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Magnetite as an indicator mineral in the exploration of porphyry deposits: a case study in till near the Mount Polley Cu-Au deposit, British Columbia, Canada

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Abstract

We investigated if the composition of hydrothermal ore-related magnetite in till could be used to locate porphyry deposits in terrain where glacial overburden overlies rocks that host porphyry Cu-Au mineralization. We test this hypothesis using 20 till samples collected in an ~900 km^2 area surrounding the Mount Polley porphyry Cu-Au deposit, in south-central British Columbia, Canada. At least 100 magnetite grains were randomly selected from the magnetic fraction of each till sample. Nineteen trace elements in ~50 magnetite grains in each sample were measured by Laser Ablation-Inductively Coupled Plasma-Mass Spectrometry (LA-ICP-MS). The large beam or raster size used for laser ablation (to 100 μm) homogenizes any heterogeneous trace element distributions in magnetite that result from oxy-exsolution and/or and dissolution/re-precipitation, avoiding this issue with the few micron size of an electron beam. Linear discriminant analysis (LDA) performed on a compilation of magnetite compositions measured by LA-ICP-MS from worldwide porphyry deposits and intrusive igneous rocks define the chemical signature (Mg, Al, Ti, V, Mn, Co, Ni) of hydrothermal magnetite exclusive to porphyry systems. Application of our LDA models to 985 magnetite compositions we measured in the till samples surrounding Mount Polley showed anomalous amounts of hydrothermal magnetite grains in till up to 2.5 km west-southwest and 4 km northwest of

the deposit – a pattern that is consistent with the ice-flow history of the region. Our LDA model for magnetite trace element compositions has strong potential to be an effective tool in exploration for buried porphyry systems.

Introduction

Indicator mineralogy is a powerful tool in exploration and has had proven success in locating a variety of mineral deposits including diamondiferous kimberlite pipes and base metal sulphides (Averill, 2001). For porphyry Cu(-Au) systems, indicator minerals have included a large variety of accessory and alteration minerals that occur in these deposits such as apatite, epidote, garnet, jarosite, rutile and tourmaline, and ore-related minerals such as pyrite, chalcopyrite and gold (Averill, 2011; Celis et al., 2014; Kelley et al., 2011; Hashmi et al, 2015). Magnetite chemistry has been applied in sedimentary provenance studies (Grigsby, 1990; Razjigaeva & Naumova, 1992; Yang et al., 2009), and as a resistate mineral ubiquitous in porphyry systems, shows potential as an indicator mineral in exploration (Dupuis & Beaudoin, 2011; Nadoll et al., 2012; Dare et al., 2014).

Magnetite (Fe_3O_4) is a member of the spinel group of minerals that are recognized for their systematic variations in composition as a function of temperature, cooling rate, fO_2, or silica activity (Buddington & Lindsley, 1964; Irvine, 1965; Sack & Ghiorso, 1991; Roeder, 1994; Barnes & Roeder, 2001; Dare et al., 2014). Several studies suggest that variation in minor and trace element contents in magnetite is systematically related to the type of mineralizing environment, including volcanogenic massive sulphide (VMS), skarn, Fe-oxide-Cu-Au (IOCG) and porphyry Cu(-Mo-Au) (McQueen & Cross, 1998; Kamvong et al., 2007; Dupuis & Beaudoin, 2011; Nadoll et al., 2012, 2014, 2015; Huang et al., 2014; Dare et al., 2012; 2014; Chen et al., 2015; Makvandi et al., 2015; Canil et al., 2016).

Despite a dearth of experimental data for the partition of trace elements in magnetite crystallized from fluid (Ilton and Eugster, 1989; Simon et al, 2004), several studies have used an empirical approach to fingerprint the chemical signature of magnetite from various hydrothermal environments. Dupuis & Beaudoin (2011) developed discrimination diagrams for a number of deposit types including IOCG, Kiruna, banded iron formation, porphyry Cu, skarn, Fe-Ti-V, Ni-Cu-PGE, and volcanogenic massive sulphide. Nadoll et al. (2012, 2014) used factor analysis to identify factors that are able to discriminate between igneous, low-temperature hydrothermal and metamorphic magnetite, identifying Mg, Al, Ti, V, Mn, Co, Zn and Ga as key elements. They determined that hydrothermal magnetite has elevated values for the Mg-Mn factor, whereas igneous magnetite can be identified by high values of Co-Ni-V. Nadoll et al. (2015) calculated discriminant measures on magnetite from porphyry Cu and skarn deposits, as well as their respective host rocks, in the southwestern United States. Dare et al. (2014) used data from a variety of hydrothermal ore deposits and igneous rocks to broadly distinguish hydrothermal from igneous magnetite using Ti, Ni and Cr. Canil et al. (2016) used principal components analysis on hydrothermal magnetite from porphyry Cu(-Au-Mo) and skarn deposits to recognize positive correlations of Al, Ti and V related to temperature, and negative correlations of Sn and Mo, with Mn and Co, governed by fluid chemistry. Few studies have examined magnetite as an indicator in till proximal to an ore deposit (McMartin et al., 2011; Sappin et al., 2014). There have been no ground-truth tests of magnetite around a known porphyry deposit.

In this paper, we directly test the potential of magnetite trace element chemistry as an indicator for porphyry Cu(-Au) deposits. We first use multivariate statistical analysis to rigorously define the signature of hydrothermal magnetite from mineralized porphyry systems. We then apply our discrimination methods to 987 magnetite compositions we measured in till surrounding the Mount Polley porphyry Cu-Au deposit, British Columbia, Canada (Fig. 1) to show the veracity and plausibility of using trace elements in magnetite as an exploration tool.

Geological Setting

The Mount Polley porphyry Cu-Au deposit occurs in the Quesnel Terrane, an assemblage of Late Triassic to Early Jurassic arc rocks in the North American Cordillera (Fig. 1). The intrusive complex hosting the Mount Polley deposit is a high-level, northwest trending alkalic stock, approximately 4 by 6 km in dimension, emplaced in metasedimentary and metavolcanic rocks of the Nicola Group that form an open northwest-plunging syncline (Fig. 2; Logan & Mihalynuk, 2005; Rees, 2013). The intrusive complex is composed of fine-grained porphyritic diorite and monzonite with subordinate plagioclase porphyry and syenite dikes with crystallization ages of 205±3 Ma based on the U-Pb zircon method

(Mortensen et al., 1995; Logan & Mihalynuk, 2005; Logan et al., 2007). Various magmatic-hydrothermal breccias are the major host for Cu-Au mineralization, which occurs as disseminated and stockwork chalcopyrite, bornite and pyrite (Fraser et al., 1992; Rees et al., 2013). The age of porphyry Cu-Au mineralization was determined by Logan et al. (2007) as 205.2 ± 1.2 Ma based on $^{40}Ar/^{39}Ar$ ages of hydrothermal biotite intergrown with chalcopyrite.

The Late Wisconsinan Fraser Glaciation deposited a large amount of glacial sediment over the Quesnel Terrane and much of the Interior Plateau. Ice build-up of the Cordilleran Ice Sheet began ~29 ka, with complete deglaciation by ~11.5 ka (Ryder et al., 1991). Ice-flow indicators as measured from erosional features on outcrop indicate two directions of ice-flow movement in the Mount Polley region: 1) early westward ice-flow (oriented at 250-275°) during advancement of valley and piedmont glaciers from the Cariboo Mountains in the east; and 2) later north-westward ice-flow (oriented at 290-330°) during the last glacial maximum related to an ice divide to the south of Mount Polley at 52°N latitude (Plouffe et al., 2013a). The NW-trending direction is the dominant ice-flow movement recorded in the study area (Hashmi et al., 2015).

Sampling and Analytical Methods

Till Samples

A total of 74 till samples from an area of ~900 km^2 surrounding the Mount Polley deposit gathered as part of a Natural Resources of Canada Targeted Geoscience Initiative 4 program (Plouffe et al., 2013b; Ferbey et al., 2014) were the basis for our study (Fig. 2). For each sampling location, ~10 kg of till was collected at a depth of ~80 cm below the soil weathering horizon. Bulk till samples were prepared at Overburden Drilling Management Ltd. (Nepean, Canada) by first wet sieving to a <2 mm size fraction and then concentrating by density on a shaking table. The recovered heavy minerals were further concentrated in methylene iodide (specific gravity 3.2) and sieved into three size fractions: 1-2 mm, 0.5-1 mm, and 0.25-0.5 mm. Magnetic separation was performed on the heavy mineral fraction of the processed till samples. Magnetite grains (2 to 0.25 mm) were picked from the ferromagnetic heavy mineral fraction using a hand magnet and binocular microscope at the University of Victoria (Victoria, Canada). Approximately 100-125 grains per till sample were randomly selected, mounted in epoxy and polished for further study (Pisiak et al., 2015). The composition of magnetite from both mineralized and barren rocks in the Mount Polley deposit were previously investigated by Canil et al. (2016).

Petrography

Reflected light microscopy was used to examine internal textures of the magnetite. Magnetite grains were classified according to their crystal form, state of preservation (e.g., intensity of pitting, alteration or fracturing), exsolution textures, and abundance of mineral inclusions. A subset of magnetite grains in each sample was further examined with a Hitachi S-4800 field emission scanning electron microscope (SEM) at the University of Victoria using an accelerating voltage of 16 kV and emission current of 20 nA. Back-scattered electron (BSE) imaging and semi-quantitative energy dispersive x-ray spectrometry (EDS) were used to identify exsolution and alteration phases, and inclusions.

Electron Microprobe Analysis

The concentrations of Mg, Al, Si, Ca, Ti, V, Cr, Mn, Fe and Ni in magnetite from a subset of the till samples were determined using a CAMECA SX 50 electron microprobe (EMP) at the University of British Columbia (Vancouver, Canada). The EMP was operated at an accelerating voltage of 15 kV, beam current of 20 nA and spot size of 5 μm. Counting times of 20 sec were used for all elements except V, Cr and Ni (40 sec). Less than 5% of all analyses for most elements in magnetite were below the detection limits of the EMP, except Cr and Ni for which ~30% of all analyses were below detection.

Laser Ablation-Inductively Coupled Plasma-Mass Spectrometry (LA-ICP-MS)

Fifty magnetite grains per till sample were randomly selected for elemental analysis by LA-ICP-MS at the University of Victoria. Grains with appreciable alteration, fracturing or a significant number of inclusions were excluded because they would not be amenable to ablation without obvious contamination of other phases. Analysis of Mg, Al, Si, Ca, Sc, Ti, V, Mn, Cr, Co, Ni, Cu, Zn, Ga, Nb, Mo, Sn, Ta and W was performed using a 213 nm Nd: YAG UV laser operating at 50-63% energy and 10 Hz, and interfaced to a Thermo Scientific XSERIES 2 Quadrupole ICP-MS.

Laser ablation using spots (25-55 μm) and rastered lines (100-150 μm) were employed depending on the mineral size and texture (Fig. 3). Data collection for each analysis included 30 sec of background signal followed by 20 to 30 sec of ablation, with minimum of 60 sec washout. External calibration using standard reference material silicate glasses NIST611, NIST613 and NIST615 was performed for every 10 analyses. Time-resolved spectra were processed off-line using a custom spreadsheet. Obvious mineral inclusions at the surface were avoided for ablation (Fig. 3), but heterogeneities in time-resolved spectra, caused by small inclusions intersected at depth by the laser, were edited to remove contaminated sections from the spectra. When such heterogeneities were too large and/or frequent in the spectra, the analysis was discarded.

Three different methods of standardization were employed: 1) internal standardization using an assumed stoichiometric amount of Fe in pure Fe_3O_4 magnetite (i.e., 72 wt.% Fe); 2) internal standardization using the Fe concentration as determined by EMP analysis; and 3) the ablation yield correction factor (AYCF) method of Liu et al. (2008). The AYCF method is an internal standard-independent calibration that corrects for the absolute amount of material ablated and calibrates to the total counts per second for each analysis assuming that all elements in the sample have been analyzed (Liu et al., 2008). For magnetite, the elements selected for LA-ICP-MS analysis cover the range of known substitutions in magnetite that occur in measurable concentrations and, therefore, safely satisfies this assumption. A comparison of the three different standardization methods is discussed further below.

Accuracy, precision and detection limits were monitored over time in each analytical session using the USGS basaltic glass reference BCR2g (Table 1). Accuracy for Mg, Si, Ca, Sc, Ti, V, Mn, Co, Ni, Nb, Mo and W is better than 5%. Elements Cr, Fe and Ta have accuracies between 5 and 10%; Al, Cu and Sn between 10 and 20%; and Zn and Ga at 20 to 30%. Precision was better than 20% for all elements except Cr, Zn, Ga and W. Detection limits are reported using the 3σ criterion, whereby the limit of detection for each element is calculated using three times the standard deviation of the gas background signal that contains zero analyte (Longerich et al., 1996).

Petrography

Magnetite grains in till exhibit a wide variety of morphologies, textures, alteration and inclusions (Fig. 3). In the till samples, magnetite grains vary in shape from subhedral-euhedral octahedrons and cubo-octahedrons, to more commonly rounded to sub-rounded forms, which likely result from sediment transport. Some magnetite grains are highly fractured or strongly pitted, whereas others are relatively pristine or homogeneous (Fig. 3A). In reflected light, magnetite is typically gray with a brownish tint, but is often characterized by exsolution of a slightly lower reflective phase. Exsolved phases of ilmenite ($FeTiO_3$) or, very rarely, ulvöspinel (Fe_2TiO_4) and/or rutile (TiO_2) typically occur as trellis-type lamellae ranging from submicroscopic to 10 μm in width (Figs. 3, 4). Parallel or sandwich-type lamellae are also observed, but are much less common. Some magnetite grains contain more than one exsolution phase. For example, Figure 4A shows thick ulvöspinel + rutile lamellae with thin interstitial trellis replaced by an unknown Si-Al-Mg phase in magnetite. Ilmenite exsolution occurs by subsolidus oxidation during cooling, whereas ulvöspinel exsolution occurs from cooling alone below 600°C (Haggerty, 1991).

Two predominant types of alteration are present in magnetite from till: 1) martitization (hematite alteration) due to oxidation, and 2) ‘sphenitization’ by late stage fluids (Haggerty, 1976). Martitization is identified in reflected light, and predominantly occurs along grain boundaries, crystallographic planes (lamellae), adjacent to fractures, or as localized patches, with no preference for any specific textural or morphological type of magnetite (Fig. 3). In comparison, alteration of magnetite to titanite (‘sphenitization’) is highly variable and in some cases cryptic. In exsolved magnetite, titanite is a replacement product of Ti-rich lamellae (Fig. 4C), forming after oxy-exsolution, whereas in magnetite grains without exsolution, titanite occurs as irregular stringers or fine disseminations (Fig. 4D). In addition, titanite may form along grain boundaries or as localized patches (Fig. 3D), similar to hematite. Mineral inclusions may be present and/or abundant in some magnetite grains (Figs. 3A, C). Apatite is the most common type of inclusion followed by quartz, K-feldspar and chalcopyrite in decreasing abundance. Magnetite grains containing multiple generations of coarser-grained magnetite (> 200 μm) recognized as dissolution/re-precipitation during hydrothermal processes in skarn deposits (Hu et al, 2015), were rare in the total population, and not recognized in any of the 985 magnetite grains analyzed. Nevertheless, as will be shown in what follows, only a subset of the till magnetites are likely hydrothermal in origin.

Chemistry of Magnetite

Comparison of Analytical Methods

The complete compositional dataset for all magnetite grains in till (n = 985) is given in an online Appendix Table A1. We tested the accuracy of three calibration methods for the processing of magnetite LA-ICP-MS spectra, in order to optimize its application in mineral exploration. The 'EMP Fe' calibration method uses the Fe concentration in magnetite as determined by EMP as the internal standard for LA ICPMS spectra, and is the most accurate. To obviate the step of obtaining EMP data, the 'stoichiometric Fe' calibration method assumes the maximum stoichiometric amount of Fe that can occur in magnetite (72%) as the internal standard and, therefore, is an approximation that produces the maximum possible trace element concentrations. The third type of calibration, the ablation yield correction factor (AYCF) method, calibrates to the total counts per second in each analysis and, therefore, requires no internal standard, and thus no prior step of EMP analysis for Fe.

For all major and minor elements in magnetite, the 'stoichiometric Fe' calibration method produced consistently higher concentrations (by 13 – 35%) than the 'EMP Fe' method (Fig. 5A). In contrast, the AYCF method produces element concentrations within only a few percent of that calculated using the 'EMP Fe' calibration method (Fig. 5B) with a maximum difference observed of 20-25%. The low mass elements Mg, Al, Si and Ca could not be compared between standardization methods because spurious concentrations arise when calibrated using Fe as an internal standard, possibly due to the low concentration of Fe in the NIST reference materials used for standards. On the other hand, the calibration curves for Mg, Al, Si and Ca using the internal standardless AYCF method are well correlated and considered reliable. Thus, the AYCF method is preferred over either internal standardization methods by providing accurate calibration of all elements in magnetite while eliminating the time for an additional analytical step in determining the concentration of the internal standard.

Elements such as Mg, Al, Ti, V, Cr and Mn occur in significant concentrations in magnetite and are often detectable by both EMP and LA-ICP-MS. Other elements that occur in much lower concentrations (<0.1 wt.%) such as Si, Ca, Cr and Ni, however, are often near or below detection for the EMP (Fig. 6A). Significant scatter observed for certain elements may be an artifact of the difference in beam size between the EMP and LA-ICP-MS (Fig. 6B). For example, extensive exsolution is observed in a large proportion of the magnetite grains in this study (Figs. 3, 4) and the lamellae are often at scales coarser than the electron beam size (5 µm). The smaller beam size of EMP is less able to homogenize entire bulk composition during analysis in this case (Dare et al, 2012). This difference in sampling would contribute significantly to differences in concentrations for elements that are heterogeneously distributed in magnetite, such as Ti, which can be concentrated in exsolution lamellae (Fig. 6B). A larger beam size is possible for EMP, but the rastering capability of the laser is more efficient for homogenizing material during analysis. In what follows, we use element concentrations determined by LA-ICP-MS analysis rather than EMP because of the greater sensitivity and lower detection limits of the former instrument, and the fact a laser spot or raster homogenizes more of the crystal during analysis and is, therefore, a better representation of the total primary magnetite composition before exsolution occurred.

"Sphenitization": High Si and Ca in Magnetite

A large number of magnetite grains sampled from tills in the Mount Polley region are characterized by elevated concentrations of Si and Ca, up to 7.3 and 6.0 wt.%, respectively. Both Si and Ca do not have extensive substitutions in spinel group minerals from the Earth's crust (O'Neill & Navrotsky, 1984). A number of studies have documented several weight percent Si in magnetite from various environments with Ca typically <1 wt.% (Vincent & Phillips, 1954; Shcheka et al., 1977; Newberry et al., 1982; Shiga, 1988; Westendorp et al., 1991; Dare et al., 2014). The high Si contents could be attributed to a Fe_2SiO_4-equivalent spinel component in solid solution with magnetite rather than contamination by minute silicate impurities (e.g., Newberry et al., 1982; Westendorp et al., 1991). Alternatively, Shcheka et al. (1977) suggested variable amounts of a Ca_2SiO_4 spinel component in magnetite based on similarly high Ca contents. The majority of Si-bearing magnetite in the aforementioned studies is described as zoned or containing discrete domains in which Si is concentrated, features that are not observed in magnetite from our till samples.

Most magnetite grains in till that are enriched in Si and Ca are characterized by approximately equal atomic proportions of these two elements (Fig. 7). This is inconsistent with substitution as a Ca_2SiO_4 spinel component, but is the expected 1:1 stoichiometry of Si and Ca in titanite ($CaTiSiO_5$), which is observed petrographically in many magnetite grains on a variety of scales (Fig. 3, 4). Using the Si and Ca contents, the maximum amount of titanite alteration in magnetite is 33% (Fig. 8).

Sphenitization of magnetite can occur by metasomatic introduction of SiO_2 and CaO in late stage H_2O-rich fluids or by the breakdown of plagioclase, followed by the removal of FeO from magnetite to form other minerals in the rock such as sulphide, chlorite or amphibole (Haggerty, 1976). This phenomenon has been documented in metasomatized intrusive rocks (Weisse et al., 1985), I- and S-type granites (Whalen & Chappell, 1988) and hydrothermally altered oceanic crust (Shau et al., 2000). Weisse et al. (1985) report preferential titanite alteration of ilmenite lamellae in magnetite from the Nahant gabbro, United States, similar to that observed in till near Mount Polley (Fig. 4C). Haggerty (1976) documents examples of titanite replacement for both exsolved lamellae and host magnetite.

In five porphyry and skarn deposits of British Columbia, sphenitization was observed petrographically in very few samples. The chemistry of magnetite from Mount Polley (< 0.3 wt.% Si, < 0.05 wt.% Ca) confirms that this is not a characteristic of the deposit (Canil et al, 2016). In fact, greater degrees of sphenitization in magnetite in till samples were observed at greater distances down-ice or away from the Mount Polley deposit. As sphenitization is relatively common during regional, low-grade metamorphism (Haggerty, 1976), grains in the till with any appreciable amount of this effect are likely sourced from the metamorphosed volcanic and sedimentary rocks of the Late Triassic Nicola Group that dominate the study area (Fig. 2) rather than being hydrothermal in origin and specific to the Mount Polley porphyry deposit.

Compositional Variability in Magnetite from Till

The large compositional variability observed in the detrital magnetite grains reflects the diverse bedrock sources for till in the Mount Polley region. For brevity, we describe only the salient relationships between a few of the trace elements. Titanium is the most variable of all elements measured in magnetite, ranging from 50 ppm to 13.5 wt.%, and shows a strong positive correlation with Al, which ranges up to 7.6 wt.% (Fig. 8A). The Ti and Al contents of magnetite are related to temperature (Buddington & Lindsley, 1964; Turnock & Eugster, 1962). Therefore, elevated Ti and Al concentrations in most magnetite grains in till is consistent with a large proportion of material derived from igneous source rock, likely the volcanic rocks of the Nicola Group that cover most of the study area (Fig. 2). In contrast, hydrothermal magnetite typically contains substantially lower Ti and Al than magnetite in igneous rocks (Fig. 8A - Dupuis & Beaudoin, 2011; Dare et al., 2014; Nadoll et al., 2014; Canil et al., 2016).

Trivalent V and Cr have maximum concentrations of 0.7 and 8.1 wt.%, respectively. High V contents are typical of magmatic magnetite (Nadoll et al., 2014); however, magnetite at the Mount Polley deposit also contains significantly high V (Fig. 8B). Magnetite with elevated Cr concentrations occurs in serpentinized mafic-ultramafic rocks as ‘ferritchromit’, a product of hydrothermally altered Cr-Al spinels (e.g. Mellini et al., 2005). Chromium-rich magnetite could be derived from serpentinites of the Slide Mountain Terrane that outcrop in the far northeast corner of the study area (Fig. 2). Elevated concentrations of Si and Ca related to secondary sphenitization are also observed in some grains (Fig. 7).

Scandium and Sn are measurable in nearly all magnetite grains but are <77 and 12 ppm, respectively. Tin concentrations are positively correlated with Mo and Sc in hydrothermal magnetite, but not in the variety of magnetite in the tills (Fig. 8 C,D). Other minor elements that are detectable in most magnetite grains include Ni (max. 3021 ppm), Cu (max. 1807 ppm), Co (max. 759 ppm) and Ga (max. 213 ppm). Gallium is weakly correlated with Mn (100- 30,000 ppm) and Zn (30 – 10,000 ppm), suggesting minor components of $MnGa_2O_4$ and $ZnGa_2O_4$. More than 10% of all analyses were below detection for Nb, Mo, Ta and W for which the maximum contents of each element is less than ~150 ppm.

Identification of Hydrothermal Magnetite

Binary Plots

Dupuis & Beadoin (2011) developed two diagrams to distinguish magnetite compositions of porphyry, iron-oxide-Cu-Au (IOCG), skarn, Kiruna, banded iron formation (BIF), and Fe-Ti-V deposits. Their Ni/(Cr+Mn) vs. Ti+V plot is unable to accurately classify magnetite from porphyry deposits in the southwest United States and British Columbia (Fig. 9). Tests of a second discrimination diagram of Ca+Al+Mn vs. Ti+V found that porphyry magnetite from till and bedrock at Mount Polley partly coincides with the porphyry field (Fig. 10), but also extends into the fields for IOCG, skarn, Kiruna and Fe-Ti-V deposits (Nadoll et al. 2014; Canil et al, 2016). The latter diagram is very problematic because Ca+Al+Mn values will be grossly modified by sphenitization, which we documented in several magnetite grains in till sourced from low-grade metamorphosed bedrock (Fig. 7). The Al+Mn vs. Ti+V diagram of Nadoll et al. (2014, 2015) define fields for distinguishing skarn and porphyry deposits from the southwest United States. With the exception of one sample from Mount Polley, magnetite from the deposit is accurately classified as porphyry (Fig. 11); however, magnetite from several unmineralized igneous intrusions (Mountjoy, 2011) and all magnetite from till are similarly classified as porphyry. Finally, a plot of Ti vs. Ni/Cr in magnetite proposed by Dare et al. (2014) broadly distinguishes hydrothermal from magmatic grains. Hydrothermal porphyry magnetite from Mount Polley (Canil et al. 2016) falls within the hydrothermal field on this plot (Fig. 12). Chromium is typically in very low concentrations in hydrothermal magnetite (i.e., <150 ppm at Mount Polley). Therefore, for a large number of analyses of magnetite from Mount Polley and a small number of magnetite grains in till, the detection limit for Cr was used, producing a minimum Ni/Cr value. Magnetite from the till samples primarily plots in the igneous field, with a small proportion of grains overlapping into the hydrothermal magnetite field (Fig. 12).

Because the discrimination diagrams shown in Figures 9, 10 and 11 are unable to properly identify hydrothermal magnetite from Mount Polley, these diagrams would be too general or ineffective in practice for an exploration program. The Ti vs. Ni/Cr diagram is straightforward and shows potential for application of magnetite in exploration for porphyry deposits (Fig. 12), but hydrothermal magnetite is poor in Cr and detection limits will introduce practical uncertainty. Hu et al. (2015) also contest the validity of these diagram in Figures 9, 10 and 11, because EMP analysis (on which some the diagrams are based) is prone to the problem of a small beam size in sampling the larger heterogeneities in hydrothermal magnetite. We show laser ablation, particularly with rastering, can sample at a scale beyond most heterogeneities, and integrate the total composition of magnetite (irrespective of the origin) and still avoid obvious mineral inclusions at the surface (Fig. 3). Nevertheless, there may be too wide of a compositional range in hydrothermal magnetite to be accurately classified using simple binary diagrams.

Discriminant Analysis

A multivariate statistical method that applies the collective significance of all elements, rather than just a few, may be a more robust approach to characterize the composition of magnetite as an indicator mineral. We used discriminant analysis (Davis, 2002) to test for differences between magnetite from various sources. We compiled a database of trace element concentrations in magnetite (n = 481) from three petrologically distinct groups to create a training dataset for the discriminant analysis (Table 2). Hydrothermal porphyry magnetite (HTP) was designated as that occurring in veins and hydrothermal breccias, thus showing direct petrographic evidence of precipitation from fluids. Ore igneous magnetite (OIG) is eu-subhedral and occurs in fresh or altered intrusive rocks located at a porphyry deposit. Barren igneous magnetite (BIG) is eu-subhedral, but from unmineralized intrusions that are remote from or show no relationship to any kind of ore deposit. The database was then divided into two subsets based on the whether the analytical data was measured all in one lab, or in different labs.

For brevity here, the extended procedures and details of the discriminant analyses are given in the Appendix 2 and elsewhere (Pisiak, 2015). We determined the elements that have the most discriminating power among magnetite compositions from these three groups (HTP, OIG, BIG) to define discriminant functions that uniquely identify the provenance of magnetite grains. A linear method of discriminant analysis (LDA) was chosen due to the simplicity of linear functions. Table 3 summarizes the results of the two discriminant analyses, subsequently referred to as LDA1 and LDA2 for the two different subsets of data, described in detail in Appendix 2. The discriminant scores for the two analyses show a clear separation of the magnetite compositional groups on a territorial plot of the linear functions (Fig. 13a,b). Factor 1 (F1) effectively separates hydrothermal magnetite (HTP) from the ore-related igneous (OIG) and barren igneous (BIG) groups, with hydrothermal magnetite generally characterized by positive F1 values. Coefficients of the standardized discriminant functions show the relative importance of each variable on the respective function (Table 3). The greatest influence on F1 are Mg and Al, suggesting these two elements have the strongest discriminating power for separating hydrothermal from igneous magnetite. Factor 2 (F2) effectively separates ore and barren igneous magnetite, with the latter tending towards positive F2 values.

Differences in the LDA1 and LDA2 models are mostly affected by Mn, Ni and V in F2, but with a smaller influence of Mn and V in LDA2. Slight variation is expected between the analyses because of the difference in variables selected. Minor overlap is observed between compositional fields, and is slightly more pronounced in LDA2 (Fig. 13b). Nevertheless, the derived compositional fields from both models show strong potential for identifying magnetite provenance in mineral indicator studies. Our approach shows a classification accuracy of 91 and 78% for the LDA1 and LDA2 models, respectively (Table 4), which suggests that this classification method can be confidently applied to magnetite compositions of unknown provenance as an exploration tool.

Application to the Mount Polley Porphyry Cu-Au Deposit

Hydrothermal Magnetite in Till

The classification methods for magnetite composition derived from discriminant analysis and binary plots discussed above were directly tested using till samples surrounding the Mount Polley porphyry deposit as a ground-truth evaluation. For a total of 20 till samples distributed up-ice, down-ice and remote from the deposit, 50 magnetite grains were analyzed in each sample to determine if hydrothermal magnetite from the deposit could be successfully identified in till (Fig. 2).

For each till sample, magnetite grains were assigned a discriminant score using each of the two sets of linear functions derived for LDA1 and LDA2. A territorial plot of F1 vs. F2 allowed for classification of magnetite grains into one of the three pre-defined groups (Fig. 13 c,d) and for the relative percentage of hydrothermal grains in each till sample from the area (Table 5). The discriminant functions of LDA1 identified 71 hydrothermal porphyry magnetite grains in the area, whereas those of LDA2 identified 93 grains. Most magnetite grains from each classification plot in the ore igneous (OIG) compositional field using both LDA1 and LDA2 functions, suggesting that the discrimination of ore from barren igneous magnetite is not well constrained.

Magnetite compositions from till were also plotted on the discriminant diagrams proposed in the literature as described above. The diagrams of Dupuis & Beaudoin

(2011) were unable to correctly classify magnetite from Mount Polley and could not identify a sufficient number of porphyry magnetite grains in till (Figs. 9,10). The Al+Mn vs. Ti+V diagram of Nadoll et al. (2015) was only useful in distinguishing porphyry from skarn magnetite, with no distinction provided for igneous magmatic magnetite (Fig. 11). The Ti vs. Ni/Cr diagram of Dare et al. (2014) correctly identified all magnetite samples from Mount Polley as hydrothermal. When the discriminant diagram was applied to magnetite from till, 93 hydrothermal magnetite grains were identified, which is similar to the classification by the LDA2 model (Fig. 12, Table 10C).

The proportion of hydrothermal grains estimated in each till sample are generally consistent between the LDA1 and LDA2 models, with the latter yielding slightly higher percentages in most samples (Fig. 14a). In contrast, there is less agreement between the LDA models and the Ti-Ni/Cr discrimination diagram (Figs. 14 b,c). A significant number of grains that were classified as hydrothermal by the Dare et al. (2014) plot were not similarly identified as hydrothermal in our discriminant analyses.

The textures of hydrothermal magnetite grains were also examined to determine if they were potentially useful as a petrographic discrimination tool. The majority of hydrothermal magnetite grains in till showed no exsolution, but the proportion varied with the classification method: 58% for LDA1 and 66% for LDA2. Although hydrothermal magnetite typically contains relatively low Ti contents, ilmenite exsolution lamellae in magnetite from porphyry systems have been documented at several localities (e.g., Celis et al., 2014; Nadoll et al., 2015; Canil et al., 2016). Additionally, magmatic magnetite from rapidly cooled volcanic rocks is commonly free of exsolution (e.g., Frost & Lindsley, 1991; Grigsby, 1990; Dare et al., 2014). The absence of exsolution in magnetite is not a reliable indication of its provenance from hydrothermal porphyry systems. Other textural features such as the presence of inclusions or alteration assemblage were not directly linked to magnetite classification.

Mineral Indicator Maps for Till Surrounding Mount Polley

The percentage of hydrothermal magnetite grains in each till sample surrounding the Mount Polley porphyry deposit as determined by LDA1, LDA2 and the Ti-Ni/Cr discriminant diagram (Table 5) are plotted in Figures 15 - 17. Samples having populations of >10% hydrothermal magnetite grains were deemed anomalies, whereas samples below this amount were considered to be the regional background signal.

Classification of magnetite by the LDA1 discriminant functions identified six till samples with anomalous concentrations of hydrothermal magnetite grains (Table 10). Five of these samples were similarly identified with the LDA2 classification and occur along a trend that begins immediately southwest of the deposit with the furthest anomalous sample approximately 4 km northwest of Mount Polley (Figs. 15, 16). To the west, the anomaly is detected ~2.5 km from the deposit. Till samples at 5 km and further afar from the deposit contain hydrothermal magnetite counts within the background range.

Northwest transport of hydrothermal magnetite from the Mount Polley deposit is consistent with the dominant ice-flow direction in the study area and documented using several ore-related and pathfinder elements, and other ore and alteration minerals (Hashmi et al., 2015). The transport of magnetite grains several kilometers west-southwest of the deposit is also consistent with the observations of Hashmi et al. (2015), who found chalcopyrite, gold, and epidote grains in till samples west-southwest of the deposit. This bidirectional transport of material is characteristic of glacial dispersal whereby material was initially transported by the earlier ice-flow movement to the west-southwest during glacial advancement from the Cariboo Mountains, followed by reworking and transport of debris by ice-flow to the northwest during the glacial maximum (Parent et al., 1996).

The Ti vs. Ni/Cr method of magnetite classification (Fig. 12) identified the same quantity of hydrothermal magnetite grains as the LDA2 discriminant functions, but spread over a larger number of till samples (Table 5). Using this discrimination method, an apparent bidirectional detrital glacial transport from the deposit to the west and northwest remains, but the dispersal of hydrothermal magnetite grains over a larger number of samples produces a more diffuse anomaly compared to those from the LDA classifications (Fig. 17). The four till samples with the greatest proportion of hydrothermal magnetite grains identified by the Ti-Ni/Cr diagram occur on a trend west from the deposit to a distance of 5 km, much further than that determined by the LDA classifications, and by Hashmi et al. (2015) for chalcopyrite, gold and epidote in till.

Hydrothermal magnetite grains would be primarily transported to the northwest based on the dominant ice-flow trajectory at Mount Polley. In contrast, classification using the Ti-Ni/Cr diagram shows hydrothermal magnetite from the deposit has been transported greater distances to the west than to the northwest, conflicting other glacial evidence from the region (Hashmi et al., 2015).

All till samples with anomalous concentrations of hydrothermal magnetite grains can be directly traced to the Mount Polley porphyry deposit based on ice-flow directions in the study area, with the exception of one till sample that occurs 5 km north of the deposit (Fig. 15,16). This sample (12 TFE 102 A01) contains 12% hydrothermal magnetite grains identified by both LDA1 and the Ti vs. Ni/Cr discrimination methods (Table 5). Hydrothermal magnetite grains at this location are not likely to be derived from the Mount Polley deposit based on known ice-flow movements and the remoteness of the sample. However, the sample is located 1 km south-east (up-ice) of an alkalic porphyry Cu-Au occurrence. We suspect that other similar covered and unknown occurrences that host hydrothermal magnetite might be present in the area and, therefore, may be prospective targets for further exploration.

Summary and Further Work

Previous studies have shown potential for magnetite as an indicator mineral in exploration of porphyry deposits but there have been few if any published tests in the field. We carried out such a test on magnetite in till sampled around a known porphyry Cu-Au deposit at Mount Polley, where the glacial history has been well studied (Plouffe et al., 2013; Hashmi et al., 2015) and the composition of magnetite from mineralized and intrusive lithologies at the deposit are known (Canil et al., 2016). Laser ablation-ICP-MS was performed on detrital magnetite grains (n = 985) from 20 till samples collected over an area of ~900 km^2 surrounding the Mount Polley deposit. The LA-ICP-MS method which employs a large beam diameter and raster homogenizes and intergrates magnetite compositions in grains showing exsolution, and when calibrated using the 'standardless' AYCF method is an efficient protocol for analyzing large numbers of grains in an indicator mineral exploration study, ensuring that all trace elements (Mg, Ti, Al, V, Mn, Co and Ni) imperative for use in the discriminant models for magnetite can be consistently measured to low detection limits.

Previously proposed binary discriminant diagrams are not optimal for distinguishing hydrothermal magnetite in an exploration study. Caution must be taken when interpreting magnetite chemistry using certain elements affected by secondary processes such as sphenitization (for Si and Ca), which was relatively common in detrital magnetite from till. We instead employed linear discriminant analysis (LDA) to successfully classify magnetite from various porphyry deposits and barren igneous rocks using Mg, Ti, Al, V, Mn, Co and Ni. Anomalous values of hydrothermal magnetite were detected in samples up to 4 km from the deposit using the LDA models developed in this study, which is consistent with the ice-flow history and known glacial transport direction of the region.

The success of the LDA models in the Mount Polley area indicates that our approach has a proven potential in mineral exploration in glacial drift for porphyry systems in glaciated terrain. Further refinement of the LDA models will be possible with a larger compilation of new magnetite compositions from a greater number of known deposits in other regions. The compositional differences between ore-related and barren igneous magnetite needs further examination because ore-related grains in till can provide an additional exploration tool for porphyry systems where mineralization has not yet been exposed at the surface.

In this study, the number of analyses per sample was arbitrarily set at 50. Analyzing fewer grains per till sample will increase classification error, especially for distal samples down-ice of a deposit where the number of hydrothermal magnetite grains in till becomes scarce. The optimum number of grains per till sample to ensure that a target of viable size can be generated requires further investigation.

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Appendix 2 – Detailed Procedures for Discriminant Analyses of Magnetite

We compiled a database (n=493) of trace element concentrations in magnetite to use as a training dataset for linear discriminant analyses (Table 2). The first dataset (MDB1, n = 222) consists of magnetite compositions from Canil et al. (2016) and Mountjoy (2011) for six porphyry Cu(-Au-Mo) deposits and four barren igneous intrusions in the Canadian Cordillera. All data for MDB1 were obtained in the same LA-ICP-MS laboratory, thus providing an internally consistent dataset. The second dataset (MDB2, n = 271) combines the aforementioned data with those of Nadoll et al. (2015) from four porphyry Cu deposits, one Climax-Mo deposit (southwest United States) and one barren igneous intrusion (Japan), all measured at different LA-ICP-MS laboratories. The larger MDB2 dataset is less internally consistent, but is a more comprehensive representation of porphyry magnetite composition from a large number of deposits, rocks and locations.

Magnetite in both datasets was classified as either of: (1) hydrothermal porphyry magnetite (HTP) by occurring in veins and hydrothermal breccias, showing direct petrographic evidence of precipitation from fluids; (2) ore-related igneous magnetite (OIG) as an igneous mineral in fresh or altered intrusive rocks in a porphyry deposit; or, (3) barren igneous magnetite (BIG) as an igneous mineral from unmineralized intrusions. Summary statistics for the compositional groupings for magnetite in the MDB1 and MDB2 datasets are given in Tables A2-1 and A2-2, respectively.

Discriminant analysis requires values for all selected elements of interest, thus only those elements that are common between all the magnetite data sources were included in the analyses. Magnetite from skarn deposits was available for compilation of MDB1 and MDB2, but was excluded because it often contains trace element abundances that are below detection/reporting limits (Nadoll et al, 2012; 2014; 2015; Canil et al, 2016). For the internally consistent MDB1 dataset, only elements Mg, Al, Ti, V, Mn, Co, Ni and Sn were selected as variables, because Cu, Nb, Mo and Sn were only measured in 60-80% of the analyses. For the MDB2 dataset, a larger number of analyses have elements that are below detection, due to the higher limit of reporting (10σ of the background) criterion used by Nadoll et al. (2015), compared to the detection limit (3σ of the background) criterion we apply. To maximize the number of samples from the Nadoll et al. (2015) dataset, only elements Mg, Al, Ti, V, Mn, Co and Ni could be included in the discriminant analyses (Sn was excluded). Censored values were not used to substitute for any elements that are below detection.

Discriminant analysis assumes that each variable is normally distributed, but geochemical data are commonly skewed and/or contain outliers (Reimann & Filzmoser, 2000). Additionally, compositional data are 'closed' to a constant sum of 100%, which conflicts with the assumption of variable independence (Chayes, 1960; Vermeesch, 2006). The problem of closure is minimal for trace elements in magnetite because they comprise only a small portion of the bulk composition (which is mostly Fe). Nonetheless, to minimize deviations from normality and prevent the effects of data closure, each element (x_i) was transformed using the additive log-ratio method of Aitchison (1986). This method transforms D-part compositional data into (D – 1)-dimensional Euclidean space to which multivariate statistical methods can be applied. Iron was chosen as the component divisor due to its consistently high concentration and minimal variation (<10% relative) in magnetite. Transformed data for magnetite was imported into SYSTAT 13, a statistical software package, to perform the discriminant analysis.

The linear method of discriminant analysis (LDA) was chosen due to the simplicity of linear functions as compared to quadratic discriminant analysis (QDA), which is more complex and contains a larger number of parameters. In LDA, equal covariance between groups is required to produce reliable discriminant functions. Although QDA is the preferred method for datasets with unequal covariance, LDA is a very close approximation and can perform better on independent test data (Vermeesch, 2006). As a comparison, LDA and QDA were carried out on both datasets and the discriminant models were tested based on their predictive power for independent test data. There was no significant improvement using QDA.

To assess the capability of LDA for distinguishing magnetite composition, the MDB1 and MDB2 datasets were each subdivided into two parts: 1) a random 65% of the data used as 'training' for the discriminant analysis, and 2) the remaining 35% left as independent 'test' data. First, the optimum set of elements was determined because some may not significantly contribute to the discriminating power of the functions and can, therefore, be eliminated from the analysis. The stepwise method of discriminant analysis was used to incrementally add elements into the discriminant analysis based on their contribution to the model as indicated by the associated change in the Wilks λ parameter, which approaches zero with increasing power for the chosen element(s). Decreasing the number of variables in the discriminant analysis will naturally increase Wilks λ. Table A2-3 lists Wilks λ values associated with each element ranked in order of discriminating power by forward-stepping LDA using the training datasets. At 0.05 level of significance, all eight elements (Mg, Al, Ti, V, Mn, Co, Ni and Sn) were statistically significant in the MDB1 training data, whereas, only Al, Mg, Ni and Co were significant in the MDB2 training data.

The stepwise method is commonly used to optimize variable selection, but it is not always the best method for selecting the most useful subset of variables (Thompson, 1995). Alternatively, multiple iterations of discriminant analyses can be performed where each element is manually removed in turn from the analysis and each discriminant model is evaluated on the ability to correctly classify samples from an independent test dataset. Classification results on the 'test' data using various manually chosen element subsets for LDA compared rankings of the percent classification accuracy and the associated Wilks λ. For MDB1, a classification accuracy of 91% was achieved when Ti and Sn were excluded from the analysis (Table A2-4). This is contrary to stepwise LDA, which determined that all eight elements (Mg, Al, Ti, V, Mn, Co, Ni and Sn) were significant in the model. Similarly, stepwise LDA determined that Mg, Al, Co and Ni were significant for the MDB2 training data, but the derived functions using only these four elements performed relatively poorly on the test data, with 66% accuracy. In contrast, 78% of magnetite samples were classified accurately when using all seven elements (Mg, Al, Ti, V, Mn, Co and Ni) (Table A2-4). The stepwise method is not ideal for variable selection in this case. Therefore, only the elements with the greatest predictive power for the independent test data were selected for further analysis. Table 3 gives the equations for the discriminant functions (F1, F2) and Table 4 is a detailed summary of the classification for the magnetite compositional groups using the preferred set of elements for each dataset. The equations for the field boundaries separating the magnetite groups (HTP, OIG and BIG) are given in Table A2-5.

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