THE MINERALOGY OF BRONZE AGE COPPER ORES FROM THE BRITISH ISLES: IMPLICATIONS FOR THE COMPOSITION OF EARLY METALWORK

Summary. Numerous analytical studies during the latter half of this century have contributed to the compilation of a large compositional database of Early to Middle Bronze Age copper-based artefacts revealing distinctive impurity patterns which appear to change over time. However, attempts to relate these data to copper ore sources proved problematic in the absence of firm evidence for the location of prehistoric copper mines. Over the last fifteen years this situation has changed dramatically with the discovery of numerous Early and Middle Bronze Age copper mines in England and Wales. This study is the first attempt at a comprehensive mineralogical survey of the principal mines investigated to date in order to define the likely composition of the copper ores as mined in antiquity for comparison with the artefact database. The study suggests that the majority of these mines can only have produced essentially pure copper. Only one mine, Ross Island, is likely to have produced copper with a significant level of impurities. The relative purity of the known ore sources is contrasted with significant levels of various metallic impurities among the analysed artefacts, leading to the conclusion that metal circulation and mixing may have been more extensive than previously thought even during the earliest part of the Bronze Age.

INTRODUCTION

Recent years have seen the development of a considerable research effort aimed at the investigation of field evidence for prehistoric copper production in Britain and Ireland. This has resulted in the discovery of a substantial number of sites at which Bronze Age copper mining is either confirmed or suspected. To date little evidence has come to light for copper smelting or fabrication processes, although one site, Ross Island, has recently yielded tantalizing evidence of onsite metal production in the form of a metal droplet.

Until the early 1980s the only Bronze Age mine site that was generally accepted in the British Isles consisted of a cluster of primitive openworks and shallow galleries at Mount Gabriel on the Mizen peninsular in south-west Munster. Today, thanks very largely to the efforts of a relatively small number of dedicated field workers, some
thirty probable or definite prehistoric copper mining sites have been identified in the British Isles. Many of these are based on surface outcrops at sites which, in the historic period, became well known and highly productive. In addition to Mount Gabriel, the best known and best investigated sites to date are the Great Orme and Parys Mountain in North Wales, Cwmystwyth in central Wales, Alderley Edge in Cheshire and Ross Island near Killarney in south-west Ireland (Fig. 1). The results of recent fieldwork at these sites have recently been summarized in a useful short book by O’Brien (1996) and in works by Timberlake (1992), Craddock (1995) and Gale (1995).

The antiquity of mining at these sites has been established from multiple radiocarbon determinations on material, mostly charcoal derived from firesetting, within secure...
contexts in mining spoil. The radiocarbon results (Table 1) suggest that the principal prehistoric extractive activity at all of the sites investigated in detail took place within the Bronze Age. In addition, Ross Island features a ‘work camp’ area adjacent to the mine from which a series of radiocarbon dates have also been obtained. Ross Island appears to be the earliest site yet excavated with dates clustering in the second half of the third millennium BC, corresponding to the early copper-using horizon, just prior to the beginning of the Early Bronze Age, in Ireland; the period associated with the introduction of metallurgy in the British Isles. The other sites, some of which appear to have been in use over extensive periods of time, are approximately coeval and have overlapping date ranges spanning the Early Bronze Age and earlier Middle Bronze Age from c.1900–1200 BC. Although a number of the sites (including Ross Island) have now produced evidence for post-Bronze Age, pre-industrial activity, no copper mining site has yet come to light with evidence for extensive Late Bronze Age or Iron Age working.

Understandably, in the initial phase of investigation, the focus of research has been to outline the extent and form of mine workings and the technology and practice of extraction which they represent. This is now being expanded with systematic programmes of investigation of stone mining tools (Gale 1995) and, where they occur, associated habitation and work areas (O’Brien, pers. comm.). Hitherto, however, there has been comparatively little detailed study of the mineralogy of the deposits mined in prehistory. A full understanding of the detailed mineralogy and petrography, and therefore composition, of the ore ‘as mined’ is an essential prerequisite to studies aimed at linking known mine sites to coeval metalwork (Ixer 1995). The work reported here is part of a larger study intended to describe the composition of the ores mined from some of the better investigated Bronze Age mine sites and model their chemical alteration in beneficiation and later smelting. In this way it is hoped to develop a more detailed understanding of the interpretation of artefact compositional data in provenance studies. This adds to a continuing reassessment of framework assumptions underlying the interpretation of metalwork analytical data (Budd and Taylor 1995; Budd et al. 1993, 1994, 1995, 1996).

Most analytical studies of Bronze Age metal artefacts have involved grouping those of similar composition or examining existing typological groups for compositional similarities. In the British Isles the latter approach has been applied by Northover (1980, 1980b, 1982) and Needham et al. (1989) who were able to outline significant changes in the impurity pattern of copper-based metalwork over time and between different regions. Although it has been possible to comment on the possible provenance of the copper used in various areas and periods on the basis of artefact distribution patterns and the general pattern of copper mineralization in the British Isles, there has, until now, been little opportunity to focus on specific mine sites. Now, for the first time, it is becoming possible to develop a clear idea of the impurity patterns likely to have resulted from smelting the ores from particular sites which we know to have been exploited in the Early and Middle Bronze Ages. A number of the best investigated mine sites are briefly described below together with an outline of their geology and the mineralogy of the deposits. Conclusions are drawn on the likely make-up of the ores as they would have been mined in antiquity and the implications for provenance studies are discussed.

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<table>
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<tr>
<th>Site and Sample</th>
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<th>Description</th>
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Prehistoric copper mining at Alderley Edge has recently been described by Gale (1995, 79–91) and O’Brien (1996, 52–5). Alderley Edge, about 20 km to the south of Manchester (Fig. 1), is a prominent north-facing scarp rising steeply from the Cheshire plain. Copper, lead, and later cobalt, were extracted in the historic period from the late seventeenth century AD and, on a small scale, throughout the eighteenth and early nineteenth centuries from mineralized veins on the scarp and its margins. The small mining area (Fig. 2) extends to the east of Wood Mine and to Mottram St. Andrew some 1.5 km to the north-east. The early historic mining operations concentrated on mineralized faults and their immediate surroundings, but from the mid-nineteenth century AD, the introduction of acid ore treatment made it economically viable to process low-grade disseminated copper in the sandstones.

New mining strategies, developed as a result of the acid treatment process, led to the discovery of much earlier mine workings and stone mining tools in the later nineteenth century AD. Old pit workings with stone mining hammers were first discovered in 1874 (Boyd Dawkins 1875, 1876). A wooden spade recovered from these excavations (Sainty 1878, 64) was lost for many years until recently rediscovered (Garner et al. 1994). The tool has now been radiocarbon dated to 1977–1526 BC (2σ error) (OxA-4050) corresponding to the earlier part of the Middle Bronze Age. Further antiquarian interest around the turn of the century led to the discovery of numerous stone mauls from recent mine tips at various other sites around the Edge including Windmill Wood, Engine Vein and Dicken’s Wood as well as at Mottram St. Andrew (Roeder 1901; Roeder and Graves 1905). At Engine Vein, Roeder and Graves recorded five superficial mining pits which survived in section in the north face of the openwork. Later examination of this mining face (Gale 1986, 1989) suggests that they are prehistoric and that their form was preserved because they remained full of mine debris until the latter half of the nineteenth century when the stopes were cleared of their deads and timber.

Limited excavations of surface workings at Brynlow, Wood Mine and along the Engine Vein fault in 1991 failed to produce further evidence for prehistoric mining (Gale 1993), and Gale (1995, 90) concludes that prehistoric mining, whilst it may have been widespread at Alderley Edge, was limited in intensity. The site is currently the subject of a detailed survey and recording programme organized by the National Trust and Manchester Museum (Timberlake, pers. comm.) which may help to define further the nature and extent of Bronze Age activity. The recent discovery of a coin hoard in a ceramic vessel in the fill of what had previously been taken to be a square profile, pick-cut, eighteenth century shaft just to the north of the Engine Vein openwork has now raised the possibility of Romano-British activity at the site. The copper alloy coins have been provisionally dated to the third century AD, but are currently in the process of conservation and are awaiting further study. Although intriguing, the coins have not, to date, provided conclusive proof of Roman mining at Alderley Edge as they were a chance find made during work to cap and clear the shaft and their exact stratigraphic relationship with it is not yet clear.

The diagenetic-epigenetic copper-lead-baryte mineralization of Alderley Edge is hosted by the Triassic Helsby Sandstone Series. Primary mineralization, comprising lead and copper sulfides (galena and
Plan of the main historical mining area at Alderley Edge in Cheshire. Selected surface features are shown together with some of the main eighteenth–nineteenth century AD workings. The plan is redrawn, with permission, from Carlon (1979, 38).
chalcopyrite) and baryte as disseminated pore-filling cements and grain replacements, is restricted to within 5 m of a number of WNW-ESE-trending fault zones; mineralization is more intense on the footwall side of the faults. Locally, replacement of the footwall sandstones by galena and baryte is intense, so giving the appearance of vein-like ore bodies; although true open-void vein-infilling is rare and only of minor importance. Secondary disseminated copper mineralization, mainly as carbonates and silicates (malachite and chrysocolla), is more widespread, infilling intergranular void spaces within porous sandstones.

The mineralization, which belongs to the ‘red-bed’ class of deposit (Ilexer 1990; Craig and Vaughan 1994; Rowe and Burley 1997), has been described by Warrington (1965), Carlon (1979), Thompson (1991) and most recently redescribed and reinterpreted by Rowe and Burley (1997). The primary mineralogy has been discussed by Ilexer and Vaughan (1982) and Holmes et al. (1983) and the secondary and supergene mineralogy by Braithwaite (1994).

Ore grades for both types of ore are low with sulfides ranging from 2% by volume up to 40% close to the fault zones (Rowe and Burley 1997). Massive ores (>50% by volume) are rare and dominated by galena and baryte rather than copper minerals. In both ore types, the simple primary sulfide mineralization is dominated by galena and chalcopyrite with minor sphalerite, nickelooan, pyrite (bravoite) and trace amounts of the nickel-cobalt-iron arsenides and sulfarsenides (gersdorffite, cobaltite, pararammelsbergite), arsenic-poor tetrahedrite (Ilexer and Vaughan 1982; Ilexer 1990) and enargite (despite Braithwaite (1994) showing that earlier records of this mineral were incorrect). An extensive suite of supergene and secondary minerals is present and is especially associated with the disseminated ores. These minerals include the copper sulfides djurleite, covellite and spionkite as well as lead, copper and zinc carbonates, sulfates and a large number of rare to very rare species. Amongst these rarities are a few lead, iron, cobalt and copper arsenates such as erythrite (cobalt) and olivenite (copper) (Thompson 1991; Braithwaite 1994). Indeed, the only potentially arsenic-bearing minerals that are widespread at Alderley Edge are members of the pyromorphite-mimetite series and even these are relatively arsenic-poor (Braithwaite 1994).

Systematic mineralogy of both the primary and secondary ores clearly shows that faehlerz ore is missing, although minerals of the faehlerz group (tetrahedrite and enargite) are present in microscopical amounts. Hence, arsenic is a very minor component of the ore at Alderley Edge despite the suggestion of Dewey and Eastwood (1925), repeated by Eager and Broadhurst (1959) and Warrington (1965), that the ores were arsenic-rich; a suggestion based on the yellow colour of some of the ores and a chemical analysis of a single hand-specimen. The proposal, based on these reports, by Pollard et al. (1991) and Budd et al. (1992) that Alderley Edge may have been a potential ore source for arsenic-bearing copper artefacts can therefore be discounted.

In summary the main copper ore minerals at Alderley Edge are malachite, djurleite/chalcocite and, more locally, azurite, chalcopyrite and chrysocolla. These minerals carry few other metals other than iron (chalcopyrite) and trace amounts of silver (djurleite/chalcocite), so any metal made from them would be free of significant metal impurities. Potential trace elements could have come from the other ore minerals, but only galena (PbS), sphalerite (ZnS) and pyrite/nickelooan pyrite/bravoite (FeNiCo)S₂ are present in
sufficient quantities that they could contribute to the composition of any metal; that contribution would, however, be extremely small.

THE GREAT ORME

Prehistoric mine workings on the Great Orme have been described in detail by a number of researchers (James 1988, 1990; Dutton 1990; Lewis 1990, 1993, 1994; Jenkins and Lewis 1991; Dutton et al. 1994; Gale 1995, 51–60; O’Brien 1996, 47–51). The Great Orme’s Head is a Carboniferous Limestone promontory rising to 207 m OD and projecting from the North Wales coastline just west of Llandudno (Fig. 1). The headland features a number of mineralized veins striking roughly north-south with steep dips to the west. Four major copper-bearing lodes, as well as at least ten minor veins have been mined for copper. The site was worked in the historic period between the seventeenth and nineteenth centuries and these operations, described in detail by Williams (1979) and Smith (1989), extended

Figure 3
Simplified oblique view of some Bronze Age workings around the 10 m level of the post-industrial Vivian’s shaft at Great Orme near Llandudno. The drawing is reproduced, with permission, from Lewis (1990).
for over 1 km from St. Tundo’s Church in the north, to Maes-y-facrell in the south, ultimately attaining depths of 70 m below sea-level.

As with many copper mining sites now known to have been worked in the prehistoric period, accounts of nineteenth century AD miners intersecting earlier workings attracted initial interest in the possibility of early mining. Mid-nineteenth century accounts describe old stopes with bone and stone tools plus a few bronze artefacts and charcoal from fires (Stanley 1850). Also, in common with many of the sites described here, the Great Orme was inspected by Oliver Davies in the 1930s as part of a programme supported by the British Association for the Advancement of Science. Systematic exploration of the underground workings started in the mid 1970s. A 1987 mining survey allowed access to three previously sealed shafts with linked workings. Initial investigations by James (1988) revealed a system of horizontal galleries up to 50 m in length reaching a vertical depth of some 30 m and associated with characteristic prehistoric spoil. An important initial indication of the antiquity of the early mining phase is the presence of characteristic mine spoil (comprising angular country rock and vein debris) in stratified sequences which incorporate bone and stone cobble tools as well as the ubiquitous charcoal associated with firesetting. This backfilling was capped and partially cemented by flowstones of dimensions normally associated with formations in natural caves.

An ongoing programme of systematic underground investigation and removal of surface spoil to facilitate the development of the site as a mining heritage centre has revealed more of the early workings and produced stratified material for radiocarbon dating from which the workings have been dated broadly to the Early to Middle Bronze Age (Table 1). It is estimated that between 5 and 10 km of prehistoric underground passages were developed. These extend laterally over 240 m and to a 70 m depth (Lewis 1990, 1993, 1994; Dutton et al. 1994). The prehistoric underground workings are of quite distinctive form (Fig. 3) and very different from the later historic workings. Gale (1995, 28) has pointed out that the prehistoric mine workings so far described in the British Isles share the characteristic of being purely extractive in nature, involving the extraction of the minimum of barren ground in order to extract useful ore-bearing material. The prehistoric workings at the Great Orme conform to this pattern. They are typically small and tortuous with extraction restricted to veins and the minimum possible development in country rock to allow access. Prior to seventeenth–nineteenth-century dumping, numerous rock exposures of the limestone strata in the Pyllau valley would have been apparent (Lewis 1990, 1993) and Gale (1995, 53) suggests that the original land surface may have taken the form of a series of stepped scars from which the copper-bearing lodes were worked initially by trenching and open casting. Dutton et al. (1994) have suggested that an underground karst drainage system may have existed providing direct underground access in prehistory. The extraction of secondary copper minerals from weathered dolomite is likely to have been a relatively simple operation, but further development into hard rock probably led to the increased use of firesetting and stone tools.

The geology of the Great Orme mines and the field relations between the mineralization and wall rocks have been described in Dutton et al. (1994) but is better in Lewis (1994) and is summarized in his figure 2 (Lewis 1994, 32). The ores have been described by Ixer (1994a), Joel et al. (1997), Ixer and Stanley
Epigenetic copper mineralization cuts and infills Dinantian and Brigantian dolostones and limestones of the Carboniferous. It is Britain’s first copper-dolomite association deposit (Ixer and Stanley 1996) rather than belonging to the lead-zinc-rich Mississippi Valley-style deposits as previously believed (Ixer and Vaughan 1993; Lewis 1994; Dutton et al. 1994).

The primary ores are very simple, namely saddle dolomite-chalcopyrite-minor pyrite/marcasite-calcite, but these have been extensively altered to a supergene assemblage of malachite and limonite accompanied by minor amounts of copper sulfides, azurite and trace amounts of silver-bearing phases, native copper and copper oxides (Ixer and Davies 1996). Trace amounts of cobalt and nickel are present in material collected from the nineteenth century AD mine tips (Ixer and Stanley 1996), but may not be representative of the Bronze Age ores. Indeed these ores appear to be rare. Two main ore types would have been available to Bronze Age miners: secondary malachite-calcite ore, irregularly present within void spaces or along minor joints in dolostones; or thin chalcopyrite-malachite-copper-bearing limonite veins cross-cutting limestones and dolostones. Although both are viable ores, the thin but continuous chalcopyrite-malachite veins would have provided a continuity of supply and their exploitation would best explain the 5 to 10 km of narrow Bronze Age mining passages believed to be present in the mine. Azurite is rare and mainly confined to one thin, non-carbonate-bearing shale horizon (Ixer and Davies 1996) and hence its role, if any, is unlikely to be as important as suggested by Lewis (1994) and Joel et al. (1997).

Galena with minor chalcopyrite is rare within the mine, being present in a single thin vein next to argillaceous dolomitized limestones. It has no importance in terms of any potential copper ore and indeed belongs to an earlier and quite different type of mineralization, namely the Mississippi Valley-style lead-zinc ores of the north-west Wales orefield (Ixer and Vaughan 1993). The efficient separation of galena from copper ores by Bronze Age miners, as seen at Cwmystwyth, suggests that it was highly unlikely that any galena was exploited from the Great Orme during the Bronze Age and even less likely that it could have been accidentally incorporated into the mined ore.

In both the primary chalcopyrite-rich and supergene malachite-limonite-rich ores the only contaminant metal is iron (chalcopyrite, pyrite/marcasite and limonite). All other metals including Co, Ni, As and Ag are only present in parts per million and no antimony-bearing phases have been recognized. Hence whichever of the two principal ore types was used, no significant contaminant metals would have been present and it seems likely that the resulting copper would have been substantially free of ore-derived, metallic impurities.

**PARYS MOUNTAIN**

Prehistoric copper mining at Parys Mountain has been described by Timberlake (1990a) and Gale (1995, 69–75). Parys Mountain is located in north-eastern Anglesey near Amlwch and rises to 147 m OD (Fig. 1). Intensive copper mining was carried out at the site during the late eighteenth and early nineteenth centuries AD with output from the mine dominating the World copper market during the earlier part of this period. Today the site is characterized by two enormous opencasts surrounded by extensive mineral tips resulting from this activity.
As with most of the prehistoric mine sites investigated to date, there are eighteenth and nineteenth century AD references to ‘old man’ workings pre-dating the historic mining. Cobblestone tools were noted from the area in the mid-nineteenth century (Stanley 1850) and the site was one of those investigated by Davies in the 1930s (Davies 1939). The northern side of the mountain is least disturbed by the historic mining operations and it is in this area that attention has been focused by those investigating earlier activity. Davies excavated an ancient tip just to the north of the summit windmill from which 24 hammerstone tools were recovered. Lenses of charcoal within the tip suggested that the material was removed by firesetting, but in the absence of datable material Davies was unable to be specific about the antiquity of the tip.

In 1988 the tip investigated by Davies was re-excavated by Timberlake (1990a). Its location suggested that it must have been derived from a working within the ‘Oxen Quarry’ (a shallow depression about 15 m deep immediately to the south), although stone quarrying in the historic period had removed any evidence of early mining. Pyrite- and, locally, chalcopyrite-bearing quartz veins are known to crop out in Ordovician shales on this part of the site. Re-excavation of the tip revealed undisturbed early waste underlying more recent and disturbed material. These lower layers included crushed veinstuff, charcoal fragments and cobblestone tools. Charcoal removed from this material for radiocarbon dating produced Early to Middle Bronze Age dates (Timberlake 1989) (Table 1). The prehistoric tip appears to be distinguished only by its small size and low profile and by the presence of stone tools and tool fragments. There is close vegetation cover over much of the site today and it is clearly possible that further Bronze Age tips remain to be discovered beneath the extensive historical mining spoil. No evidence of in-situ prehistoric workings have been identified and, in view of the extent of the historic activity, this seems unlikely to change.

The mineralization at Parys Mountain is different from that at any other British Bronze Age copper mine in that it is characterized by a wide range of polymetallic ores that were present in Lower Palaeozoic host rocks. It is a synsedimentary volcanogenic deposit of Ordovician-Silurian age showing some similarities with the Tertiary Kuroko deposits of Japan (Pointon and Ixer 1980). The primary ore was partially remobilized into a series of cross-cutting quartz-sulfide veins during the Caledonian Orogeny (Greenley 1919; Nutt et al. 1979) so that the resulting ore types have a very complex spatial distribution and mineral assemblage.

The geology, mineralization and genesis of Parys Mountain have long been contentious and have been described by Wheatley (1971), Pointon and Ixer (1980), Southwood (1982), Westhead (1991) and are currently being re-evaluated by Charter et al. 1996). The ores have been described in detail by Pointon and Ixer (1980) and Ixer and Pointon (1980), whilst lists of primary and secondary minerals are given in Southwood and Bevins (1995). At least three major ore types are present: silicified pyritic ore with a low copper content; chalcopyrite-rich ore with pyrite and minor amounts of other sulfides; and bluestone ore — a complex, fine-grained intergrowth of galena-sphalerite with minor pyrite and chalcopyrite and significant amounts of silver, antimony and arsenic.

Unsurprisingly, the evidence for Bronze Age mining at Parys Mountain has been almost totally obliterated by the post-industrial activity which concentrated on the chalcopyrite-rich ores and bluestone ores.
leaving much of the pyritic ore in situ. However, the excavations by Timberlake (1988a, 1990a) suggested that the Bronze Age mining activity centred on the pyrite-chalcopyrite-quartz veins on the northern slopes of the hillside away from the main copper and bluestone ores. This suggestion accords with the mineralogy of the ores; the very hard pyrite-silica ores have a low copper content and so were unattractive and difficult to mine and the bluestone ores, carrying only c.2 wt.% copper, are unlikely to have been amenable to prehistoric beneficiation and smelting. Indeed, even during the nineteenth century AD mining, the bluestone ores were used as road metalling (Southwood and Bevins 1995). The copper-pyrite ores have an average grade of 3.5 wt.% copper (Greenley 1919) and contain minor amounts of sphalerite and galena and lesser amounts of tetrahedrite-tennantite and arsenopyrite (Wheatley 1971; Pointon and Ixer 1980).

As all of the natural outcrops at Parys Mountain show strong signs of glaciation, as at Morfa du for example, it is unlikely that Bronze Age miners were presented with much gossan, although Lentin (1800) noted the presence of tenorite and native copper in gossans ‘beneath the soil’ above copper-rich portions of The Great Lode. However, as Timberlake (1994) suggests, in the absence of strong evidence for the exact location of Bronze Age mining, and therefore of exactly which ores were exploited (although sulfides rather than secondary minerals seem more likely), it is not possible to be sure of the composition of metal smelted from Parys Mountain ores during the Bronze Age. There is therefore the possibility that ores from Parys Mountain could produce copper with detectable contaminant metals including lead, zinc, arsenic, antimony and silver but not cobalt nor nickel.

COPA HILL, CWMYSTWYTH

The prehistoric copper mines on Copa Hill, Cwmystwyth have been described in detail by Timberlake and co-workers (Timberlake 1987, 1988b, 1990b, 1991a, 1991b, 1993; Timberlake and Mighall 1992; Timberlake and Switsur 1988) with more general descriptions by Gale (1995, 61–8) and O’Brien (1996, 42–5). The site is in central Wales, approximately 23 km east of Aberystwyth (Fig. 1). A number of mineralized lodes at Cwmystwyth were mined for lead in the seventeenth–nineteenth centuries AD (Hughes 1981). Prehistoric activity, however, appears to be restricted to a single opencast working where the copper-bearing Comet Lode crops out on the scarp slope of Copa Hill high above the Ystwyth valley at approximately 420 m OD (Fig. 4). Prehistoric activity appears to have been entirely restricted to copper extraction.

Stone tools associated with an old working at Cwmystwyth were noted in the nineteenth century AD (Smythe 1848) and the site was investigated by Davies in the 1930s (Davies 1946). He excavated some small trenches and recovered a number of stone tools, but was not convinced of the antiquity of the site. More recently systematic investigation dates from 1986 with small-scale excavations and survey work by Timberlake (1987) which led to radiocarbon determinations on three charcoal samples sealed with the tip material (Timberlake 1988b; Timberland and Switsur 1988; Ambers 1990) dating the mining activity to the Middle Bronze Age (Table 1).

The prehistoric opencast on Copa Hill is approximately 45 m in length, up to 17 m wide and of uncertain depth (Timberlake 1988a). Downslope there are extensive tips of mine spoil extending for some 85 m. Hundreds of cobblestone tools and tool fragments have been recovered from this
material. Ongoing fieldwork by the Early Mines Research Group has concentrated on the investigation of the form and infill of the working. Excavations in 1989 adjacent to the north wall of the opencast revealed the top of a small fireset gallery-type working at a depth of approximately 1.5 m. The gallery was thoroughly investigated in 1990 and found to be irregular in shape, some 1.5 m in depth and height and 2 m wide, corresponding to the shape of the extracted vein pocket. Indications of pounding by stone tools were noted on the walls of the gallery and main working and an in situ peat deposit was revealed on the floor of the gallery working (Timberlake 1990a). These excavations also showed that the main working was at least 4–5 m in depth. Subsequent work (Timberlake 1993) has increased this figure to at least 7 m from the modern land surface giving a total depth of some 12 m from the top of the opencast.

The infill consists of shale scree and silt with interspersed peat horizons. Further radiocarbon dates have been obtained for three of these layers and for two contexts at the floor of the working. The stratigraphy and the dating evidence supported the proposal that the opencast is essentially an intact Bronze Age working progressively infilled after abandonment (Timberlake 1990b, 1992). This interpretation has been strengthened by

Figure 4
Simplified section and oblique view of the Bronze Age opencast at Copa Hill, Cwmystwyth showing the location of key radiocarbon samples. The drawing is reproduced, with permission, from Timberlake (1994).
fieldwork in 1993 (Timberlake 1993) which exposed two entrance areas at the lower end of the opencast with evidence of pounding marks from stone tools. Workings in this area were found to follow mineralized veins leaving barren rock undeveloped. A small working area was also located with a rock-cut bench associated with stone tools, fireset debris and crushed quartz. The excavation also led to the recovery of a number of wooden objects including two launders made from split and hollowed trunks. One of these appeared to be in situ, lying on the rock cut floor of the entrance. One of the launders has now been radiocarbon dated to the Early Bronze Age (Timberlake 1993) (Table 1). There has been some disturbance to the opencast as a result of operations in the historic period, most notably by an intrusive timber feature interpreted as a medieval shaft (Timberlake 1991b) and by the construction of a dam on the downslope side of the working in order to create a reservoir for hushing.

The Comet Lode on which the Bronze Age mining activity is based is part of the major Ystwyth fault on the southern margin of the Central Wales Mining District. Within this orefield, polyphase, epigenetic, ENE-WSW-trending lead-zinc-copper sulfide-bearing quartz-ankerite veins cross-cut Ordovician-Silurian metasediments. Despite a long history of exploitation, the orefield is not well known, but its ores and mineralogies have been described by Raybould (1974) and, more recently, by Mason (1994, 1996). A brief account and map of the orefield is given by Bevins (1994).

Chalcopyrite-rich ores occur separately from the galena-rich ores at the Copa Hill mine site. In the copper ores, chalcopyrite is the main sulfide accompanied by nickel-bearing pyrite, galena, chalcopyrite disease-free sphalerite and minor marcasite as well as a white cubic mineral with the optical properties of ullmannite. Alteration/weathering of chalcopyrite has formed the secondary iron-copper sulfides bornite and ‘idaite’, plus the secondary copper sulfides digenite, covellite, spionkopite and the oxides cuprite and limonite. Galena has altered to anglesite and cerussite. The galena-rich ores comprise galena and minor chalcopyrite with trace amounts of pyrite and the mineral provisionally identified as ullmannite. Here galena also shows extensive alteration to covellite, anglesite and cerussite.

The copper ores of Copa Hill would produce few metallic impurities if smelted, only iron with traces of nickel (nickeloan pyrite, ullmannite) and antimony (ullmannite); arsenic is absent from the mineral suite. The earlier identification of arsenopyrite from the orefield (Raybould 1974) has been shown to be a misidentification of a number of antimony-bearing minerals (Mason 1996). Indeed, the results of the exhaustive mineralogy of the ores of the Central Wales Orefield by Mason (1994, 1996) show that arsenic phases are extremely rare, restricted to the Gwaith-yr-Afon Mine (Rust and Mason 1994) and that the ores are lead-copper or lead-zinc dominated with subordinate Fe, Ag, Sb, Co, Ni and Au (Rust and Mason 1994; Mason 1996).

The presence of galena-rich material in the mine spoil suggests a fairly sophisticated degree of beneficiation at the site. This careful separation of lead and copper minerals presents a potential problem in lead isotope provenance studies, for if lead isotope ratios taken from galena are taken to represent those belonging to nearby copper ores or smelted copper, care must be taken to ensure that both copper and lead mineralization belong to the same mineralization event.
MOUNT GABRIEL

Mount Gabriel is a Devonian sandstone mass rising to 408 m OD which represents the highest point of the 8 km ridge bridging the eastern end of the Mizen Peninsula in south-western Ireland (Fig. 1). The evidence for prehistoric copper mining at the site has recently been comprehensively published by O’Brien (1994). In contrast to the other sites discussed here, Mount Gabriel was not subject to large-scale exploitation in the historic period and there is no mention of early mining in records dating before the current century. Primitive workings at the site were recorded in the late 1920s (Duffy 1932), but systematic investigation did not begin until the 1960s when Jackson (1968) excavated a small trench obtaining a Bronze Age radiocarbon date for charcoal stratified within mine spoil (Table 1). A subsequent programme of field survey and excavation by O’Brien has confirmed many of the workings noted by Jackson and added a number of others as well as producing further radiocarbon dates from stratified mine spoil (Table 1). Twenty-five early workings have now been confirmed and a further seven are suspected. These are concentrated along the eastern slope of Mount Gabriel at altitudes ranging from 122–213 m OD. The workings occur singularly or as small groups intersecting the copper-bearing beds along their strike length. The primitive mine workings take the form of shallow, inclined openings ranging up to 12 m in depth. As with the other sites discussed here, there is no development within barren ground, with the form of the workings apparently entirely dictated by following surface exposures of secondary mineralization. Again the distinctive, rounded, morphology of the mines indicates firesetting.

A comprehensive suite of ore types, both from the spoil heaps and in situ locations within the mine area, have been collected and described (Ixer 1990, 1994; Ni Wen et al. 1996). Other, mainly vein-style ores from the region have been described by Reilly (1986), Ixer (1990), Ni Wen (1991) and Ni Wen et al. (1991, 1996). The mineralization at Mount Gabriel is very low grade, <0.5 wt.% copper (Snodin 1972; Jackson 1980), and comprises stratiform disseminated sulfides infilling pore spaces within sandstones/siltstones; or later sulfide concentrations lying along spaced cleavage/joint planes within thin (<5 cm), quartz-chlorite, veinlets cutting the host rocks. The host rocks are green-grey siliclastics and minor carbonate-bearing horizons (cornstones) of the Devonian Castlehaven Formation. The mineralization is an example of the red-bed class of deposit (Ixer 1990), but has undergone reconcentration by later metamorphism during the Hercynian Orogeny (Ni Wen et al. 1996) and, yet later, supergene alteration.

Both primary and secondary mineral assemblages are simple with only copper ± iron minerals being present. At the present time, within the Bronze Age mines area, visible signs of mineralization are restricted to malachite-limonite staining on joint planes and it is difficult to determine what was being exploited in the Bronze Age. Despite this, a full mineralogical description of the potential ore types has been possible. Much of the copper in the ores is very fine grained, <0.01 mm in diameter, and is present as disseminated intergrowths of chalcocite-bornite or as thin concentrations of the iron-copper sulfides, bornite, ‘idaite’ and the copper sulfides digenite, covelline and spionkopite lying along the spaced cleavage/jointing. Later supergene alteration has altered the sulfides to malachite and limonite with trace amounts of azurite. All other phases are present in trace amounts, but

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include a single grain of arsenopyrite (c.10 μm diameter) and very minor chalcopyrite. Tetrahedrite-tennantite is absent from the ore despite an earlier hand-specimen identification of fahlerz (Jackson 1968, 1980). The paucity of arsenic is confirmed by chemical analysis of the ores showing <25 ppm As (O’Brien 1994). Any copper metal produced from these ores essentially would be impurity-free (O’Brien et al. 1990), other than a little iron.

ROSS ISLAND

The Ross Island copper mine is situated on the eastern edge of Lough Leane, the largest of the Killarney Lakes, in Co. Kerry (Fig. 1). An intensive excavation and research programme is currently underway and full publication will follow, but some preliminary descriptions are available (O’Brien 1995; 1996, 33–6). The site was worked for copper in the early nineteenth century AD revealing older workings described as ‘Danes’s Mines’ in contemporary literature. Systematic excavation and radiocarbon dating have now shown that the earliest of these workings date to c.2400–2000 BC making them the earliest yet identified in the British Isles. Copper extraction appears to have started around the time of the late Neolithic copper-using horizon making the site crucial to our understanding of the inception of metallurgy in the region. The workings take the form of large cave-like openings in the limestone host rock with the smooth profiles commonly associated with firesetting and cobblestone tool use. The best investigated is a large opening in the western part of the site, but a large, and now partly flooded, trench at the eastern end of the site, the so-called ‘Blue Hole’, is thought to be of similar date. Large spoil deposits associated with the Bronze Age workings contain abundant charcoal as well as large numbers of stone hammers (many carefully waisted to take withy handles) and cattle scapula shovels.

In addition to the early dates, Ross Island is particularly significant for the discovery of a ‘work camp’ area located on a level escarpment immediately adjacent to the Bronze Age workings in the Western Mine area. The work camp has produced evidence for temporary habitation including shelters, animal bone food waste, pottery and a small amount of worked flint. There is also evidence for activities associated with the mine including ore crushing and possibly beneficiation as well as pit-features with burnt debris which may have a metallurgical function, although further investigation is required to confirm this. The work camp is securely dated by a series of radiocarbon measurements (Table 1) and this is strengthened by the recovery of numerous early Beaker pottery sherds. The Beaker period activity can be distinguished from later, Early Christian period, metallurgy for which evidence has recently come to light elsewhere on the site (O’Brien, pers. comm.).

Stratabound copper-lead-zinc ores with minor cobalt and silver sulfides and sulfarsenides occur for about 2.5 km from Ross Island to Muckross Mine, within a sequence of up to 20 m of heavily recrystallized, dolomitized and partly silicified bioclastic limestones of Courceyan age that are thrust over barren, silicified, argillaceous micrites. A wide range of primary copper-iron-lead-zinc sulfides and sulfarsenides, together with minor amounts of cobalt, silver and molybdenum minerals are present (Ixer and Pattrick 1995; Ixer et al. 1995). Supergene and oxidation zone mineralization are poorly developed so that the availability of secondary ores would have been very limited.

The mineralization at Ross Island

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comprises epigenetic copper sulfide-fahlerz veinlets cross-cutting recrystallized basal limestones of Carboniferous age plus banded polymetallic massive sulfides. It has not been determined whether these represent a single phase or multiple phases of mineralization. The deposit belongs to a small group of Cu-Ag ± Hg deposits found within the basal Carboniferous limestones in the north of the Munster-Shannon Basin (Andrew 1993; Ixer and Patrick 1995). However, the copper sulfide-fahlerz veins share many similarities with those present in the Devonian siliclastics in the south of the Munster Basin, most notably at Ballycummisk close to Mount Gabriel (Ixer 1990). The fine-grained banded sphalerite-galena ores are not characteristic of either the Cu-Ag ± Hg nor the copper-polymetallic veins of South Munster but are very like the bluestone ores of Parys Mountain.

At the Bronze Age Western Mine site chalcopyrite and silver-poor tennantite with minor pyrite, cobaltite, stromeyerite and molybdenite form disseminations and veins cutting the recrystallized limestone. More massive ores, comprising chalcopyrite, bornite, tennantite, spionkopite and molybdenite, present in the nearby nineteenth century AD spoil, may represent local, more extensive development of these veinlet-style ores at depth. A number of quite different, fine-grained, massive sulfide ores are present at the Blue Hole opencast, approximately 100 m east of West Mine. Here, chalcopyrite-pyrite with minor galena and sphalerite ores pass upwards into very finely banded sphalerite-galena with lesser amounts of pyrite and chalcopyrite. Minor to trace amounts of arsenopyrite and tennantite are present in all these ores, but are only of minor importance (Ixer and Patrick 1995). All of the ore types have been recovered from the excavated work camp area although the vast majority of fragments comprise chalcopyrite-tennantite veinlets in limestone. However, two caches of walnut-sized, massive chalcopyrite-pyrite, plus minor polymetallic sulfide ore have been found. These ores are not from the Western Mine and were probably derived from the Blue Hole opencast: ores that are arsenic-poor compared to those of the Western Mine. A single sample from the work camp is an open meshwork of charcoal-cubenite-löllingite. This is interpreted as a product of the incomplete roasting of charcoal mixed with crushed chalcopyrite-arsenopyrite ore (one of the minor ore types present at Ross Island).

For some of the mine sites discussed here it has been difficult to determine the exact nature of the ores as mined as grades are so low (Mount Gabriel) or the ores variable and the evidence of workings patchy (Parys Mountain). Neither problem is encountered at Ross Island where there is a superabundance of high grade ore and plentiful mining evidence. It seems clear that chalcopyrite-tennantite vein-style ores were being extracted from the Western Mine area which could have been processed to yield an arsenical copper product. However, the presence at the work camp of caches of the fine-grained chalcopyrite-pyrite ± galena, sphalerite, arsenopyrite/tennantite suggests a role for the arsenic-poor ores which may have been mined at the Blue Hole. Although it is necessary to be cautious regarding evidence from a single specimen, the part-roasted ore might suggest that a subtype of these ores, namely a fine-grained chalcopyrite-arsenopyrite was also being produced. Therefore it is possible, even likely, that the chalcopyrite-tennantite, or even the fine-grained polymetallic ores from Ross Island could have been smelted to produce an impurity-rich copper and, in particular, an arsenic-rich copper.
Conclusions

Archaeometallurgists will be familiar with data suggesting a significant minor element impurity component in much of the analysed Early Bronze Age metalwork from Britain (Northover 1980a, 1980b, 1982; Needham et al. 1989) and Ireland (Coghlan and Case 1957; Junghans et al. 1974). Even though it is a small proportion of the total metal recovered, Junghans et al. (1974), for instance, analysed more than one thousand Irish Early Bronze Age artefacts and found over 66% contained more than 0.1% As with 15% having more than 0.05% Ni. In this context, it is perhaps surprising that, of the mines investigated, only one or two could, realistically, have produced anything but essentially pure copper ore. Of the mines investigated here, it is highly likely that the Great Orme, Cwmystwyth, Alderley Edge and Mount Gabriel produced ores which did not contain significant levels of the impurity elements (As, Sb, Ni, Ag, Co) often considered diagnostic in interpretations of the metalwork analytical data. The nature of the field evidence at Parys Mountain makes it more difficult to predict the composition of the mined ore, but here too the balance of probability is that the ore is likely to have been free of significant impurities with the possible exception of iron and, maybe, zinc and lead.

Of the mines surveyed only Ross Island has produced unambiguous evidence for the exploitation of polymetallic ores. It is likely that the West Mine at Ross Island produced an arsenic-rich chalcopryite-tennantite ore and that this may have been processed together with a relatively arsenic-poor Cu, Fe ± Pb, Zn ore from the Blue Hole. The virtual absence of secondary/supergene mineralization at Ross Island makes it highly likely that the primary sulfide mineralization was the target of Bronze Age mining. If, as seems likely, such ores were mined in quantity at Ross Island, this would dispel previous doubts over the plausibility of a sulfide-based metallurgy at the beginning of the Early Bronze Age in the British Isles (Budd et al. 1992), although it poses interesting questions as to the technology employed. This observation does not, of course, rule out the possibility that secondary copper-arsenic ores may have been exploited elsewhere in the British Isles where they occur in considerably greater abundance. It is noteworthy that secondary copper-arsenic ores appear to have been used at about this time in south-east Spain for instance (Hook et al. 1991).

Although the use of arsenic-bearing ores at Ross Island might explain the occurrence of arsenic in Irish (and more widespread) metalwork of the period, in the majority of the artefacts analysed to date the element is accompanied by significant levels of antimony and silver (Coghlan and Case 1957; Northover 1980b). This has usually been explained in terms of the use of a tennantite-tetrahedrite (fahlerz) ore source. Interestingly, although the Ross Island, West Mine ores do feature minerals of this series, they occur predominantly as tennantite and are therefore antimony- and silver-poor. Indeed microprobe analyses show the fahlerz to be almost pure end-member tennantite (Pattrick, pers. comm.). If the currently available artefact analyses are representative, it would seem unlikely that Ross Island ores alone could have been responsible for the composition of much of the early metalwork given that no discrete antimony phases have been recognized from the site to date.

It is worth considering that Ross Island may not have been the only source of As ± Sb ± Ag-bearing copper in south-west Ireland. There are a number of locations in
the Mount Gabriel area of Co. Cork where Devonian sandstones are cut by major copper-bearing quartz-sulfide veins which carry these metals (Reilly 1986; Ni Wen et al. 1991). These include Dhurode, where chalcopyrite-arsenopyrite-tetrahedrite is present within mineralogically complex ores (Ni Wen et al. 1991) and, more importantly, Ballycummisk, a few kilometres south of Mount Gabriel, where coarse-grained chalcopyrite-tennantite-hematite ores are abundant. To date, there is no evidence of ancient mining at these sites, although this may have been obscured by mining in the last century (Ixer 1990; Ni Wen 1991; Ni Wen et al. 1996). These sites could be worthy of further investigation.

Another element which has been considered important in metal artefact analysis, nickel, does not appear to have been a significant constituent of the ores from any of the mines investigated. Northover (1980a) postulated that the appearance of significant levels of nickel in early tin-bronzes related to a shift in copper production from established Irish metal sources to deposits in the upland areas of Britain, especially Wales. Although the earliest dates for mines such as the Great Orme and Cwmystwyth are concordant with this interpretation and despite the presence of trace.microscopic amounts of nickeloan pyrite and possible ullmannite at Cwmystwyth, neither mine can realistically be considered to have been a source of the nickel. It may be that other Welsh sites will come to light with nickeliferous ores, or it may simple be that nickel was entering the metal supply from nickel-rich copper sources elsewhere which came into use at the same time.

Although this survey is probably far from comprehensive, in that those Bronze Age mines which we have investigated in detail so far are unlikely to be more than a small proportion of those that once existed, the results do provoke some reflection on our understanding of the organization of early metal production and use. Numerous studies have made use of artefact impurity patterns to define metalworking traditions with distinctive geographical and chronological distributions and links to particular technological entities. The underlying assumption was that metal production centred on particular ore sources during specific periods with little mixing of the finished product beyond a ‘hinterland’ enjoying the same source of supply. Under these circumstances it was reasonable to consider the impurity pattern of the metal as a fairly direct reflection of that ore, in which case the high proportion of analysed metalwork containing significant impurities implied extensive exploitation of copper-rich polymetallic ores. Distinctive changes in the impurity pattern of the metalwork over time were seen as a reflection of the decline of old ore sources, or the communities using them, and the emergence of new groups with control over different ore sources each with a characteristic impurity pattern.

The current study shows, however, that the majority of Bronze Age mine sites investigated to date, including some which appear to have been highly productive, were producing copper which must have been, essentially, impurity-free. Clearly, the possibility of additional copper production in other, less well investigated, metalliferous areas cannot be discounted, but if the pattern revealed by this study were typical it would be necessary to consider alternative hypotheses for the apparent predominance of impure copper-based metalwork.

Some impurity elements could have found their way into the metalwork as a result of deliberate, post-extractive, metal or mineral additions or as an unintentional by-product of
fluxes or refractories. Many of the artefacts in question are tin bronzes and are, almost certainly, deliberately alloyed; but it seems unlikely that the addition of tin or tin minerals would account for the suite of impurity elements (As, Sb, Ag, Ni, Pb) commonly found, or that arsenic would have been deliberately added at levels too low (less than 0.2%) to have been noticeable (Budd 1991). The most likely explanation is that the impurity elements are, ultimately, derived from copper ores, and that the widespread occurrence of impurities in Early and Middle Bronze Age copper-based metalwork reflects considerable mixing between copper derived from relatively pure ores like those of the Great Orme and Cwmystwyth and that from polymetallic deposits like Ross Island.

In a recent discussion of the Mediterranean Bronze Age, Sherratt (1994) emphasizes that the importance of metal as an exchange medium is closely linked to the possibilities it offers for accumulation and convertibility and therefore its flexibility for a wide range of social transactions. Given the implied importance of metalwork for personal wealth and prestige it seems highly unlikely that overall rates of deposition would have matched those of production. It seems probable that a significant proportion of metalwork stayed in circulation, to be re-melted, mixed and re-used, even in the earliest phase of the Bronze Age.

Although the possibility of widespread mixing and re-melting of British Early Bronze Age metalwork was discussed in previous analytical studies (e.g. Needham et al. 1989), it was felt that statistical procedures could nevertheless be used to outline broad changes over time and geography. This may indeed be a useful approach as it is reasonable to think in terms of the exhaustion or abandonment of old mines and the development of new sources resulting in gradual changes to the impurity pattern of the stock of metalwork in circulation and giving rise to geographically distinctive patterns which vary over time. It would be less likely that any of the compositional groups that may emerge would be closely related to an individual, specific ore source. The only exception might be metalwork associated with the very inception of metallurgy in a particular region.

The data presented here could be interpreted as support for the argument of Coghlan and Case (1957) for south-west Ireland as the earliest source of copper to be exploited in the British Isles, with its distinctive As-Sb-Ag impurity pattern gradually fading away as pure copper sources in England and Wales increased their contributions to the pool of metal in circulation. On the other hand, such speculation might be unwise given our incomplete picture of prehistoric mining in other areas of arsenic-rich copper mineralization, especially in south-west England. The current lack of evidence for a suitable source for the later Early Bronze Age nickel impurity urges caution in this respect.

Acknowledgements

The authors would like to thank all of those who allowed access to their sites, made samples and illustrations available and discussed their own data and results, often in advance of their own publications; Dr David Gale, Tony Hammond, Andy Lewis and their colleagues at Great Orme Mines, Simon Timberlake and colleagues in the Early Mines Research Group and Dr Billy O’Brien. The authors would also like to thank Drs Richard Pattrick and Quentin Fisher for their analytical support and advice. The work was supported by SERC/NERC grant GR/J26175 and by the Palaeologos Research Fund. One of the authors (PB) is currently supported by the NERC under the Advanced Fellowship scheme.

Oxford Journal of Archaeology
GLOSSARY OF SOME MINING AND GEOLOGICAL TERMS USED IN THE TEXT

Backfilling  The practice of infilling redundant workings with mine waste from later operations.

Barren Ground  Non ore-bearing rock in the area of mine workings.

Beneficiation  The practice of processing ore (usually by crushing and density separating in water or air) to improve the ore grade to the point at which it can be smelted.

Country Rock  Rock bodies intrusive mineral veins.

Deads  Waste material from mining, especially that left underground.

Dolomitization  Secondary alteration of limestone in which calcium carbonate is converted to calcium magnesium carbonate.

Epigenetic  Mineralization that post-dates its country rock.

Fahlerz Ore  Copper, iron, ± arsenic, ± antimony sulfide ores made up of minerals of the tennantite/tetrahedrite series.

Fault  A fracture in rock along which there has been an observable displacement.

Firesetting  The practice of lighting a fire adjacent to the rock surface to be worked. Thermal expansion leaves the rock more friable when the fire is extinguished allowing easier working, especially using stone tools.

Footwall  The wall of country rock under the lode left after mining.

Gallery  A horizontal or slightly inclined mine working.

Gangue  Waste material from ore beneficiation.

Joint  A fracture in rock between the sides of which there is no observable displacement.

Lode  A vein, especially one from which ore is being mined.

Open Cast  A surface mine working.

Openwork  A surface mine working.

Ore  Mineralization extracted for metal production.

Ore Body  Mineralization which is (or was) considered exploitable for metal production.

Ore Grade  The amount of metal of interest in the ore.

Outcrop  A surface exposure.

Shaft  A vertical working allowing access to and ventilation of mine workings.

Spoil  Mine waste, especially that deposited on the surface.

Strike  The direction along which a fault, vein or lode intersects the surface of the Earth.

Stope  A horizontal bed from which ore is extracted layer after layer either from above (underhand stoping) or below (overhand stoping).

Supergene  Processes involving water (often with dissolved minerals) percolating down from the Earth’s surface. In the context of ore bodies these processes usually result in the oxidation of primary minerals to their secondary products.

Tips  Surface dumps of mine waste.

Trenching  The practice of excavating narrow surface workings, especially to locate lodes for subsequent openwork.

Vein  A tabular or sheet-like body of minerals often a long joint or fissure in rocks.

Veinlet  A small vein.

Veinstuff  Material extracted from a vein.

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