

**Platinum-group elements in Ordovician magmatic Ni-Cu sulphide  
prospects in northeast Scotland**

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## **Abstract**

Previous work on early Palaeozoic mafic-ultramafic intrusions in northeast Scotland identified two Ni-Cu-PGE exploration targets. The first prospect is at Arthrath, near the town of Ellon, and the second in the southeastern corner of the Knock intrusion, near the town of Huntly, around the farms of Littlemill and Auchencrieve. Both prospects occur within a group of mafic-ultramafic rocks known as the Younger Basic intrusions emplaced broadly synchronously with the later stages of Ordovician amphibolite facies metamorphism of the Dalradian Supergroup metasediments. In this study we have re-examined the available Ni-Cu exploration samples in terms of their PGE potential, placing particular emphasis on the less-well-known Arthrath prospect. PGE abundances of up to 418 ppb Pt (Littlemill) and 458 ppb Pd (Arthrath) associated with zones of Cu-Ni-Fe sulphide mineralisation have been identified. At Arthrath Pd is hosted primarily in merenskyite enclosed in base metal sulphide and the platinum-group mineral occurrences define a bimodal distribution. At Littlemill platinum-group mineral occurrences also involve mobilisation and redeposition of precious metals in sheared rocks enclosed within the base-metal sulphide zones. When the mineral compositions of all the rocks associated with the zones of Cu-Ni-PGE mineralisation are considered relative to the known differentiation series present within these intrusions, a model involving the mixing/mingling of relatively late, primitive magma and pre-existing differentiates is favoured for both prospects. In particular, a cumulate orthopyroxenite unit at Arthrath closely associated with a zone of Cu-Ni-PGE mineralisation is directly interpreted as the result of the influx into a magma chamber of a late, primitive magma. Previous work had suggested crustal contamination as the mechanism of sulphur immiscibility, and, although

evidence of crustal contamination is present, it is not favoured as the direct mechanism for the formation of the sulphide-rich zones that comprise the principal Ni-Cu-PGE exploration targets.

**Keywords:** Platinum-group elements, magmatic Ni-Cu sulphide, Littlemill, Auchencrievie, Arthrath, Scotland.

## **Introduction**

Of the possible exploration targets for platinum-group elements (PGE) in Britain, the potential for economic PGE deposits is greatest in the mafic-ultramafic intrusions of northeast Scotland (review of Gunn & Styles 2002). Anomalous concentrations of the PGE in these intrusions occur in a number of settings, but the highest PGE values are associated with magmatic Ni-Cu sulphide mineralisation in a suite of ultramafic-mafic rocks known as the Younger Basic intrusions. Fletcher (1989) suggested that reef-style stratiform deposits may occur in the Belhelvie Younger Basic intrusion, whereas Gunn & Styles (2002) noted similarities between mineralisation in the Knock and Arnage - Haddo House Younger Basic intrusions and contact-type deposits, a subclass of basal accumulation-type magmatic sulphide deposits (Thériault & Barnes 1998; Peck et al. 2001). This study has re-examined drillcore remaining from the Ni and Cu commercial exploration programmes from the two best prospects to assess the distribution and abundance of PGE and to conduct the first systematic search for platinum-group minerals. In addition we provide new silicate mineral chemical data that allows a new assessment of the petrogenesis of the rocks hosting the Ni-Cu-PGE mineralisation.

## **Regional Geology**

The mafic-ultramafic intrusions of northeast Scotland were first surveyed by Read (1919, 1923) who subdivided them into two main categories: an Older group (Older Basic) which was intruded prior to regional deformation and metamorphism; and a Younger group (Younger Basic), which, broadly speaking, postdated these events. Whether this distinction accurately describes the magmatism and associated deformation is still not

clearly understood (Gunn et al. 1996 and references therein). Rocks assigned to the Older group mostly occur in regional shear zones and are particularly abundant in the Upper Deveron area of the Portsoy shear zone (Fig. 1). They comprise ultramafic and mafic lithologies and are generally highly deformed and pervasively metamorphosed (e.g. Gunn et al. 1996). In contrast the Younger Basic rocks generally preserve magmatic textures, except where deformed in discrete shear zones (Ashcroft et al., 1984). They comprise several major intrusions including, amongst others, the Huntly, Knock, Inch, Arnage and Maud intrusions (Fig. 1). The Younger Basic rocks were emplaced into Neoproterozoic metasedimentary rocks of the Dalradian Supergroup at  $470\pm 9$  Ma, coincident with the later stages of amphibolite facies metamorphism in the country rocks (Dempster et al. 2002). The emplacement of the Younger Basic intrusions was part of the Grampian orogenic episode ( $\sim 480$ - $465$  Ma) that is postdated by isostatic uplift ( $465$  –  $435$  Ma) and the ‘main’ phase of Caledonian orogenesis in Scotland ending at  $\sim 395$  Ma (Oliver 2001). The Grampian orogenic episode may relate to the subduction of a back-arc basin (Highland Border back-arc), or the collision of the Midland Valley Arc with Laurentia due to the subduction of the northern Iapetus Ocean (Oliver 2001). Either way, in relation to the Younger Basic magmatism, both models imply that partial melting of the upper mantle was subduction-related and this is broadly consistent with the low ratio of high-field-strength elements to light-rare-earth elements found in these rocks (Thompson et al. 1984).

## **Previous exploration (Ni, Cu and PGE)**

Despite the limited exposure and structural complexity of the mafic-ultramafic rocks in the region, substantial exploration progress has been made over the last 30 years. The Younger Basic intrusions were first explored systematically for metallic minerals in the late 1960s when Rio Tinto Zinc (RTZ, now Rio Tinto plc) and Consolidated Goldfields (CGF) initiated independent programmes of exploration for nickel. Subsequently, in 1969, RTZ and CGF formed a joint-venture company, Exploration Ventures Ltd., (EVL), to explore throughout the region. EVL conducted extensive programmes of geochemical and geophysical surveys, together with geological mapping.

Most geochemical samples were analysed for Cu and Ni only, although in some areas Mo, Pb and Zn were also determined and analyses for the PGE were restricted to a small suite of rock samples. CGF, working mainly on the Huntly and Knock intrusions in the west of the region, completed 59 diamond drillholes for a total of 9224 m. Their principal discovery (the Littlemill prospect) was at the south-eastern corner of the Knock intrusion, near to the town of Huntly, at the farms of Littlemill and Auchencrieve (Fig. 1 and Fig. 2), where a resource of 3 Mt grading 0.52 % Ni and 0.27 % Cu was identified in two sub-parallel, near-conformable zones dipping towards the north-west (Wilks 1974). The second prospect, located in the east of the region, at Arthrath, near the town of Ellon, occurs within unexposed intrusive rocks that lie immediately to the east of the Arnage-Haddo House intrusion and has been named the Arthrath prospect (Fig. 1 and Fig. 3). Here thirty-six diamond drillholes led to the identification of sulphide mineralisation in five zones over a strike length of about 4 km. At Arthrath EVL outlined a resource of 17

Mt grading 0.21% Ni and 0.14% Cu (Wilks & Smith 1976). Subsequent investigations demonstrated the presence of enhanced PGE values in both areas and these further enhance their attractiveness as exploration targets under current market conditions (Fletcher 1989; Fletcher & Rice 1989; Fletcher et al. 1997).

Exploration for PGE deposits has thus far mainly focused on the Ni-Cu sulphide mineralisation identified within the Knock (predominantly the Littlemill prospect) and adjacent Huntly intrusion (Fletcher 1989; Fletcher & Rice 1989). Drillcore and outcrop samples covering the known range of sulphide contents and rock types (including one borehole section, RD15) yielded Ni and Cu values in the range <0.01 to 3.02 wt % and <0.01 to 6.46 wt %, respectively. Mean Cu/(Cu+Ni) values are in range 0.25 - 0.60, with ultramafic rocks having lower values than mafic rocks. Precious metals showed rare, sporadic low-tenor enrichment. In the Knock intrusion, Pt values range from 2 to 18 ppb, Pd from 5 to 381 ppb and Rh from 2 to 6 ppb, whereas, in the adjacent Huntly intrusion, Pt ranges from 2 to 584 ppb, Pd from 2 to 93 ppb and Rh from 2 to 6 ppb. The highest Pd and Au values were found in gabbro-norite with net-textured sulphide from 120.8 m depth in borehole RD15 located in the Littlemill Ni-Cu prospect; whereas the highest Pt value was found in the Huntly intrusion in an outcrop sample termed a 'graphitic pyroxenitic pegmatite' from the Bin Quarry (Fletcher & Rice 1989). Few details are known of the mineralogical hosts to the PGE, although, rare, unidentified PGM and a rhenium-bearing sulphide have been reported from the Knock intrusion (Jedwab & Fletcher 1991).

In addition to the anomalies present in the Knock and Huntly intrusions, reconnaissance studies of drillcore from other Younger Basic intrusions led to the identification of two other locations with elevated PGE concentrations (Fletcher 1989). In the Belhelvie Younger Basic intrusion on the east coast near Aberdeen (Figure 1), Pt values of 2-88 ppb, Pd of 2-113 ppb and Rh of 2-7 ppb were reported in intrusive rocks named 'picrite/dunite' of which little else is known (Fletcher 1989). The second discovery was at Arthrath where magmatic Ni-Cu sulphide mineralisation was found in noritic rocks in borehole AD25 (generally 2-5 volume % sulphide, but locally up to 80%) and in this borehole Pt values of 2 - 201 ppb, Pd of 2 - 138 ppb and Rh of 2 - 7 ppb were reported in samples containing up to 1% Ni and 0.6% Cu (Fletcher 1989; Fletcher et al. 1997).

### **Sampling and analytical techniques**

Samples representative of the rock types and styles of mineralisation present, most between 10 and 60 cm in length, were collected for geochemical analysis from Exploration Ventures Limited (EVL) drillcore held by the British Geological Survey. In particular the zones of Ni-Cu mineralisation were targeted for the two principal prospects. At Littlemill, three boreholes that preserve zones of massive sulphide were sampled (n=38; boreholes RD7, RD20 and RD22). Subsequently, mineralogical investigations targeted the relatively PGE-rich massive sulphide zones, with polished sections made from samples taken at 43 points. At Arthrath, 33 geochemical samples were taken from the only available borehole that intersects zones of sulphide mineralisation (AD25). Forty-four samples for mineralogical investigation were taken



throughout borehole AD25 and these were supplemented by a few additional samples from a second borehole, AD24.

All whole-rock samples were analysed at the British Geological Survey by XRF analysis of pressed powder pellets for the following elements: Ag, Ba, Ce, Co, Cr, Cu, Fe<sub>2</sub>O<sub>3</sub>, La, Mo, Nb, Ni, Pb, Rb, Sr, V, Y, Zn and Zr. Analysis of the rocks for Pt, Pd and Au was carried out by Acme Analytical Laboratories Ltd, Vancouver, by lead fire assay of 30 g aliquots with an ICP-MS finish. The same laboratory also determined concentrations of As, Sb, Bi, Te and Se by aqua regia digestion of 1 g samples and ICP-MS finish. Total sulphur was determined by Leco induction furnace.

All polished sections were examined optically in transmitted and reflected light. Electron-microprobe analysis of silicate phases (n=~5 per phase per sample) was performed at the British Geological Survey using a Cameca SX50 wavelength-dispersive microprobe operating at 15 kV and 20 nA. Forty-six out of 87 polished sections were subjected to automated electron microprobe searching for platinum-group minerals (PGM) on a 1- or 2- $\mu$ m grid, using the back-scattered electron signal to locate minerals with high atomic number. PGM thus located were characterised using energy-dispersive spectra (EDS) and, if possible, wavelength-dispersive analysis (WDS) operating at 15 kV and 20 nA. In total, 37 PGM occurrences were characterised by WDS analysis and a further set of occurrences (~20) were imaged and characterised by qualitative EDS analysis.

Sample details and the results of these analyses are available from the Depository of Unpublished Data, CISTI, National Research Council, Ottawa, Ontario K1A 0S2, Canada.

## **Mineralogy and geochemistry**

### **Arthrath**

The principal rock types identified in the samples from Arthrath are norite, micronorite (e.g., Fig. 4A) and olivine gabbro-norite (e.g., Fig. 4B). Also present are orthopyroxenite (e.g., Fig. 4C), microgabbro-norite, spinel-bearing norite, microgabbro and tonalite. All of these rocks are biotite-bearing. The rocks are medium- to coarse-grained, subhedral granular lithologies (granular, in this context, refers to a texture where all the main rock-forming minerals are approximately of equal size and randomly orientated). Interstitial phases, where present, are plagioclase  $\pm$  clinopyroxene  $\pm$  biotite and an interstitial texture is most obvious in the orthopyroxenite and olivine gabbro-norite lithologies (Figs. 4B and C). Some textural heterogeneity occurs on the scale of an individual thin section with fine- to medium-grained granular domains (e.g., Fig. 4D) coexisting with the predominant medium- to coarse-grained domains. Amphibole is common as an overgrowth on pyroxene, and olivine is serpentinised to varying degrees. In some samples, particularly from the lower part of borehole AD25, there is a pervasive alteration to muscovite, carbonate and clay minerals. Similar rock types occur in boreholes AD24 and AD25, although correlations between them cannot be made on account of lateral variations and structural complexity. Sulphides present are pyrrhotite, pentlandite and chalcopyrite, with pyrrhotite predominant, and in the majority of samples

do not exceed 5% by volume. Sulphide is commonly interstitial to net-textured (e.g., Figs. 5A, B) but disseminated and submassive to massive textures also occur (e.g., Fig. 5C).

Exploration Ventures Limited sub-divided AD25 into six lithological units and identified two sulphide-rich zones; the sulphide zones did not correlate with the location of unit boundaries (Fig. 6). The upper sulphide zone is associated with the local development of an orthopyroxenite lithology (sample ATD11) characterized by relatively coarse orthopyroxene with interstitial plagioclase, biotite and sulphide (Fig. 4C). In addition, the orthopyroxenite is characterized by a relatively abrupt increase in En content of orthopyroxene and a less well-defined decrease in An content of plagioclase (Fig. 6; Table 1). By contrast, the lower sulphide zone is associated with microgabbroic rocks without obvious changes in mineral compositions (Fig. 6). In the lowermost part of the borehole, corresponding to EVL's units 4, 5 and 6, olivine-bearing rocks are present, but with only trace amounts of sulphide. These rocks are texturally similar to each other and, as there is little change in mineral compositions with depth, it is therefore suggested that these olivine-bearing rocks may represent a single olivine-gabbroic unit (3) (Fig. 6). The olivine from these olivine gabbroic rocks is characterised by low Ni contents (average  $0.09 \pm 0.05$  wt% Ni (n=20) in olivine of average composition  $\text{Fo}_{80 \pm 0.7}$ ). These olivine-bearing rocks contain rare sulphide but in the smallest amounts found in any rocks in borehole AD25 (interstitial and enclosed in olivine and orthopyroxene; <1%).

At Arthrath, only two samples analysed in this study contain more than 1% Ni and only one sample has a Cu value greater than 0.5% (Table 2). The highest Pd value, 458 ppb, occurs in massive sulphide with 1.69% Ni, 0.41% Cu and very low Pt and Au contents. The second highest Pd value (207 ppb) occurs in a rock with disseminated sulphide with local narrow bands of massive sulphide. It also contains 42 ppb Pt, 57 ppb Au, 0.85% Ni and 0.59% Cu. Only three samples have Pt contents exceeding typical values for mafic and ultramafic rocks (Crocket 2002), ranging from 35–47 ppb Pt. The downhole distribution of Pd illustrates that the highest abundances are associated with the two sulphide-rich zones described by EVL (Fig. 6). There are positive correlations between Pd and Fe<sub>2</sub>O<sub>3</sub>, Ni, Cu, Co, S, Bi, Te, Se, Ag, and Au, suggesting that these elements occur in the same or coeval phases (e.g., Figs. 7A to D); Pt has a positive correlation with S, Cu, Fe<sub>2</sub>O<sub>3</sub>, Ag and Bi (e.g., Figs. 7E and F). Pd/Pt values vary widely; the median value is 2, but in five samples Pd/Pt values exceed 10 (e.g., Table 2; Fig. 7E).

The majority of PGM occurrences found at Arthrath are in the sulphide-rich zones in AD25, between depths of 102.75- 109.75 m, and 176.2 - 179.3 m (Table 3). However, four PGM occurrences were also found in four polished sections from borehole AD24, at depths ranging from 133.6 m to 237 m. All PGM occurrences at Arthrath for which WDS microprobe data were obtained are of merenskyite (PdTe<sub>2</sub>) and one occurrence of melonite (NiTe<sub>2</sub>) was also found. The merenskyite present contains appreciable Ni substituting for Pd (up to (Pd<sub>0.52</sub>Ni<sub>0.44</sub>)(Te<sub>1.80</sub>Bi<sub>0.16</sub>)) and the melonite contains substantial Pd ((Ni<sub>0.65</sub>Pd<sub>0.30</sub>)(Te<sub>1.67</sub>Bi<sub>0.27</sub>)). All merenskyite analyses show Bi substituting for Te (up to (Pd<sub>0.94</sub>)(Te<sub>1.09</sub>Bi<sub>0.82</sub>)), defining two compositional populations; in some cases high-Bi

merenskyite plots near to the Bi-rich end-member michenerite (Fig. 8). Other precious metal-bearing minerals found at Arthrath were native gold, hessite ( $\text{AgTe}_2$ ), and a BiTe mineral. Merenskyite at Arthrath most commonly occurs included in pyrrhotite or pentlandite (Figs. 9A and B) but it has also been found along sulphide-silicate grain boundaries and more rarely in fractures within sulphide or silicate (Fig. 9C).

### **Littlemill**

A few samples of host igneous lithologies were studied from boreholes RD7, RD20 and RD22 at Littlemill. The rock types are (micro)gabbro-norite, tonalite, and norite, and, as at Arthrath, all are biotite-bearing and replacement of pyroxene by amphibole is common. Noritic rocks are texturally homogeneous and predominantly medium-grained, subhedral granular (e.g., Figs. 4E and F). Sulphides are predominantly submassive to massive, although some vein and disseminated sulphides also occur (Figs. 5D to H). As at Arthrath, the sulphide assemblage comprises predominantly pyrrhotite, with lesser chalcopyrite, pentlandite and, more rarely, pyrite. In addition to abundant crystal and rock fragments of the host rocks, submassive to massive sulphide also contains xenoliths of mylonitic metanorite, foliated amphibolite and pyroxene-bearing tonalite.

Mineral compositions of the host lithologies from the sulphide zones at Littlemill indicate substantial differences to those of AD25 from Arthrath (Table 1). In boreholes RD7 and RD22 orthopyroxene ranges from  $\text{En}_{39}$  to  $\text{En}_{48}$ , and is therefore more Fe-rich than in sulphide rich zones at Arthrath ( $\text{En}_{70-78}$ ), as are clinopyroxene compositions (Mg# of 0.59 to 0.62 versus 0.78 to 0.82) (Table 1). Plagioclase compositions range from  $\text{An}_{54}$  to  $\text{An}_{67}$

at Littlemill and are more similar to the sulphide-rich zones at Arthrath ( $An_{62-67}$ ). There is a large contrast, and no overlap, in the composition of biotite between the rocks studied here from the two prospects: the Mg# of biotite ranges from 0.69 to 0.87 at Arthrath and from 0.42 to 0.50 at Littlemill (Table 1). Thus all minerals from the host rocks to the sulphide zones show more evolved compositions at Littlemill by comparison with AD25 at Arthrath.

Fifteen samples contain more than 20 ppb Pd. The highest values are found in massive sulphide ores of various types from the sulphide-rich zones defined by EVL (Fig. 10). The maximum value (105 ppb Pd) occurs in a massive sulphide from borehole RD22 containing 2.4% Ni, 1774 ppm Cu and 61 ppb Au. The second highest value (101 ppb Pd) is in a massive sulphide ore with high Ni (2%), Cu (1.2%) and Au (2669 ppb). The third highest is in a banded massive sulphide ore with high Ni (2.2%) but low Cu and Au contents. The Pt values in these three samples are low (<5 ppb). A highly anomalous Pt value occurs in a single sample from borehole RD22 at Littlemill. In this sample 418 ppb Pt is accompanied by 2.2% Ni, but the Cu and Pd contents are low. Only two other samples contain above background Pt values. All other samples, both ores and host rocks, have low precious metal contents. Two samples (LMD 108 and 114) of vein-style mineralization dominated by chalcopyrite do not contain high precious metal values. Pd/Pt values vary widely in the mineralised samples from Littlemill; the median value is 3, but 12 samples have Pd/Pt values greater than 10. By comparison with Arthrath, inter-element correlations are much weaker in the Littlemill boreholes. Pd is correlated with  $Fe_2O_3$ , Co, Ni, Bi, Te, Se and S, but the data are much more scattered (e.g., Figs. 7A to

D). Pd is not correlated with Pt, Au or Cu. Pt has no significant correlation with any other elements determined and is largely independent of S content (e.g., Figs. 7E and F).

Automated searching of 26 samples from Littlemill led to the location of PGM in two polished sections: in RD7, at depth of 51.8 m, and RD20, at a depth of 138.5 m (Table 3). A single example of a Bi-rich merenskyite-michenerite (PdTeBi) was found in RD20 along a sulphide-silicate grain boundary (Fig. 8, 9D). As at Arthrath, some substitution of Pd by Ni is present ( $\text{Pd}_{0.91}\text{Ni}_{0.05}$ ), as is Bi by Sb ( $\text{Bi}_{0.73}\text{Sb}_{0.22}$ ). In borehole RD7, at 51.8 m, the occurrences are of a different type. Here, the PGM have been found in a mylonitic metanorite rock enclosed within the massive sulphide. Froodite ( $\text{PdBi}_2$ ) and a PdSb mineral have been found in addition to numerous grains of native Au and Bi within the fractures of this xenolith. In detail, these grains of native Au and native Bi are anhedral and regions of Pd-Bi alloy have been identified within them by WDS mapping techniques (Fig. 11). Unidentified Ni and As minerals were also found in this sample. Another occurrence of a mineralized xenolith is found in RD22 at 153.9m; here numerous grains of native gold and bismuth were identified, but no PGM.

## **Discussion**

Re-examination of the exploration samples of nickel-copper sulphide ores from the prospects at Arthrath and Littlemill has shown that they contain enrichments in Pd and, to a lesser extent, in Pt. The maximum reported values in this study are 456 ppb Pd and 418 ppb Pt. These values are in agreement with the previous studies of PGE, confirming the

overall relatively low tenor of these rocks (Fletcher & Rice 1989; Fletcher et al. 1997). Enrichment of Au is also locally present (up to 2669 ppb). In general, all samples containing high PGE values are massive or sub-massive ores but many such samples do not have anomalous PGE contents. The mineralogical data presented in this study suggests however that substantial differences exist in the nature of the host rocks to these PGE enrichments and these data now permit a new assessment of the petrogenesis of the mineralised rocks, in particular for the prospect at Arthrath.

#### *Crustal contamination*

The process whereby mantle-derived magmas assimilate sulphur-bearing crustal rocks has been cited by many authors as a key factor in the genesis of Ni-Cu  $\pm$  PGE sulphide deposits (e.g., Naldrett 1999; Ripley et al. 2002; Arndt et al. 2003). The Younger Basic rocks have for many years been regarded as a contaminated series on the grounds that metasedimentary xenoliths are abundant and some metasediments show evidence of partial melting of the country rock which was experiencing amphibolite facies regional metamorphism at the time of emplacement (e.g., Read 1966; Gribble 1970). As a result, crustal contamination has previously been investigated as a possible cause of sulphur saturation for both of these prospects, but the geochemical evidence for crustal contamination is ambiguous (Fletcher & Rice 1989; Fletcher et al. 1997). For example, isotopic studies have found average  $\delta^{34}\text{S}$  for igneous rocks and sulphides in both prospects to be broadly 'magmatic' falling in the range  $0.5 \pm 2.4\text{‰}$  at the Huntly and Knock intrusions (igneous rocks) and  $-0.9 \pm 0.5\text{‰}$  (igneous rocks) and  $-1.2 \pm 1.0\text{‰}$  (sulphide) at Arthrath (Fletcher et al. 1989; Fletcher et al. 1997). These studies invoked a



model whereby, on intrusion, the magma was undersaturated but near to sulphur saturation, and a modest amount of crustal contamination then resulted in sulphur immiscibility causing the formation of the massive sulphides.

In this study the role of crustal contamination has been investigated using the S/Se ratio of the rocks. Variations in S/Se, in particular increasing S/Se with increased magmatic differentiation, can discriminate between crustal (>3000) and magmatic (<3000) sources of sulphur in Cu-Ni-PGE-bearing mineral deposits (e.g. Thériault & Barnes 1998). S/Se values for both Arthrath and Littlemill exceed 3000 in almost all samples (Table 3). At Arthrath, S/Se values average 5985, whereas at Littlemill the average value is 4980, and for both prospects S/Se values are relatively constant with depth (Figs. 6 and 10). It appears therefore that the S/Se ratios of the mineralised rocks are consistent with their derivation from crustally contaminated magmas. However, in borehole AD25 at Arthrath there is no correlation between the location of sulphide-rich zones and increasing S/Se suggesting that, although the magmas appear contaminated, they may have been contaminated at an earlier stage in their petrogenesis and thus there is no link to suggest that the zones of Cu-Ni-PGE mineralisation formed as the direct result of crustal contamination.

Thus far all grains of olivine analysed from Arthrath are characterised by low Ni contents (Fletcher et al. 1997; this study). Furthermore, the rocks with low Ni-in-olivine examined in this study contain sulphide (<1%), sometimes found as inclusions within the olivine, and these rocks are also biotite-bearing. It is suggested therefore that, for the ol-

bearing rocks in the lower part of borehole AD25 at Arthrath, sulphide saturation preceded crystallisation of low-Ni olivine. This, along with the relatively low Ni and PGE tenors of both prospects, points to a possible role for an earlier sulphur saturation event in these rocks (Fletcher et al. 1997) and would be consistent with crustal contamination of the rocks at a relatively early stage in their petrogenesis.

#### *Orthopyroxenite formation*

This study has found that one of the Ni-Cu-PGE-rich zones in borehole AD25 is associated with a distinctive orthopyroxenite lithology co-incident with cryptic variations in the composition of orthopyroxene (Fig. 6). The texture of the orthopyroxenite suggests that cumulate processes were involved in the formation of this lithology (Fig. 4). A sulphide-bearing orthopyroxenite unit displaying some cryptic mineralogical variations within a sequence of noritic rocks has been interpreted in the Proterozoic Bjerkreim-Sokndal layered intrusion, Norway, to result from the mixing of relatively primitive and more differentiated magmas (Jensen et al. 2003). It is suggested therefore that the Arthrath orthopyroxenite unit associated with the upper sulphide zone in borehole AD25, and the occurrence of texturally heterogeneous noritic rocks with it, indicates that mixing/mingling of primitive and more differentiated rocks occurred in the Arthrath intrusion providing an alternative mechanism (review of Maier et al. 1998) for the development of sulphur immiscibility. The bimodal nature of PGM occurrences found in borehole AD25 at Arthrath (Figure 8) would also be consistent with this interpretation.

### *Differentiation trends of the mineralised zones*

If, as argued above, mixing/mingling of primitive and more differentiated rocks was the cause of sulphur immiscibility then this process should be evident in the differentiation trends of the rocks from the zones of Ni-Cu-PGE mineralisation. In order to test this theory further the rocks described in this study, and those by Fletcher (1989) from the Littlemill-Auchencrieve mineralised zone (contact zone), were compared to examples of the Younger Basic Lower, Middle and Upper differentiation series (Wadsworth 1986, 1988 and 1991). The Lower, Middle and Upper differentiation series are essentially unmineralised rocks in that they do not contain the sulphide-rich zones that occur at Arthrath and Littlemill-Auchencrieve. In this study the Middle and Upper series are represented by data from the 'type' intrusion at Inch. However the Lower Series has been taken from the Belhelvie intrusion because these rocks are similar to the Lower Series at Inch but are better exposed and have been examined in more detail (Wadsworth 1991).

The rocks from Arthrath and Littlemill-Auchencrieve associated with zones of Ni-Cu-PGE mineralisation (this study; Fletcher 1989) have been subdivided into three broad groups on Figure 12A. Group 1 rocks are the most primitive ( $Mg\# \text{ opx} > 70$ ) and they comprise rocks from both prospects. They define a scattered distribution that does not correspond to any of the differentiation series and only some of the most evolved Group 1 rocks show some similarity with the compositions found in the Lower and Middle Series (Figure 12A). In detail, these Group 1 rocks are sometimes olivine bearing, characterised by a bimodal distribution ( $Mg\# \text{ opx}$  is either  $>81$  or  $<78$ ), and, at least in

the case of the rocks from Arthrath examined in this study, can have variable plagioclase contents (Figure 12B). In summary, Group 1 mineralised rocks are a heterogeneous group of relatively primitive rocks without a clear differentiation trend and include the orthopyroxenite interpreted in the previous section as the product of mixing/mingling between primitive and more evolved magmas. A second group of distinctive, relatively primitive mineralised rocks is defined using four rocks described by Fletcher (1989) from the Littlemill-Auchencrieve prospect and these too do not correspond to any of the differentiation series on Figure 12 (Group 2 rocks).

Finally, a third group of mineralised rocks is defined (all of which are from Littlemill-Auchencrieve) and they are grouped together because they all show an affinity with one or other of the differentiation series. In particular these Group 3 mineralised rocks are similar to the Middle Zone differentiation series defined by Wadsworth (1988). In detail, some of the Group 3 mineralised rocks show an affinity with Middle Zone granular rocks, whereas others are most similar to Middle Zone rocks with more coarse-grained texture interpreted to be of cumulate origin (Figure 12). Thus it can be suggested that the relatively evolved mineralised rocks from Littlemill-Auchencrieve do show considerable affinity with the differentiation series, in marked contrast to the relatively primitive mineralised rocks that predominantly do not (Figure 12).

The differences illustrated on Figure 12 between the rocks associated with zones of Cu-Ni-PGE mineralisation of the Arthrath and Littlemill-Auchencrieve prospects and the Younger Basic differentiation series can be understood if processes of magma mixing/mingling involving relatively late, primitive magmas containing enhanced Ni-Cu-

PGE concentrations are emplaced into pre-existing intrusions undergoing differentiation. This would explain why only the most evolved mineralised rocks show strong affinity with the differentiation series (i.e. they are products of differentiation that have been modified at a late stage by the arrival of new primitive magmas resulting in the formation of sulphide-rich zones). Whereas the relatively primitive mineralised rocks define new compositions and 'trends' because they have an entirely different differentiation history from the differentiation series. Thus the composition of the host rocks to the zones of Ni-Cu-PGE mineralisation can vary depending on which part of the differentiation series (Lower, Middle, Upper) is invaded by the newer magma. However, it should be noted that the mineralised rocks, even the most primitive ones, have characteristics (biotite-bearing; high, relatively invariant Se/S ratios; and olivine with relatively low Ni-contents) that would suggest that some, if not all, of these relatively late magmas have experienced an earlier differentiation history involving crustal contamination and sulphur saturation (Fletcher et al, 1997).

#### *Magmatic and Hydrothermal processes*

Previous studies of the Littlemill prospect have suggested that the geochemical (PGE-Ni-Cu-Fe) patterns indicate remobilization of base and precious metals during amphibolite facies shearing (Fletcher & Rice 1989). Our observation of Pd-bearing minerals at Littlemill associated with native Au and native Bi in fractures in a mylonitic metanorite hosted within the massive sulphide further supports this interpretation. By contrast, at Arthraath, Pd values correlate with Fe<sub>2</sub>O<sub>3</sub>, Ni, Cu, Co, S, Bi, Te, Se, Ag, and Au indicating an orthomagmatic control by primary sulphide liquid remains the dominant mechanism.

In addition, the mode of occurrence of the PGM at Arthrath, where the Pd-bearing mineral merenskyite is most commonly found enclosed in sulphide or along sulphide-silicate grain boundaries, supports this hypothesis. Thus we conclude that the varied distribution of Pd at Littlemill reflects, at least in part, the dissolution and remobilization of primary PGE and host sulphide by hydrothermal fluids, and deposition within the fractured, sheared xenoliths. This is consistent with the variability and commonly higher values of Pd/Pt observed at Littlemill. In contrast, at Arthrath, the distribution of PGE retain a greater degree of orthomagmatic control by primary sulphide liquid.

## **Conclusions**

1. Crustal contamination is not favoured as the mechanism for sulphur immiscibility recorded in the zones of Cu-Ni-PGE mineralisation in these rocks because of the lack of correlation between high S/Se ratios and sulphide-rich zones in either of the prospects.
2. An orthopyroxenite lithology at the Arthrath prospect, associated with cryptic variations in orthopyroxene composition and a zone of Cu-Ni-PGE mineralisation, is interpreted as the product of the mixing/mingling between relatively late, primitive magmas and earlier magmas that had already differentiated to more evolved compositions.
3. The mineral compositions of all the rocks associated with the sulphide-rich zones from both prospects are consistent with a model involving mixing/mingling between new influxes of primitive magmas into the chamber as the mechanism for sulphur immiscibility and the formation of the sulphide-rich zones.

4. The Ni-Cu-PGE abundances at Arthrath are predominantly of magmatic origin, whereas those at Littlemill-Auchencrieve have a similar origin but have also been substantially modified by later hydrothermal activity.

### **Acknowledgements**

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## **Figure captions:**

Figure 1: The geology of northeast Scotland showing the locations of the prospects examined in this study, at Littlemill-Auchencrieve and at Arthrath. The illustrated subdivision of the Older and Younger Basic intrusions is not clearly established, although it is known that intrusions with Older Basic characteristics are predominantly to be found in the Upper Deveron area.

Figure 2: The geology of the southeast corner of the Knock intrusion around the farms of Littlemill and Auchencrieve (adapted from British Geological Survey 2000). The locations of boreholes used in this study are shown. As bedrock exposure is sparse, the geology was largely compiled from borehole data (drilled by Aberdeen University, Exploration Ventures Limited and British Geological Survey) and ground geophysical surveys, chiefly magnetic.

Figure 3: The geology of the Arthrath prospect (adapted from Fletcher et al. 1997). The locations of boreholes used in this study are shown. The geology was compiled from sparse exposure, float mapping, geophysical surveys and drilling.

Figure 4: Cross-polarised light photomicrographs illustrating selected igneous lithologies and textures identified in samples from Arthrath borehole AD25, and Littlemill boreholes RD7, 20 and 22. (A): AD25, sample ATD7, micronorite, 85.9 m; (B): AD25, sample ATD28, serpentinised olivine gabbro-norite, 305 m; (C): AD25, sample ATD11, orthopyroxenite, 109.8 m; (D): AD25, sample ATD19, microgabbro domain in a

texturally heterogeneous microgabbonorite, 161.5 m; (E): RD7, sample LMD2, microgabbonorite, 52.2 m; and (F): RD22, sample LMD16, microgabbonorite, 153.7 m.

Figure 5: Scans of selected polished thin sections from Arthrath borehole AD25 (A to C) and Littlemill boreholes RD7, RD20 and RD22 (D to H) to illustrate the variety of sulphide textures found in these rocks. A) AD25, sample ATD12, 120.7 m, interstitial to net-textured sulphide in norite; B) AD25, sample ATD17, 148.3 m, interstitial sulphide in norite; C) AD25, sample ATD35, 109.3 m, disseminated to sub-massive sulphide; D) RD22, sample LMD14, 150.7 m, cross-cutting vein sulphide in microgabbonorite; E) RD22, sample LMD32, 158.0 m, banded to disseminated sulphide in orthopyroxene-bearing microgranitic rock; F) RD22, sample LMD13, 151.8 m, disseminated sulphide in micronorite; G) RD7, sample LMD2, 52.2 m, banded massive sulphide in microgabbonorite; and H) RD22, sample LMD15b, 153.9 m, sub-massive sulphide.

Figure 6: Downhole variations in lithology, sulphide content, mineral chemistry and selected trace elements for borehole AD25 at Arthrath. The lithological sub-division of the borehole (column 1) is from Exploration Ventures Limited and all other data are from this study. All errors are  $\pm 1$  sigma.

Figure 7: Scatter plots comparing the geochemistry of samples from borehole AD25 at Arthrath with samples from boreholes RD7, RD20 and RD22 at Littlemill. For clarity two Pd values have been reduced from 207 ppb and 456 ppb to 150 ppb, and one Pt value has been cut from 418 ppb to 60 ppb.

Figure 8: Triangular plot of the atomic concentrations of Pd(+Ni, Pt), Te and Bi(+Sb) illustrating the composition of platinum-group minerals found at Arthrath and Littlemill. The fields for merenskyite and michenerite are defined by the range of compositions for these minerals given in Tables 97 and 102 of Cabri (2002).

Figure 9: BSEM micrographs illustrating the occurrence of platinum-group minerals at Arthrath (A to C) and Littlemill (D). A) AD25, sample ATD33, 102.8 m, merenskyite (white) inclusion in pyrrhotite (darker grey); B) AD25, sample ATD34, 103.1 m, two merenskyite (white) inclusions in intergrown pentlandite (lighter grey) and pyrrhotite (darker grey); C) AD24, sample ATD44, 237.0 m, merenskyite (white) in quartz-filled fracture (black) crossing pyrrhotite (grey); and D) RD20, sample LMD24, 138.5 m, michenerite (white) located at the grain boundary of quartz (dark grey) and pyrrhotite (light grey).

Figure 10: Downhole variations in lithology, sulphide content and selected trace elements for boreholes RD7 and RD22 at Littlemill. The lithological sub-division of the borehole (column 1) and the sulphide content (column 2) are from Exploration Ventures Limited and all other data are from this study. For clarity one Pt value has been reduced from 418 ppb to 120 ppb in RD22.

Figure 11: Selected plane-polarised light photomicrographs, BSEM micrographs and WDS microchemical maps illustrating the occurrence of Pd-, Bi- and Au-bearing minerals in sample LMD25 (51.8 m) from borehole RD7 at Littlemill. A) Plane-polarised light photomicrograph showing a mylonitic metanorite rock within the sulphide; B) BSEM micrograph of a string of native bismuth and gold grains, and fracture-filling sulphide, hosted within the metanorite illustrated in A; C) BSEM micrograph of another occurrence of native bismuth and gold, also found within the metanorite illustrated in A (not the same location as B); and D and E) WDS microchemical maps of a Pd-bearing zone (E, bottom right) within intergrown native bismuth (E, bottom left) and gold (D, bottom right) found in the metanorite illustrated in A (not the same location as B or C).

Figure 12: The Mg# of orthopyroxene versus the anorthite content of plagioclase for rocks from the Younger Basic differentiation series compared to the rocks associated with the Cu-Ni-PGE mineralisation. The data labeled 'Arthrath' and 'Littlemill' are from this study, those labeled under the heading 'Littlemill-Auchencrieve' are from Fletcher (1989) and those labeled under the heading 'Lower, Middle, Upper series' are from Wadsworth (1991), Wadsworth (1988) and Wadsworth (1986) respectively. The subdivision of the mineralised rocks into three groups (1, 2 and 3) is explained in the text (see the discussion section).



TABLE 1: SUMMARY OF THE AVERAGE MINERAL COMPOSITIONS FOR ARTHRATH BOREHOLE AD25 AND LITTMILL BOREHOLES RD7 AND RD22.

BH	Sample	Depth (m)	Ol	Opx				Cpx				Pl	Bt	n=
				Fo	En	Fs	Wo	Mg <sup>#</sup>	Wo	En	Fs			
AD25	ATD2	41.90	-	78	18	4	0.82	-	-	-	-	65	0.75	20
AD25	ATD5	62.60	-	72	25	4	0.75	na	na	na	na	72	0.80	21
AD25	ATD8	92.45	-	78	19	3	0.81	-	-	-	-	67	0.73	21
AD25	ATD9	103.00	-	73	23	4	0.77	48	15	37	0.78	67	0.69	22
AD25	ATD10	109.40	-	70	27	3	0.73	42*	12	46	0.82	63	na	28
AD25	ATD11	109.75	-	78	18	4	0.82	46	11	43	0.81	62	0.76	21
AD25	ATD13	123.80	-	71	25	3	0.74	44	10	45	0.81	73	0.72	18
AD25	ATD18	152.40	-	74	23	3	0.77	-	-	-	-	69	0.87	17
AD25	ATD19	161.45	-	73	24	4	0.76	45	13	43	0.79	69	0.70	13
AD25	ATD20	176.20	-	74*	22	4	0.77	42*	12	46	0.78	63	-	7
AD25	ATD24	216.15	80	79	17	4	0.83	48	12	40	0.85	66	0.83	24
AD25	ATD26	254.70	81	81	15	3	0.84	51	10	39	0.86	63	0.77	19
AD25	ATD31	341.66	80	81	17	2	0.85	46	9	45	0.94	64	0.87	19
RD7	LMD1	48.00	-	41	58	1	0.42	-	-	-	-	54	0.43	13
RD7	LMD25	51.80	-	44	54	2	0.46	34*	24	42	0.59	57	0.50	15
RD7	LMD-2	52.20	-	48	50	2	0.49	-	-	-	-	67	0.50	19
RD22	LMD12	151.45	-	46	52	2	0.47	34*	21	45	0.62	63	0.49	25
RD22	LMD16	153.70	-	39	59	2	0.41	-	-	-	-	62	0.42	23

\* n=1; ol = olivine; opx = orthopyroxene; cpx = clinopyroxene; pl = plagioclase; bt = biotite.

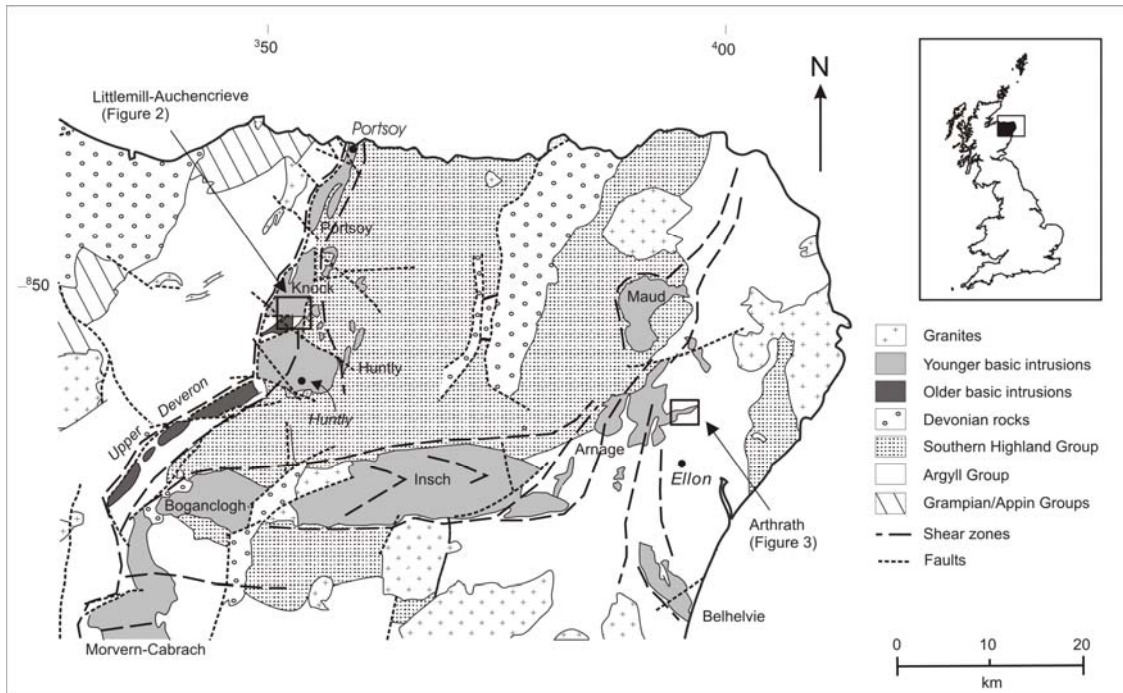
TABLE 2: SUMMARY STATISTICS OF THE GEOCHEMICAL DATA FOR THE ARTHRATH AND LITTLEMILL PROSPECTS.

	Arthrath (n=33)					Littlemill (n=38)				
	min	max	mean	median	90%	min	max	mean	median	90%
Au (ppb)	1	57	12	8	27	1	2669	83	10	30
Pt (ppb)	1	47	11	7	27	1	418	16	3	14
Pd (ppb)	1	456	42	17	93	1	105	23	18	46
Pd/Pt	0.7	228	11.27	2	10	0.08	101	11.36	2.07	37.80
Pt+Pd	2	458	52	24	121	2	451	39	23	83
Fe <sub>2</sub> O <sub>3</sub> (%)	2.22	40.25	15.58	14.71	21.28	7.06	47.38	21.77	15.90	41.42
Cr (ppm)	147	1305	769	772	1195	92	869	360	310	566
Co (ppm)	12	980	203	150	364	25	1990	526	161	1424
Ni (ppm)	126	16940	2930	1661	6772	8	33480	7755	1670	24833
Cu (ppm)	34	5920	1518	1146	3204	10	25600	3198	1102	8758
Cu/ Cu+Ni	0.08	0.67	0.37	0.38	0.48	0.02	0.92	0.40	0.39	0.69
Zn (ppm)	16	111	77	80	96	37	238	118	111	168
Mo (ppm)	na	na	na	na	na	1	33	11	10	23
Ag (ppm)	1	4	1	1	2	1	3	1	1	2
Pb (ppm)	1	41	16	16	27	1	21	9	9	15
As (ppm)	0.05	144.1	5.9	0.8	2.6	0.80	233.2	22.1	12.7	40.4
Sb (ppm)	0.01	25.26	0.82	0.01	0.10	0.02	0.54	0.15	0.11	0.29
Bi (ppm)	0.01	3.96	1.05	0.81	2.09	0.01	9.86	1.36	0.64	3.51
Se (ppm)	0.1	42.5	6.1	3.7	11.3	0.1	78.2	20.1	8.0	50.8
Te (ppm)	0.01	4.71	0.88	0.56	1.69	0.01	3.09	0.66	0.39	1.59
Total S (%)	0.05	21.3	3.7	2.2	8.4	0.06	33.4	8.8	3.2	23.7
S/Se	2500	10000	5985	5977	7153	2750	11000	4980	4401	8000

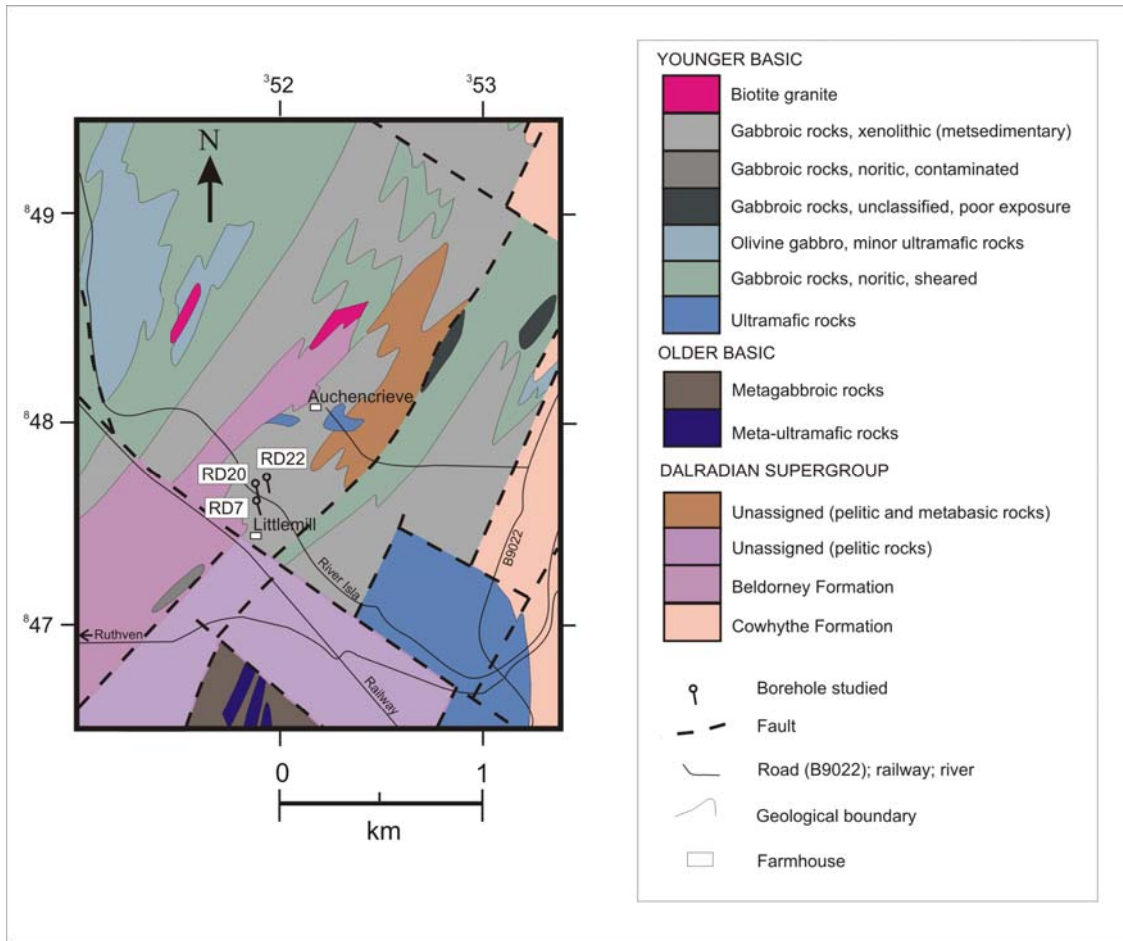
TABLE 3: SUMMARY OF PLATINUM-GROUP MINERAL OCCURRENCES.

BH	Depth (m)	Sample	Platinum Group Minerals	Other(s)	Occurrence(s)
AD25	102.75	ATD33	merenskyite melonite	AgTe mineral; native Au	inclusions in Po/Pn; with Ccp in fracture in plagioclase
AD25	103.00	ATD9	merenskyite	-	inclusions in Po/Pn; sulphide-silicate grain boundaries; in fracture in plagioclase
AD25	103.10	ATD34	merenskyite	hessite; native Au	inclusions in Po/Pn/Ccp
AD25	109.4	ATD10	merenskyite	BiTe mineral; hessite	inclusions in Po; fracture in Po/Pn; sulphide-oxide-silicate grain boundaries
AD25	109.75	ATD36	merenskyite	-	inclusion in amphibole (?)
AD25	176.2	ATD20	merenskyite	-	sulphide-silicate grain boundaries
AD25	176.2	ATD21	merenskyite	-	inclusion in Po; sulphide-silicate grain boundary
AD25	179.3	ATD39	BiPdTe mineral	BiTe mineral	inclusions in Po
AD24	133.6	ATD41	merenskyite		inclusion in Po/Pn
AD24	154.9	ATD42	-	BiTe mineral	inclusion in Po (?)
AD24	165.5	ATD43	merenskyite		inclusion in Po.
AD24	237	ATD44	BiPdTe mineral	BiTe mineral	in qtz-filled fracture in Po; inclusion in Po (?).
RD7	51.8	LMD25	froodite; PdSb mineral	native Au; native Bi; NiAsSb mineral; NiAs mineral;	many grains in fractures in a mylonitic gabbroic xenolith with fracture-filling Po and Ccp.
RD20	117.5	LMD23B	-	native Au.	inclusion in quartz
RD20	138.5	LMD24	michenerite	-	sulphide-silicate grain boundary
RD22	153.9	LMD15B	-	BiTe mineral; native Au; native Bi.	inclusion in Po; many grains in fractured gabbroic xenolith
RD22	158.65	LMD29B	-	native Au.	inclusion in Po (?)

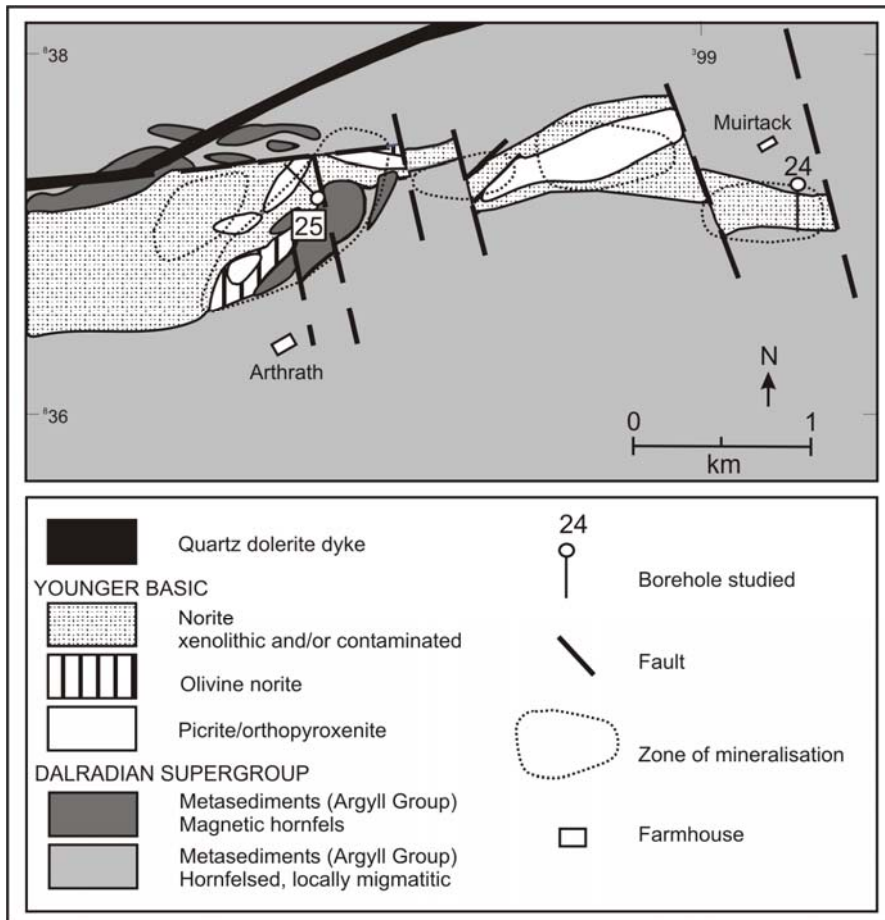
Po = pyrrhotite; Pn = pentlandite; Ccp = chalcopyrite;



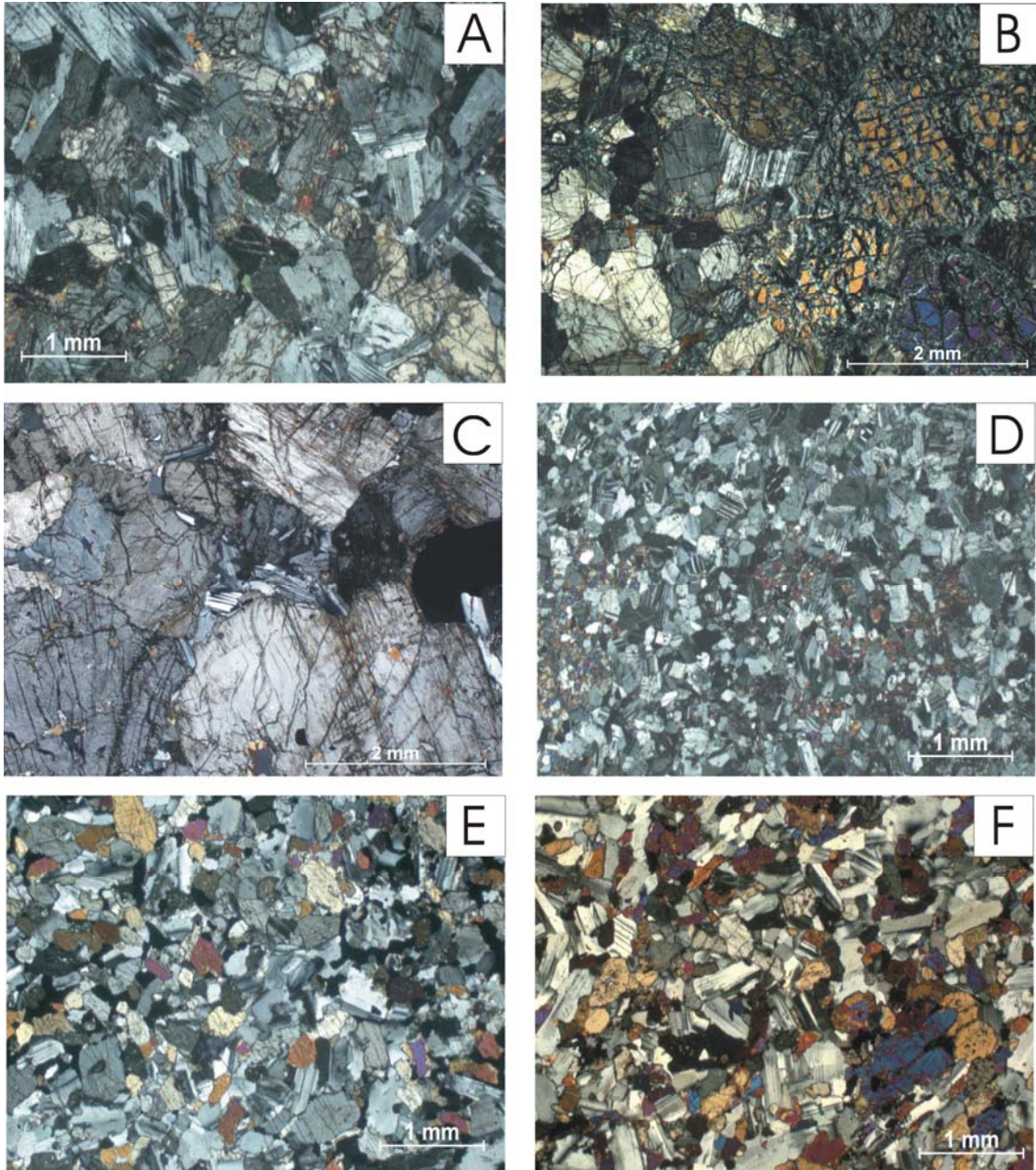
**Figure 1**



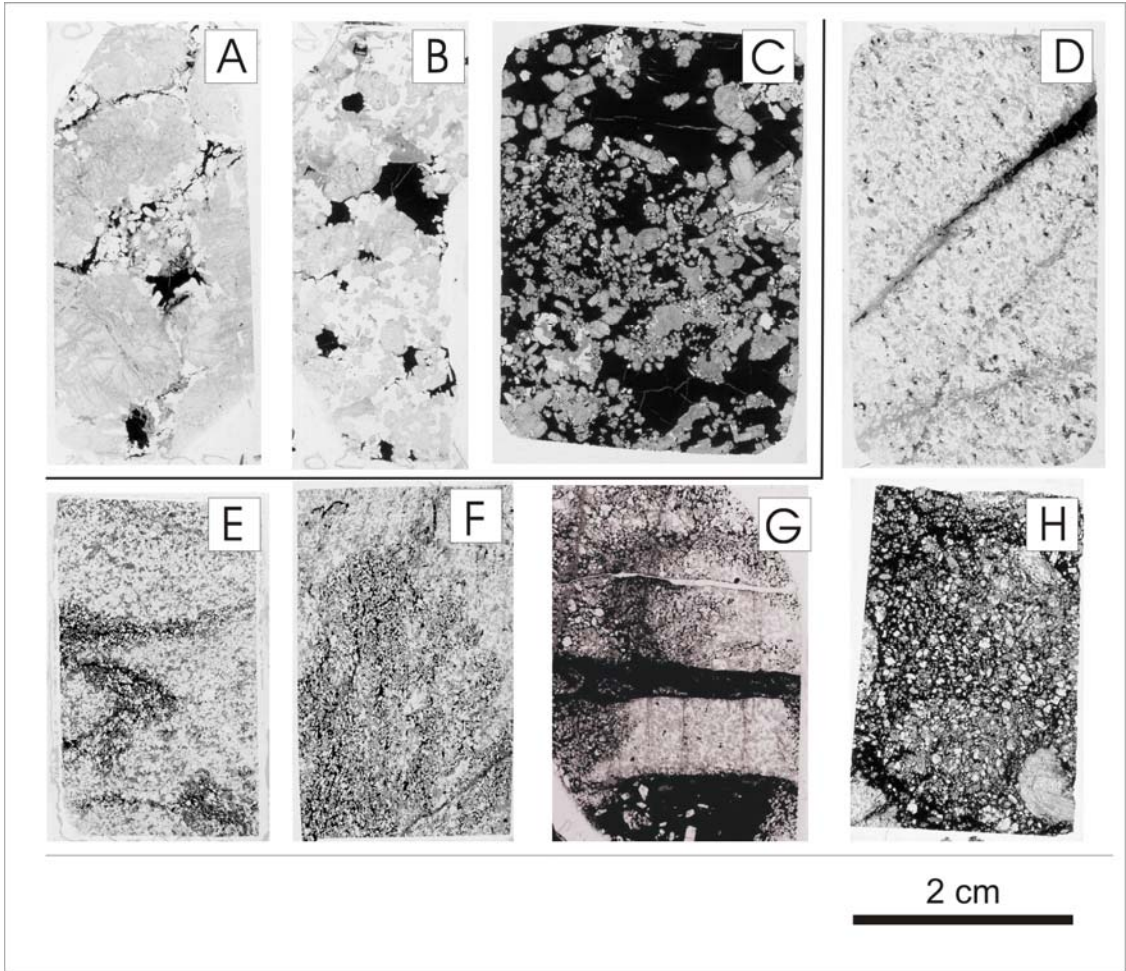
**Figure 2**



**Figure 3**

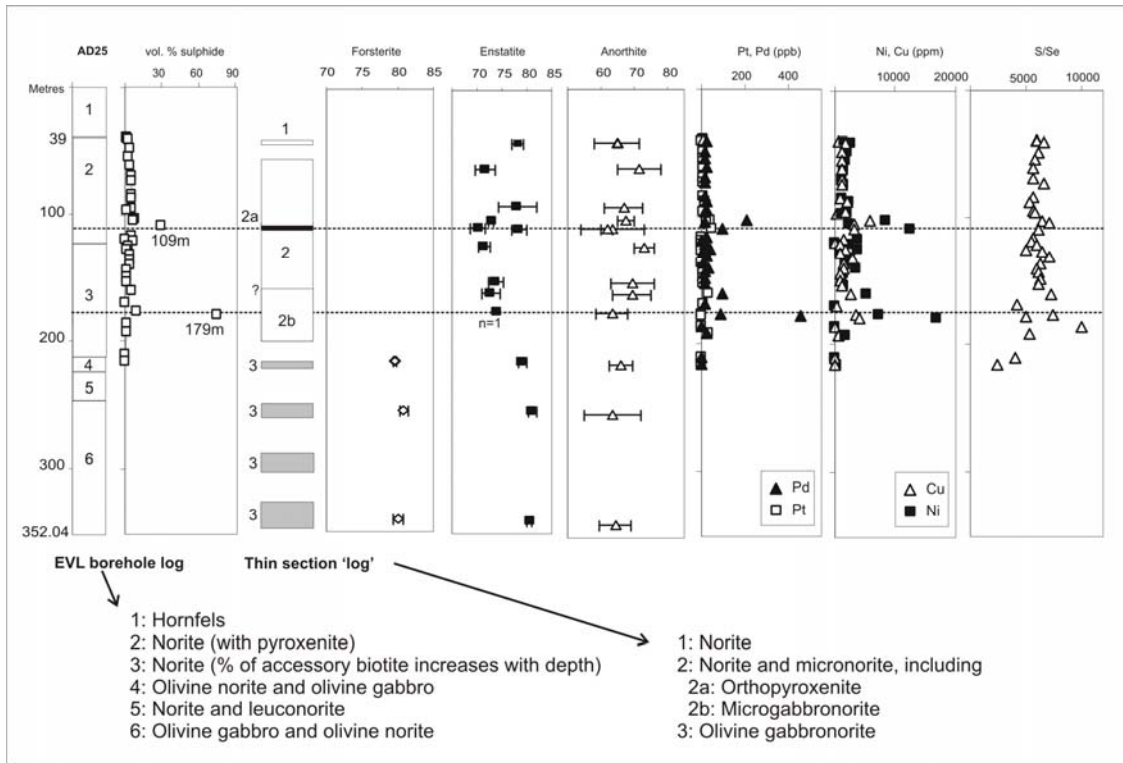


**Figure 4**

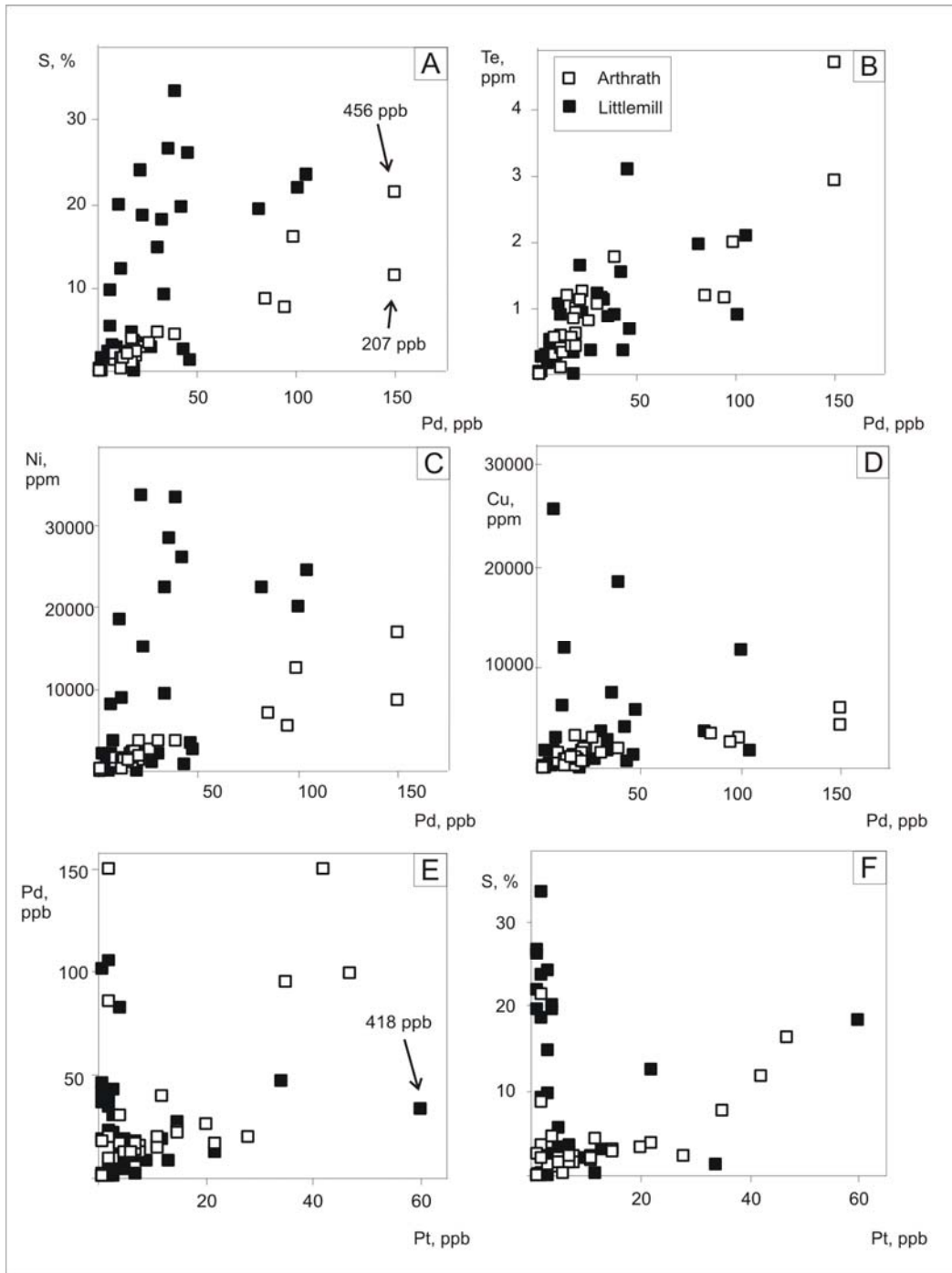


**Figure 5**

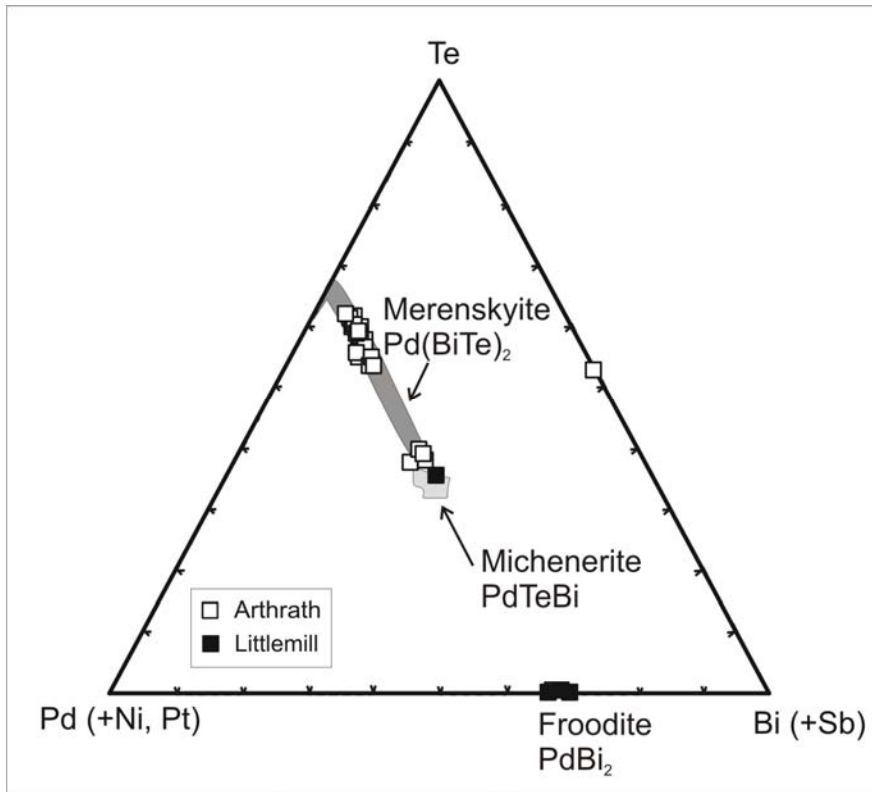




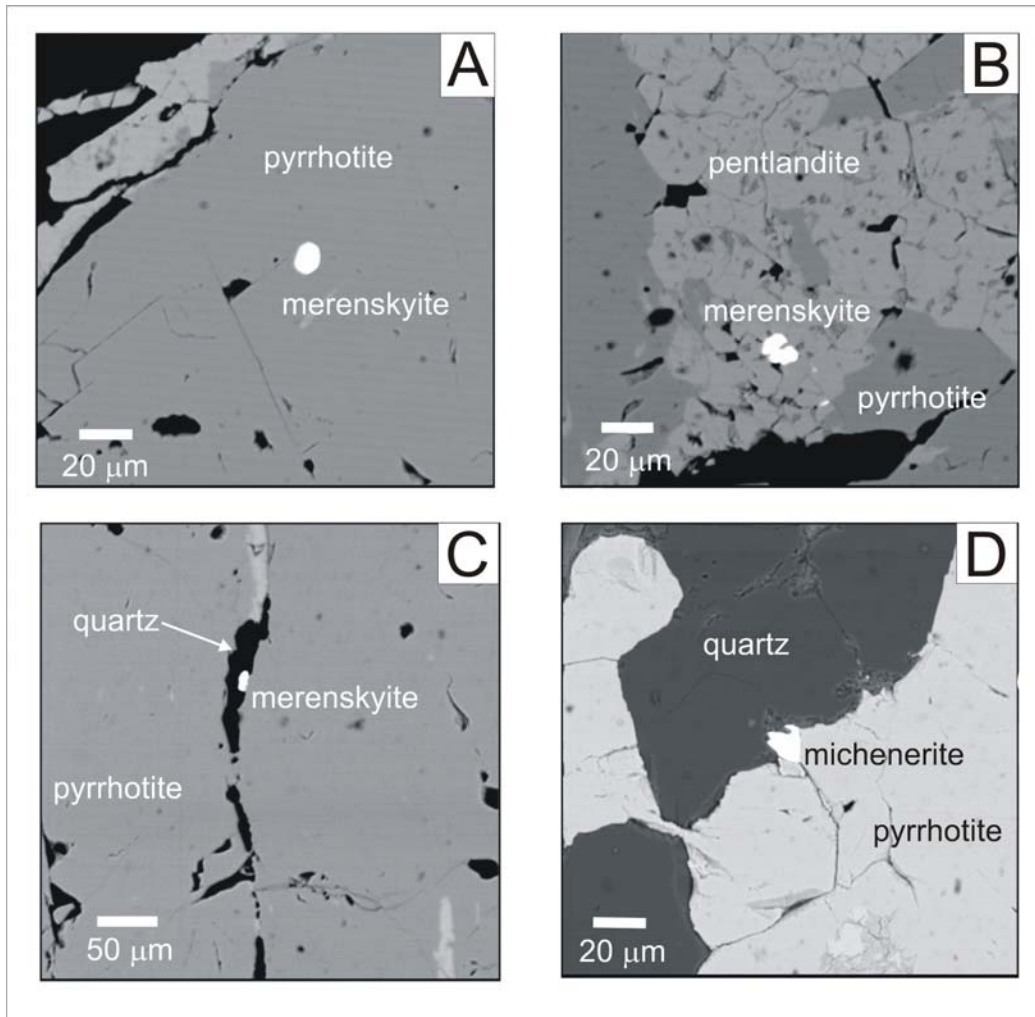
**Figure 6**



**Figure 7**



**Figure 8**



**Figure 9**

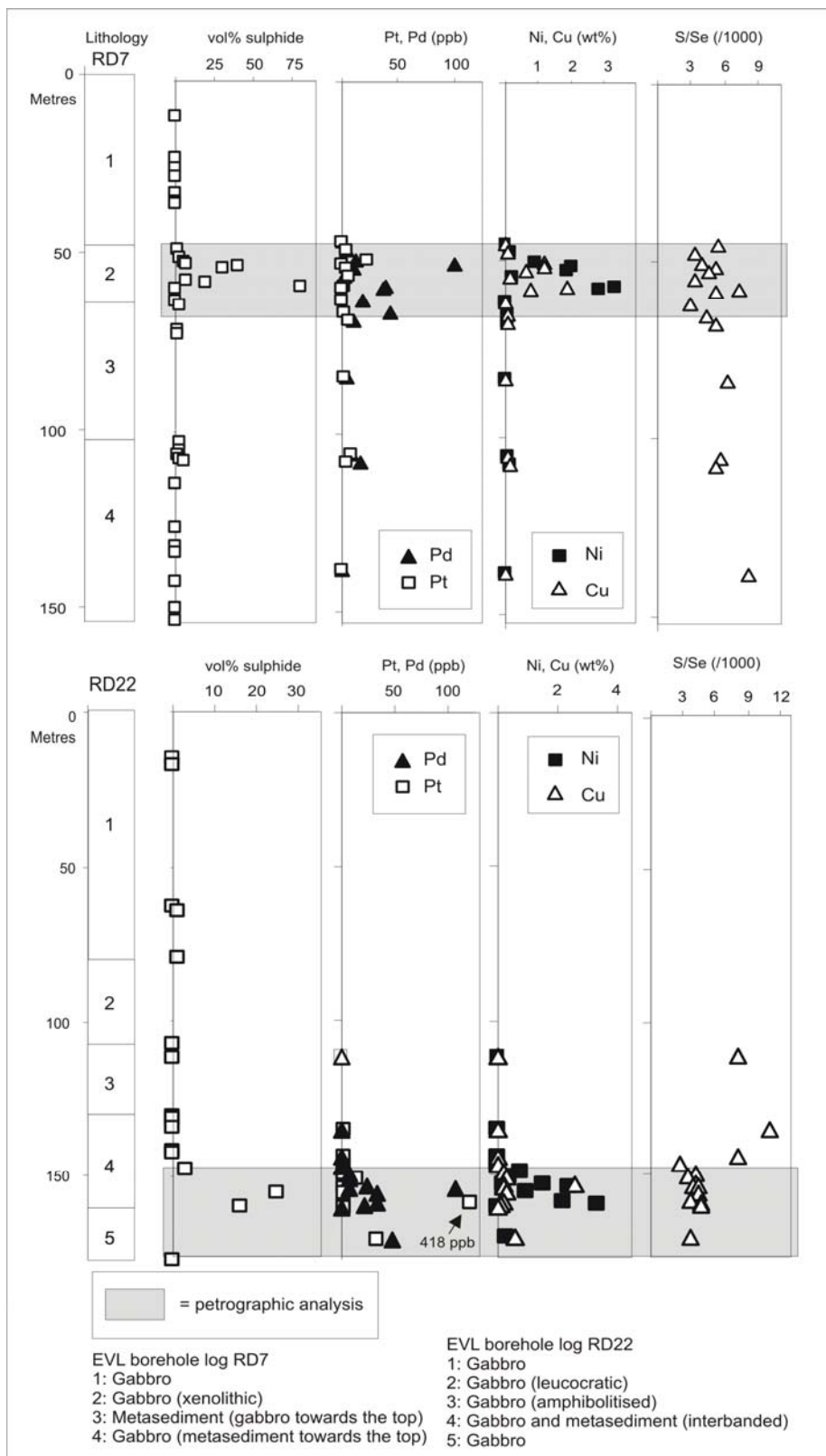
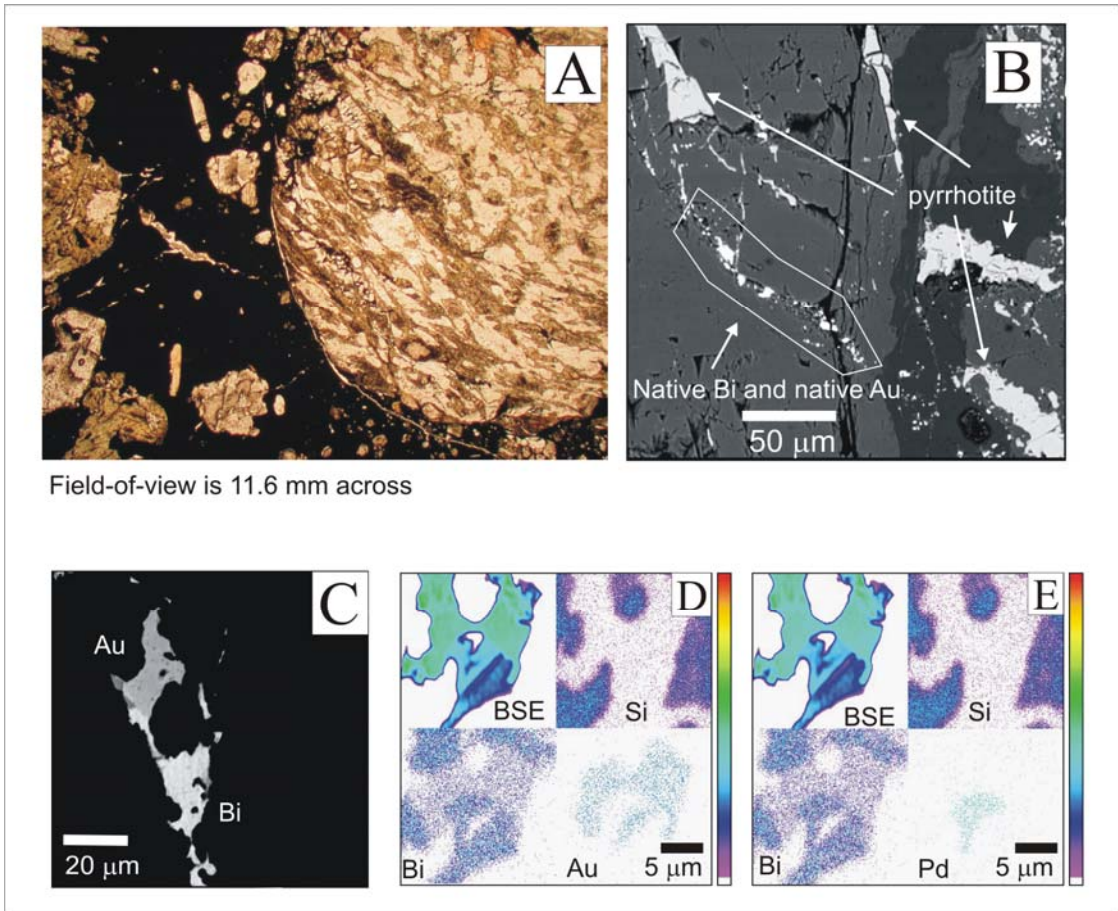


Figure 10



**Figure 11**

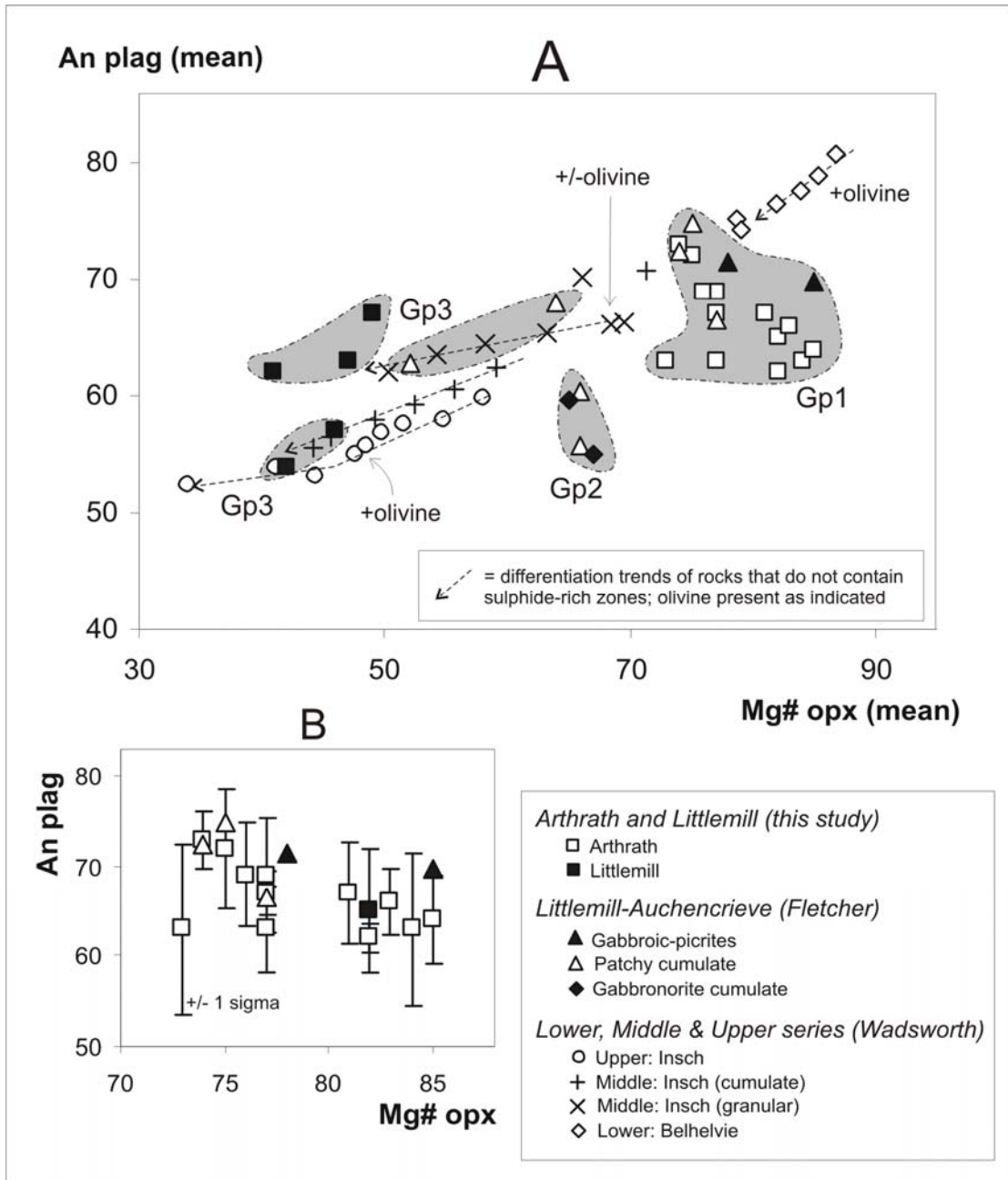


Figure 12