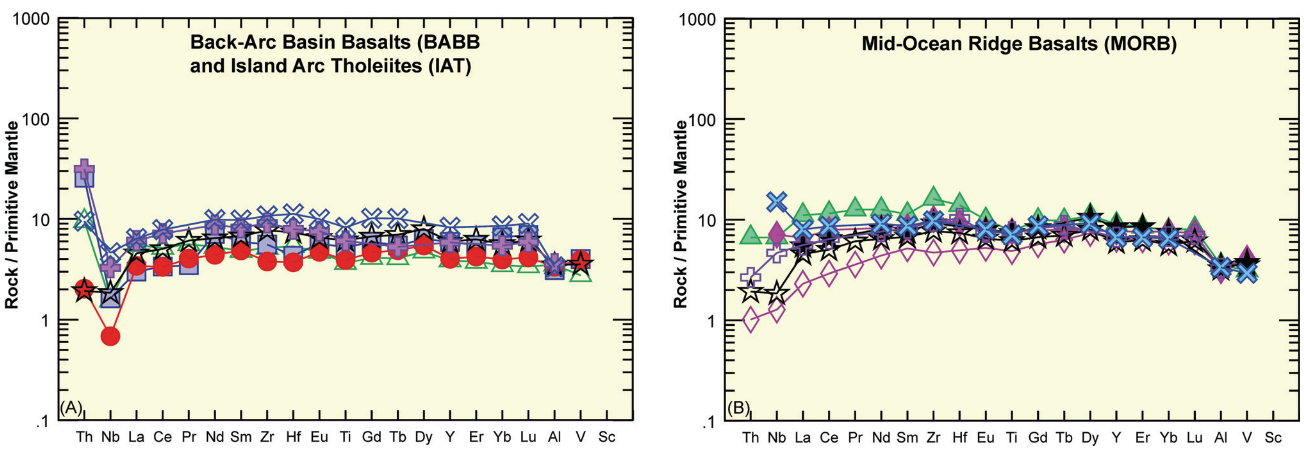

Figure 7: Primitive mantle normalized plots for mafic rocks associated with VMS deposits in modern oceans, including: A) back-arc basin basalts and island arc tholeiites; and B) mid-ocean ridge basalt (MORB). Data sources in Appendix 1. Symbols as in Figure 5.

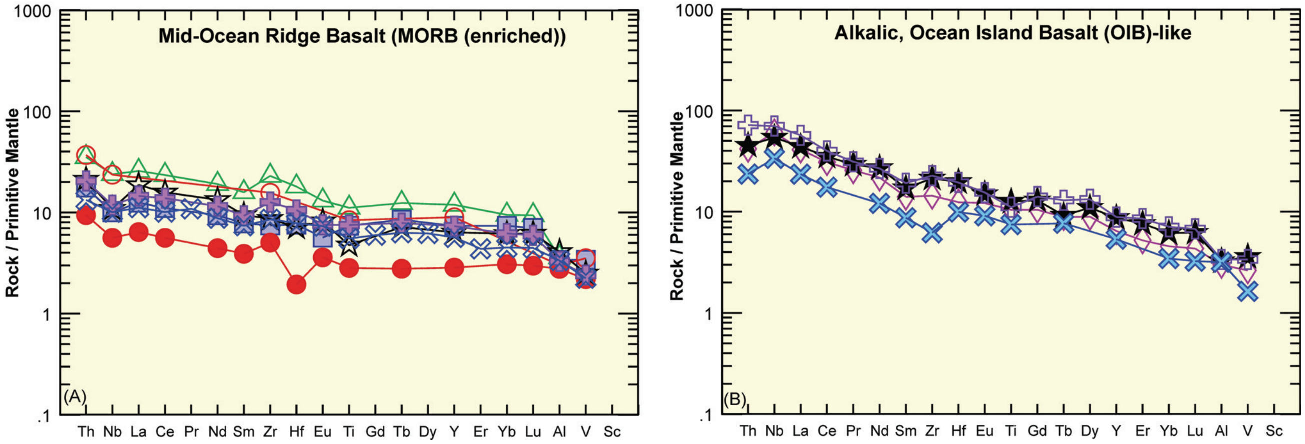

Figure 8: Primitive mantle normalized plots for mafic rocks associated with VMS deposits associated with continental crust including: A) mid-ocean ridge basalt (MORB) (note the enrichment in incompatible elements, typical of enriched-MORB); and B) alkalic, ocean island basalt-like mafic rocks. Data sources in Appendix 1. Symbols as in Figure 5.

Felsic Geochemistry

Considerable research has been undertaken on the geochemistry of felsic rocks associated with VMS systems (e.g., Lesher et al. 1986; Barrie, Ludden, and Green 1993; Lentz 1998; Hart, Gibson, and Lesher 2004). Felsic rocks formed via melting or interaction with continental crust are fundamentally different than those associated with melting a more mafic substrate, thus leading to different signatures for strata in each of these VMS environments. Furthermore, Archean felsic rocks, although similar in some cases to their younger counterparts, have signatures that are somewhat unique and must be dealt with separately from VMS-associated Proterozoic and Phanerozoic felsic rocks.

In Archean terrains considerable work has been undertaken on felsic volcanic geochemistry, particularly in the Superior Province of Canada (e.g., Lesher et al. 1986; Barrie, Ludden, and Green 1993; Prior et al. 1999; Hart, Gibson, and Lesher 2004). In these belts previous workers have outlined a tripartite subdivision of rocks, the FI to FIII suites of rhyolites (Lesher et al. 1986). This classification has been recently modified by Hart et al (2004) to include a fourth suite, FIV, but FIV felsic rocks are largely restricted to juvenile terranes in the post-Archean. The FIII suite of felsic rocks have low Zr/Y and La/Yb ratios, high HFSE contents (e.g., Zr>200 ppm), and have flat, tholeiitic chondrite-normalized REE patterns (Figures 9 and 10). The FI have high Zr/Y and La/Yb ratios, lower HFSE contents, and LREE-enriched, calc-alkalic chondrite-normalized REE patterns (Figures 9 and 10). The FII have signatures intermediate between the two groups Figures 9 and 10). The majority of Archean VMS deposits are hosted by FIII and FII felsic rocks (Figures 9 and 10), which are interpreted to have formed within Archean rift sequences from high temperature melts (T>900oC) derived from melting of hydrated basltic crust at shallow levels in the crust (e.g., Lesher et al. 1986; Barrie 1995; Hart, Gibson, and Lesher 2004). The formation at shallow depths (i.e., <10 km) allowed these melts to rise to the surficial environment without losing their heat of fusion (T>900°C), thus, giving them greater ability to drive long-lived hydrothermal systems (e.g., Barrie et al. 1999). In contrast, the other suites are interpreted to have formed from lower temperature melts (<900°C) at deeper levels in the crust (>10 km) (e.g., Lesher et al. 1986; Barrie 1995; Hart, Gibson, and Lesher 2004). These melts have less potential to drive hydrothermal systems due to their lower temperatures of fusion and loss of heat upon transport to the surface of the Earth from depth.

In Proterozoic and Phanerozoic terrains, the behavior of felsic magmatism is dependent on whether the it is associated with juvenile or evolved environments. In post-Archean evolved environments felsic rocks have a range of signatures, but most VMS deposits are associated with rhyolites that have elevated HFSE and REE contents (Figures 11-13). These rhyolites are typically FIII to FII rhyolites (Figure 11), but some deposits have FI rhyolites (e.g., Wolverine). These rocks typically have calc-alkalic chondrite-normalized trace element patterns and there is a tendency for rocks in these settings to have FII affinities (Figures 11 and 14) (e.g., Lentz 1998; Piercey, Paradis et al. 2001; Hart, Gibson, and Lesher 2004; Piercey et al. in press). Some rocks in these evolved settings, particularly those associated with continental rift or continental back-arc rifts (e.g., Delta-Bonnifield, Avoca), have rhyolites with extremely elevated HFSE contents (e.g., Zr>500 ppm; Figure 13)(e.g., Mortensen and Godwin 1982; McConnell, Stillman, and Hertogen 1991; Dusel-Bacon, Wooden, and Hopkins 2004). Rocks of these evolved environments typically have within-plate (A-type) affinities on discrimination diagrams (Figure 12). Like their Archean equivalents, felsic rocks associated with evolved settings represent high temperature (>900°C) melting of crust within rift environments (e.g., continental rift, intracontinental rifts, and continental back -arc rifts).

In post-Archean juvenile environments felsic rocks are unlike both Archean and evolved post-Archean settings. The rhyolites in juvenile environments mirror the petrology of the mafic rocks and have tholeiitic to boninite-like affinities (Figure 15-17). Tholeiitic rhyolites have low Zr/Y (<4), flat REE and primitive mantle-normalized patterns, commonly with negative Nb anomalies (not shown), are depleted in HFSE and REE (e.g., Zr<50-100 ppm), and M-type affinities on discrimination plots (Figure 15-17). Boninite-like rhyolites are similar to tholeiitic rhyolites but are more depleted in HFSE and REE with U-shaped REE patterns (Figure 17). Rhyolites from these juvenile environments have FIV affinities (Figure 15). These rhyolites typically form from melting of mafic (to andesitic) substrates often associated with forearc rifting, intra-arc rifting, or rifting during the initiation of back-arc basin activity (e.g., Shukuno et al. 2006).

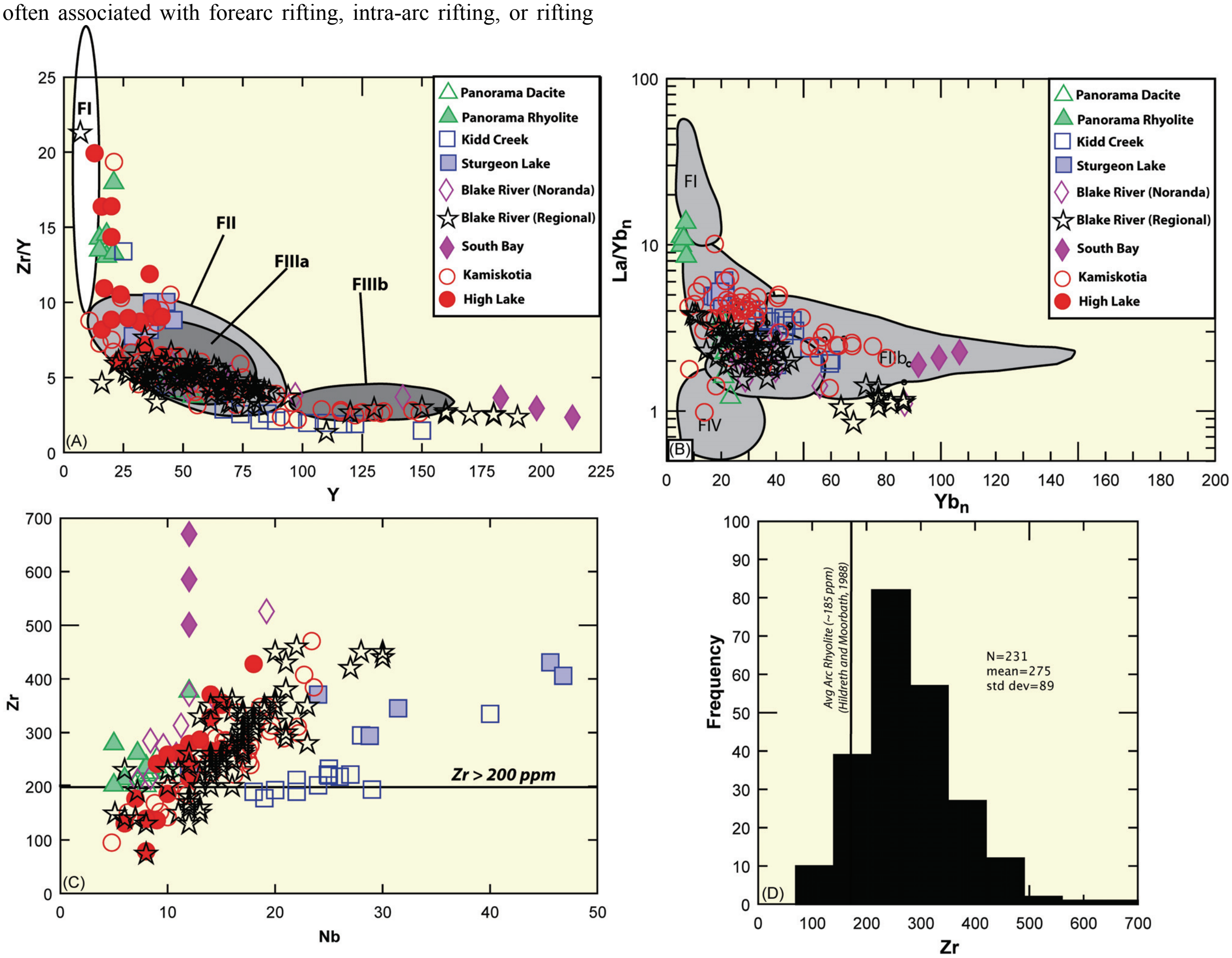

Figure 9: Key diagrams for felsic rocks associated with Archean VMS deposits. A) Zr/Y-Y diagram with FI to FIII affinities (from Lesher et al., 1986); B) La/Yb$_n$-Yb$_n$ with FI-FIII affinities (from Lesher et al., 1986 and Hart et al., 2004); C) Zr-Nb plot (from Leat et al., 1986); and D) frequency histogram of Zr for Archean rhyolites associated with VMS deposits. Note that in both (C) and (D) that most Archean VMS have felsic rocks with Zr>200 ppm. Data sources in Appendix 1.

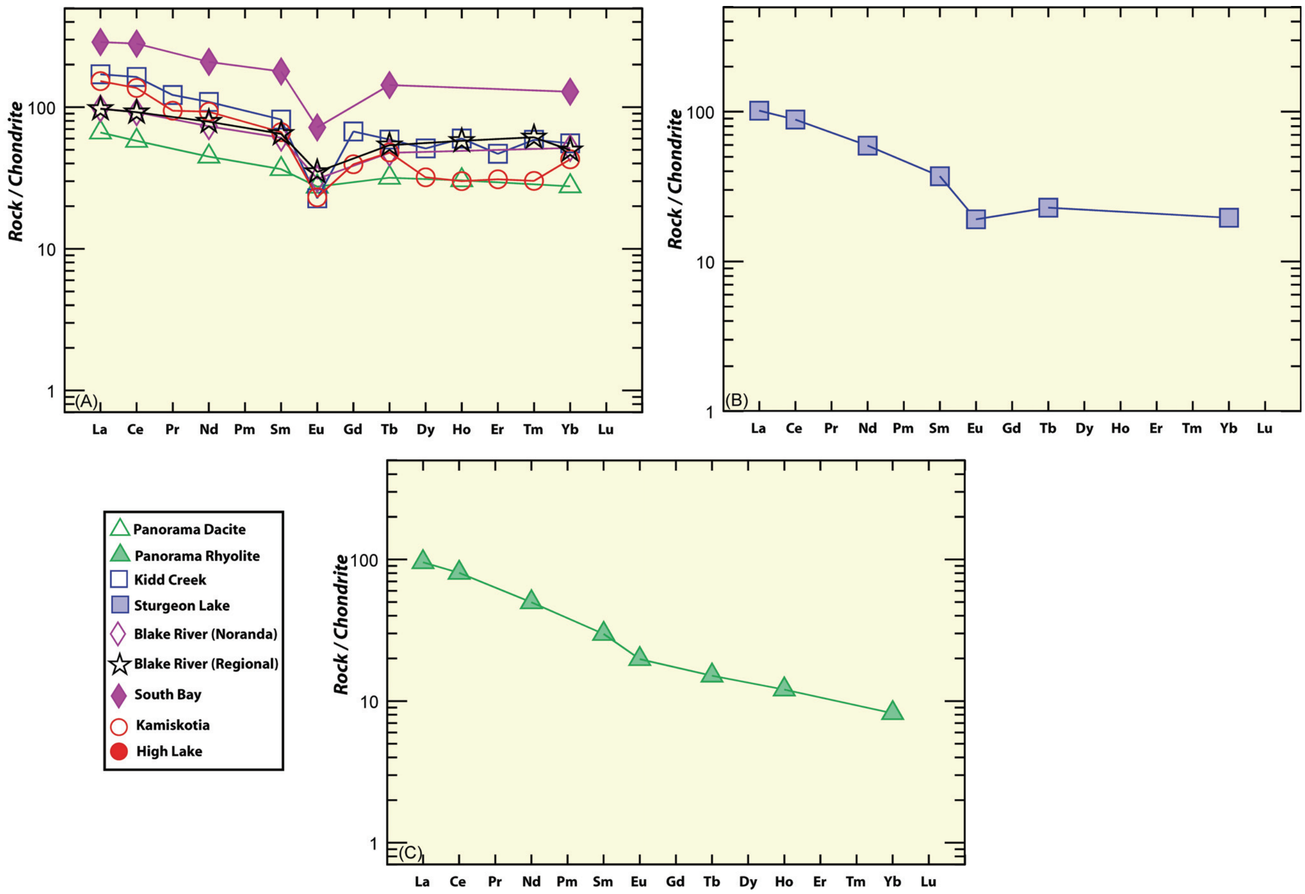

Figure 10: Chondrite-normalized REE plots for averages of: A) Archean FIII rhyolits; B) Archaen FII rhyolits; and C) Archean FI rhyolite. Data sources in Appendix 1.

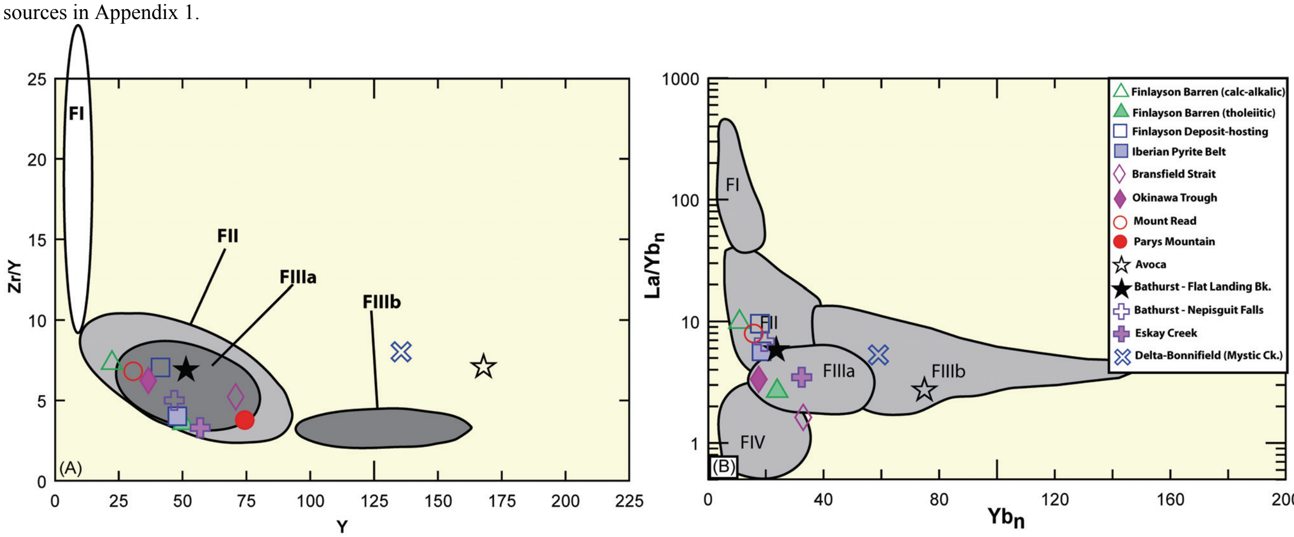

Figure 11: Rhyolite discrimination diagrams for average values for post-Archean continental crust-associated rhyolites. A) Zr/Y-Y diagram with FI to FIII affinities (from Lesher et al., 1986); and B) La/Yb$_n$-Yb$_n$ with FI-FIII affinities (from Lesher et al, 1986 and Hart et al., 2004). Data sources in Appendix 1.

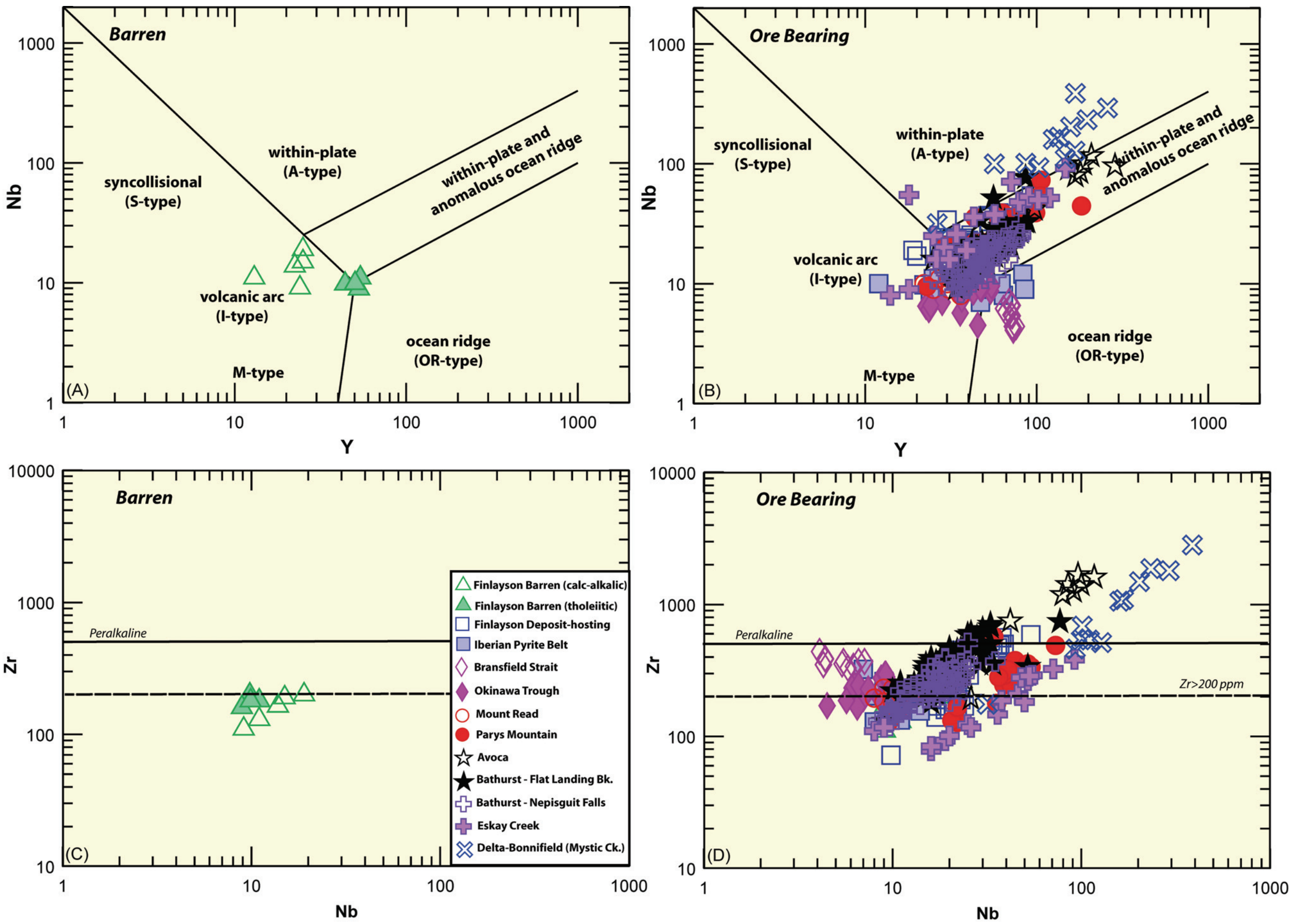

Figure 12: Tectonic distribution diagrams for post-Archean continental crust-associated rhyolites. Nb-Y discrimination diagram for A) VMS barren rhyolites and B) VMS-hosting rhyolites. Note the tendency towards within-plate (A-type) (non-arc) affinities (diagram after Pearce et al., 1984). Zr-Nb plot of A) VMS barren rhyolites and B) VMS-hosting rhyolites. Although overlapping the VMS-hosting rhyolite tend towards higher HFSE contents (e.g., Zr>20 0ppm), with some tending towards peralkalic compositions. Data sources in Appendix 1.

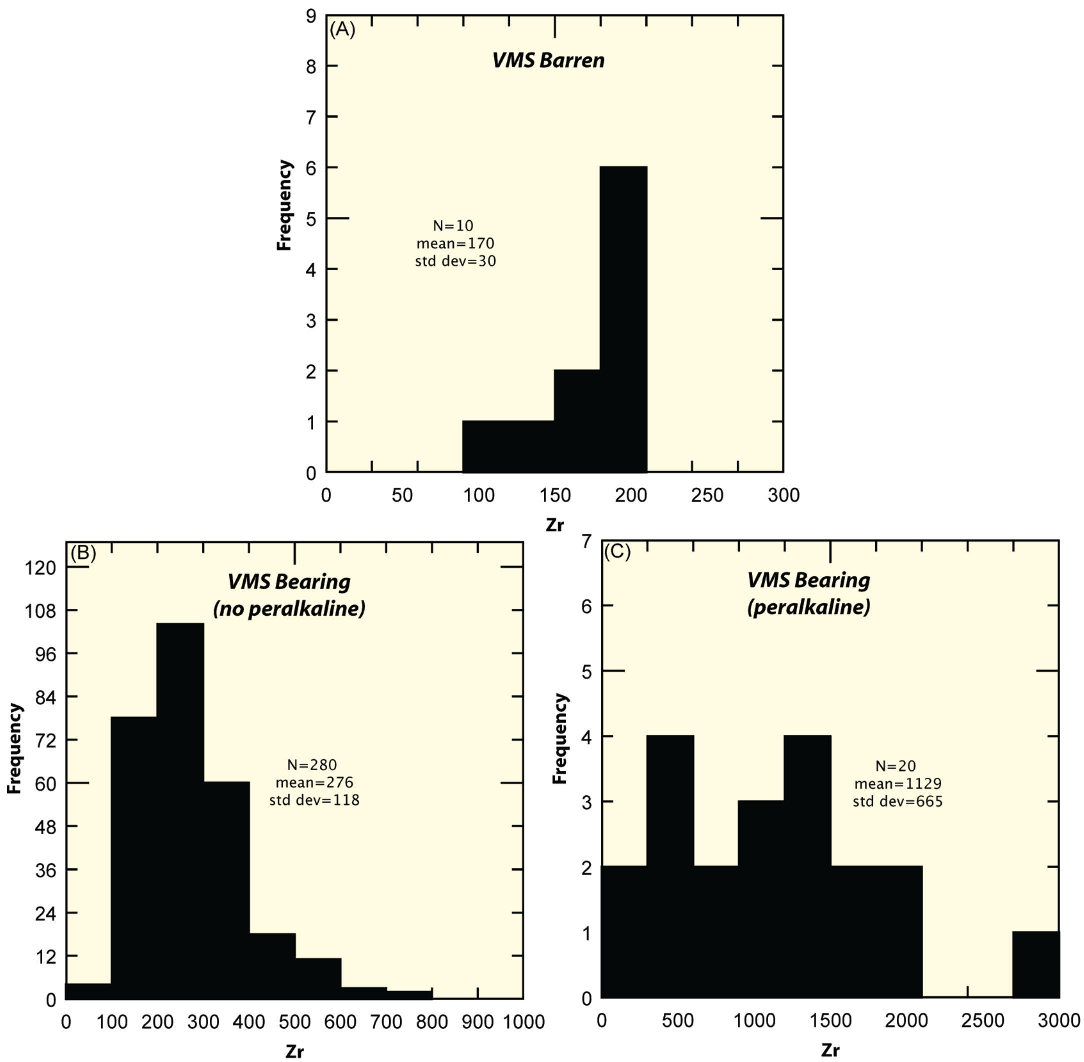

Figure 13: Frequency histograms for Zr in post-Archean rhyolites associated with continental crust for: A) VMS-barren rhyolites; B) VMS-associated rhyolites without peralkalic affinities; and C) VMS-associated rhyolites with peralkalic affinities. Note that all VMS-associated rhyolites have elevated Zr contents. Data sources in Appendix 1.

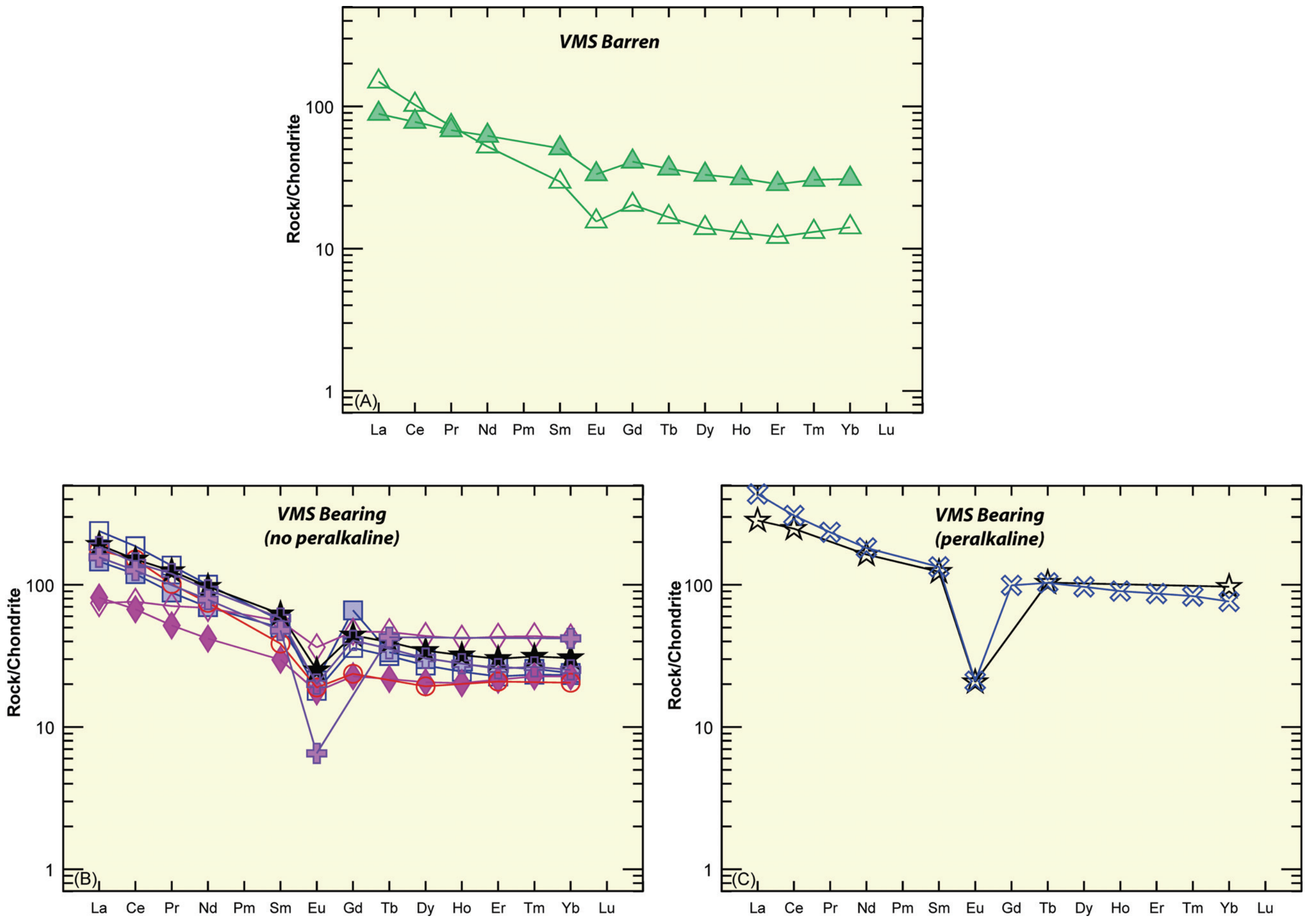

Figure 14: Chondrite-normalized rare earth element plots for post-Archean rhyolites associated with continental crust for: A) VMS-barren rhyolites; B VMS-associated rhyolites without peralkalic affinities; and C) VMS associated rhyolits with peralkalic affinities. Symbols as in Figure 12. Data sources in Appendix 1.

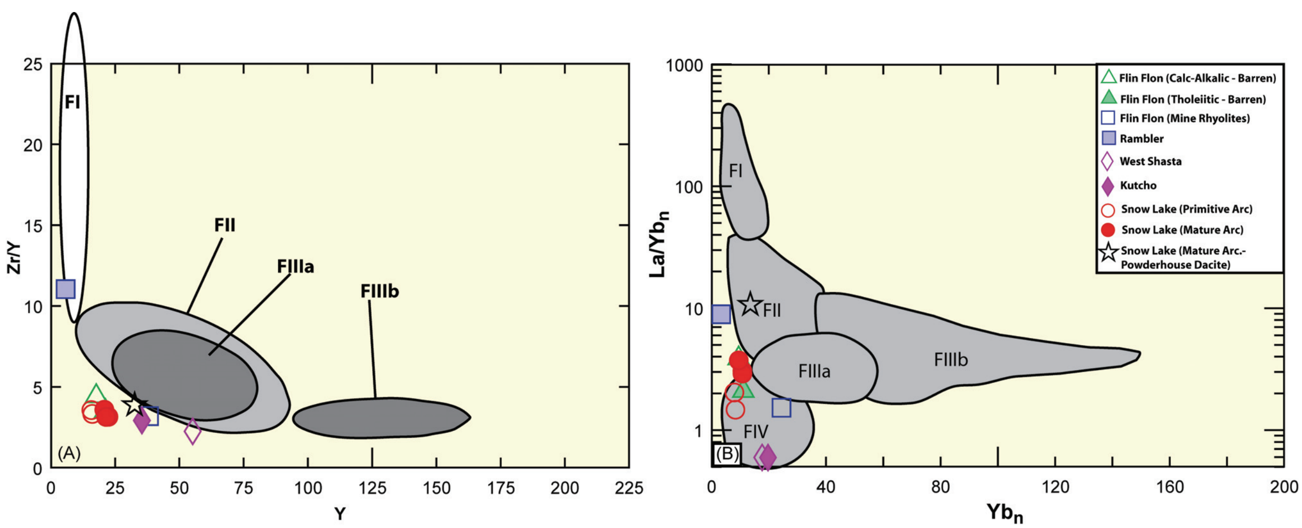

Figure 15: Rhyolite discrimination diagrams for average values for post-Archean mafic-associated rhyolites. A) Zr/Y-Y diagram with FI-FIII affinities (From Lesher et al., 1986)l and B) La/Yb$_n$-Yb$_n$ with FI=FIII affinities (from Lesher et al., 1986 and Hart et al., 2004). Data sources in Appendix 1.

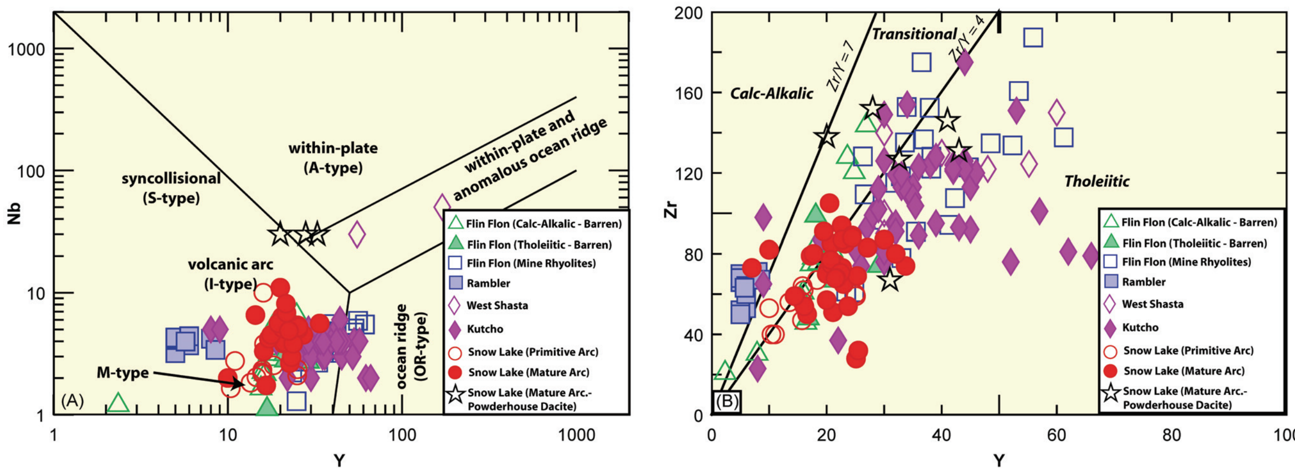

Figure 16: Discrimination diagrams for post-Archean mafic-associated rhyolites. A) Nb-Y diagram illustrating that most VMS-associated rhyolites in mafic-dominated settings have M-type affinities (diagram after Pearce et al., 1984). B) Zr-Y plot illustrating the theoleiitic character of most VMS-associated rhyolites in mafic-dominated settings (diagram modified from Barrett and Maclean (!999). Data sources in Appendix 1.

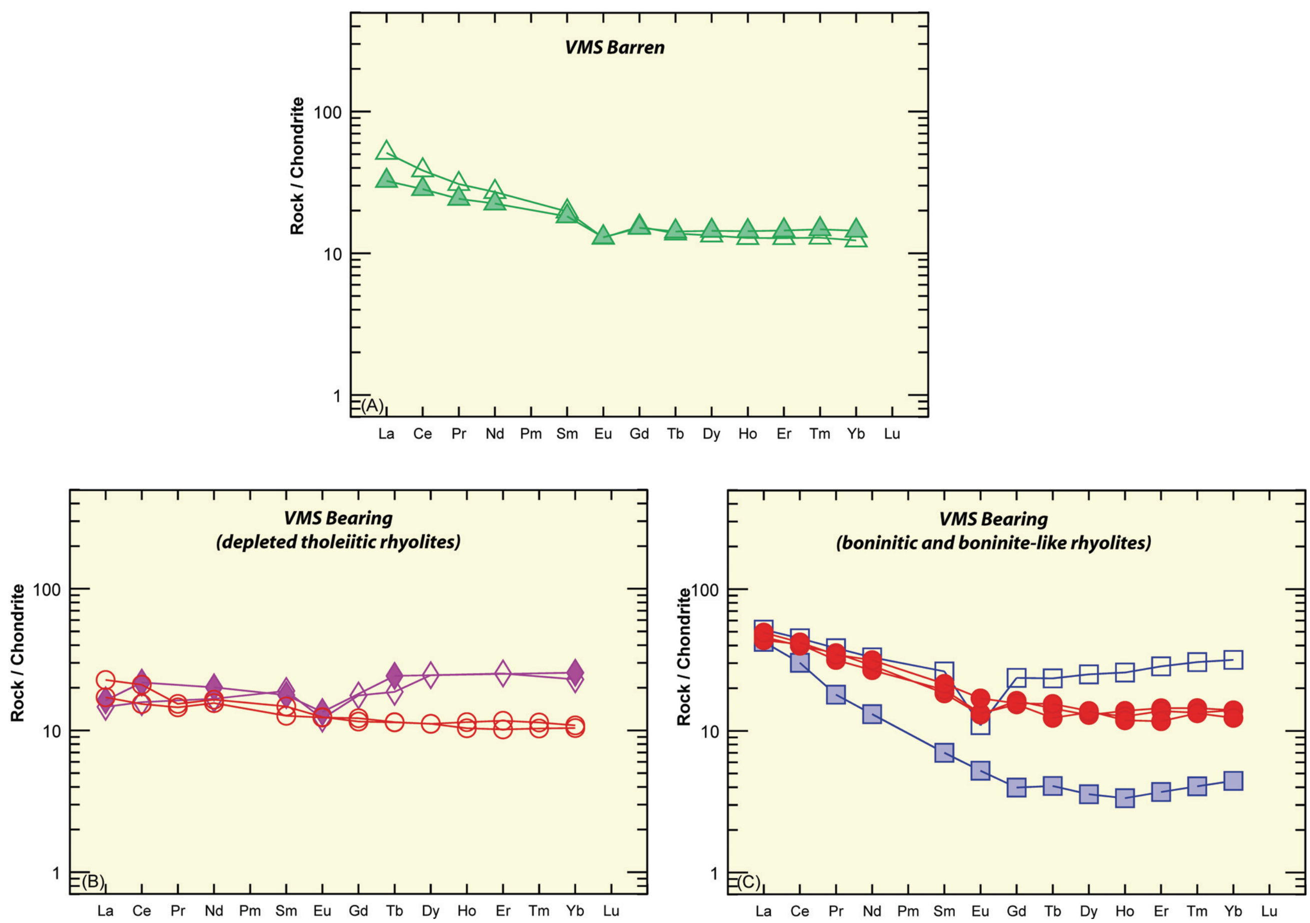

Figure 17: Chondrite-normalized rare earth element plots for average post-Archean rhyolites associated with mafic dominated settings. A) VMS-barren rhyolites; B)VMS-associated rhyolites with tholeiitic affinities; and C) VMS-associated rhyolites with boninitic affinities. Symbols as in Figure 16. Data sources in Appendix 1.

Table 2. Petrochemical assemblages of mafic and felsic rocks commonly associated with different VMS deposit classes.

VMS Deposit Class	Mafic	Felsic
Mafic	Boninite, low-Ti tholeiite, MORB	-
Mafic Siliciclastic	MORB, alkalic, boninite (rare)	-
Bimodal Mafic	MORB, boninite, low-Ti tholeiite (calc-alkalic and island arc tholeiites present but rarer)	Archean - FIII rhyolites. Proterozoic-Phanerozoic - tholeiitic rhyolites, boninitic rhyolites.
Bimodal Felsic	MORB, alkalic	HFSE-enriched rhyolites (A-type), peralkaline and calc-alkalic rhyolites (rarer)
Felsic Siliciclastic	MORB, alkalic	HFSE-enriched rhyolites, peralkaline, and calc-alkalic rhyolites (rarer)

PETROCHEMICAL ASSEMBLAGES

Very few past studies have considered coincidently the geochemical attributes of both felsic and mafic rocks; most studies focus on felsic (e.g., Lesher et al. 1986; Lentz 1998; Hart, Gibson, and Lesher 2004) or mafic (e.g., Swinden et al. 1989; Swinden 1991) rocks. In most VMS camps, however, there are specific lithogeochemical associations between mafic units in mafic-dominated settings and between mafic and felsic rocks in bimodal and felsic-dominated settings (Table 2). The groups of lithogeochemical signatures associated with different rock assemblages and deposit classifications are termed petrochemical assemblages (Table 2).

In mafic-dominated VMS environments boninites and/or LOTI are commonly hosting the VMS deposits, but are often overlain (or underlain) by MORB- or BABB-type rocks, indicative of forearc rifting or initiation of back-arc spreading (e.g. Swinden 1991; Piercey, Jenner, and Wilton 1997; Bedard et al. 1999). In mafic-siliciclastic environments the deposits are commonly associated with MORB (e.g., Escanaba Trough, Guaymas, Middle Valley), or more rarely OIB (e.g., Windy Craggy) or boninite (e.g., Fyre Lake), indicative of formation within sedimented rifts or sedimented-back-arc rifts (Saunders et al. 1982; Davis, Clague, and Friesen 1994; Stakes and Franklin 1994; Peter and Scott 1999; Piercey, Murphy et al. 2001). Plumes may have been significant in the case of the OIB-type rocks at Windy Craggy (Peter and Scott 1999).

In bimodal-mafic environments boninites and LOTI are commonly spatially associated with depleted, boninite-like or tholeiitic rhyolites with the rhyolites hosting the ores (e.g., Kerrich et al. 1998; Syme 1998; Bailes and Galley 1999; Syme et al. 1999; Wyman, Bleeker, and Kerrich 1999; Bailey 2002). These are overlain by MORB-type basaltic rocks (Piercey, Jenner, and Wilton 1997; Bailes and Galley 1999; Syme et al. 1999; Wyman, Bleeker, and Kerrich 1999; Bailey 2002). For example, in the Rambler Camp in the Newfoundland Appalachians boninite forms the footwall to the deposits, which are hosted by boninite-like rhyolites, and cross-cut by MORB dykes (e.g., Piercey, Jenner, and Wilton 1997; Bailey 2002). Similarly, in the Flin Flon camp the deposits are hosted by tholeiitic rhyolites, yet are spatially associated with LOTI, and are cross-cut and regionally associated with MORB- type rocks (Syme 1998; Syme et al. 1999). In other bimodal mafic environments the deposits are hosted by rhyolites yet the underlying mafic rocks are MORB in affinity. For example, in the Noranda camp most of the deposits are spatially associated with FIII felsic rocks, yet the bulk of the underlying stratigraphy are MORB (e.g., Lafleche, Dupuy, and Bougault 1992; Lafleche, Dupuy, and Dostal 1992; Hart, Gibson, and Lesher 2004). In all these cases, the stratigraphic sequences are indicative of formation within rift environments, either via true spreading centres (e.g., Noranda), or via a transition from normal arc volcanism to back-arc related magmatic activity (e.g., Rambler, Flin Flon). In most cases, the felsic rocks that occur within these mafic dominated environments mark the rift episode and reflect melting of the preexisting mafic-dominated substrate via mantle upwelling during the rift event (e.g., Barrie, Ludden, and Green 1993).

In bimodal felsic and felsic-siliciclastic environments felsic rocks predominate over mafic rocks with felsic rocks typically being calc-alkalic with within-plate (A-type) to peralkalic affinities (e.g., McConnell, Stillman, and Hertogen 1991; Lentz 1999; Piercey, Paradis et al. 2001; Dusel-Bacon, Wooden, and Hopkins 2004). These rocks are typically spatially associated, cross-cut, and overlain by OIB-like, alkalic basalts and/or MORB-type basalts (e.g., van Staal, Winchester, and Bédard 1991; Almodóvar et al. 1997; Piercey, Murphy et al. 2002; Piercey, Paradis et al. 2002; Rogers and van Staal 2003). For example, in the Bathurst camp the deposits are hosted by HFSE-enriched felsic rocks (Flat Landing Brook and Nepisiguit Falls formations) and cross-cut and overlain by alkalic basalts (Brunswick alkali basalts), which are overlain by MORB-type basalts (Boucher Brook formation)(Rogers and van Staal 2003). Similarly, in the Finlayson Lake district the Wolverine deposit is hosted by HFSE-enriched footwall rhyolitic tuffs and porphyries which are overlain by the MORB-type basalts (Wolverine basalts)(Piercey, Paradis et al. 2002); a similar situation exists at Eskay Creek (Barrett and Sherlock 1996). In other cases, like Avoca and the Delta-Bonnifield districts the deposits are associated with calc-alkalic and peralkalic rhyolites that are cross-cut and spatially associated with OIB-like, alkalic basalts (e.g., McConnell, Stillman, and Hertogen 1991; Dusel-Bacon, Wooden, and Hopkins 2004). In some cases, HFSE- and REE-enriched rhyolites are absent and the rhyolites have normal, calc-alkalic affinities (i.e., Zr/Y>7 but with Zr<200 ppm and volcanic-arc affinities on discrimination plots), but these rocks are cross-cut and/or overlain by OIB and/or MORB-type mafic rocks (Stolz 1995; Dusel-Bacon, Wooden, and Hopkins 2004). The occurrence of MORB and alkalic basalts in any felsic dominated setting is indicative of rifting and the upwelling of mantle beneath a continental crust-dominated substrate.

FUTURE DIRECTIONS

In the last ten years significant knowledge has been gained in understanding volcanic and intrusion geochemistry related to VMS mineralization. It is clear that rifting is an important process in localization of VMS mineralization, and identification of rifts using petrochemistry is relatively straightforward. Major challenges exist, however, in identifying the specific volcanoes within a rift that are prospective (Gibson et al., this volume). Distinguishing productive versus unproductive volcanoes will be a focus of VMS research in the next decade and will require a combination of field methods, new analytical technologies (see below), and thermodynamic modeling to better understand the interplay of tectonic, igneous, and hydrothermal processes.

Recent work has noted that Fe-Ti-P-rich andesites (icelandites) are present in the hanging wall of VMS deposits in many bimodal mafic and mafic VMS environments (e.g., Galapagos, Flin Flon, Noranda, Kamiskotia, Tambo Grande, San Nicolas, Josephine; Embley et al. 1988; Barrie and Pattison 1999; Perfit et al. 1999; Danielson 2000; Harper 2003; DeWolfe, PhD in progress; Winter, PhD in progress; Winter et al. 2004). Although minor, these units likely mark rift events there is still considerable discussion as to their origins and relationships to VMS mineralization. Research on the genesis of these Fe-Ti-P-rich andesites and their relationship to VMS mineralization will be an important avenue of research in the coming decade (e.g., DeWolfe, PhD in progress; Winter, PhD in progress).

Improvements in, and increased accessibility to, analytical technology will also likely lead to knowledge advances in the coming decade. Instruments such as the multi-collector ICP-MS will make radiogenic isotopes like Nd, Sr, Pb, and Hf more readily available to the explorationist providing an even greater understanding of the petrogenetic histories of VMS-associated and VMS-barren rocks. Similarly, techniques such as the laser ablation ICP-MS and laser ablation multi-collector ICP-MS may allow researchers to use the geochemical and isotopic signatures of resistant minerals, such as zircon, and trapped melt inclusions in igneous rocks, to predict how prospective igneous rocks and their host settings are. Further avenues of research on VMS deposits for the upcoming decade are also provided in a companion paper by Gibson et al. (this volume).

SUMMARY

Volcanic geochemistry has been and will remain to be a key tool in the delineation of prospective belts for VMS mineralization on a regional scale. It is a tool for area selection to outline prospective regions that could host VMS hydrothermal systems. In different VMS-hosting environments different groups of petrochemical assemblages are found. In mafic (e.g., ophiolitic or Cyprus-type) VMS environments, mafic rocks that host deposits are typically boninites and low-Ti island arc tholeiites , which are commonly overlain or underlain by basalt of mid-occean ridge basalt or back-arc basin basalt affinities. In bimodal-mafic environments (e.g., Noranda-type), similar mafic rock assemblages exist, but deposits are often hosted by depleted tholeiitic to boninitic rhyolites, except for in Archean environments where they are typically high field strength element-enriched rhyolites. In mafic-siliciclastic (pelitic-mafic) environments (e.g., Besshi-type), mafic rocks typically have mid-ocean ridge basalt affinities and to a lesser extent alkalic/ocean-island basalt-like (e.g., Windy Craggy) or boninitic (e.g., Fyre Lake) signatures. In bimodal-felsic environments (e.g., Kuroko-type) felsic rocks are calc-alkalic to HFSE- and REE-enriched with within-plate (A-type) to peralkalic affinities. These felsic rocks are typically cross-cut and/or overlain by mafic rocks with mid-ocean ridge basalt to alkalic/ocean-island basalt-like affinities. In felsic-siliciclastic environments (e.g., Bathurst-type or Iberian Pyrite Belt-type) felsic rocks are predominantly to HFSE- and REE-enriched with within-plate (A-type) to peralkalic affinities. These felsic rocks are typically cross-cut and/or overlain by mafic rocks mid-ocean ridge basalt to alkalic/ocean-island basalt-like affinities.

In all of these environments, regardless of setting or style, there are two common themes: 1) rifting and formation within an extensional geodynamic regime; and 2) the presence of high temperature magmatism. These two ingredients are critical in the identification of prospective environments that: 1) have the correct ground preparation to focus hydrothermal fluid flow; and 2) have sufficiently elevated geothermal gradients to drive robust and sustained hydrothermal systems. The concept of petrochemical assemblages combined with geology and lithogeochemical data well constrained by field relationships, provides a powerful tool in the delineation of potentially fertile belts for VMS mineralization.

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APPENDIX 1: DATA SOURCES

Mafic Rocks

Mafic Dominated VMS Settings

Snow Lake and Flin Flon: Stern et al. (1995); Kamiskotia: Hocker, Thurston, and Gibson (2005); Kidd Creek: Kerrich et al. (1998) and Wyman, Bleeker, and Kerrich (1999); Kutcho: Barrett, Thompson, and Sherlock (1996); Rambler/Ming: Piercey, Jenner, and Wilton (1997) and Bailey (2002); Blake River Group (Noranda): Lafleche et al. (1992; 1992); West Shasta: Brouxel et al. (1988), Bence and Taylor (1985), and Lapierre et al. (1985); Betts Cove: Bedard (1999); Troodos: Cameron (1985) and Rogers (1989); Ice Deposit: Piercey (unpublished data); Josephine (Turner Albright): Harper (2003); Fyre Lake: Piercey et al., (2001; 2004); and Windy Craggy: Peter and Scott (1999).

Modern VMS Environments

Bransfield Strait: Keller et al. (2002); Okinawa Trough: Shinjo (1999); Manus Basin: Sinton et al. (2003); Juan de Fuca (Axial

Seamount): Rhodes, Morgan, and Liias (1990); East Pacific Rise: Allan Batiza, and Lonsdale (1987); Middle Valley: Stakes and Franklin (1994); Lau Basin: Ewart et al. (1994); TAG hydrothermal field (Mid-Atlantic): Smith and Humphris (1998); Escanaba Trough: Saunders et al. (1982); Guaymas: Davis and Clague (1987).

Continental Crust-Associated VMS Settings

Avoca: Leat et al. (1986) and McConnell, Stillman, and Hertogen (1991); Eskay Creek: Barrett and Sherlock (1996); Kudz Ze Kayah (Finlayson Lake): Piercey et al. (2002); Parys Mountain: Barrett, MacLean, and Tennant (2001); Tulsequah: Sebert and Barrett (1996); Bathurst: Rogers and van Staal (2003); Delta-Bonnifield: Dusel Bacon, Wooden, and Hopkins (2004); Iberian Pyrite Belt: Almadovar et al. (1997) and Mitjavila, Marti, and Soriano (1997).

Felsic Rocks

Archean Felsic Rocks

Pilbara: Vearncombe and Kerrich (1999); Kidd Creek: Prior et al. (1999); Sturgeon Lake: Lesher et al. (1986); Noranda: Lesher et al. (1986) and Peloquin (1999)(regional); South Bay: Lesher et al. (1986); Kamiskotia: Hart (1984), Barrie and Pattison (1999); and High Lake: Petch (2004).

Post-Archean Felsic Rocks from Mafic-Dominated Settings

Flin Flon: Syme (1998); Rambler (Ming): Bailey (2002) and Piercey, Jenner, and Wilton, (1997); West Shasta: Bence and Taylor (1985) and Lapierre et al. (1985); Kutcho: Barrett, Thompson, and Sherlock (1996); and Snow Lake: Bailes and Galley (1999; 2001).

Post-Archean Felsic Rocks from Continental Crust-Dominated Settings

Eskay Creek: Barrett and Sherlock (1996); Delta-Bonnifield: Dusel-Bacon, Wooden, and Hopkins (2004); Finlayson Lake: Piercey et al. (2001); Iberian Pyrite Belt: (1997); Bransfield Strait: Petersen et al. (2004); Okinawa Trough: Shinjo and Kato (2000); Mount Read: Crawford, Corbett, and Everard (1992); Parys Mountain: Barrett, MacLean, and Tennant (2001); Avoca: Leat et al. (1986) and McConnell, Stillman, and Hertogen (1991); and Bathurst: Rogers et al. (2003).