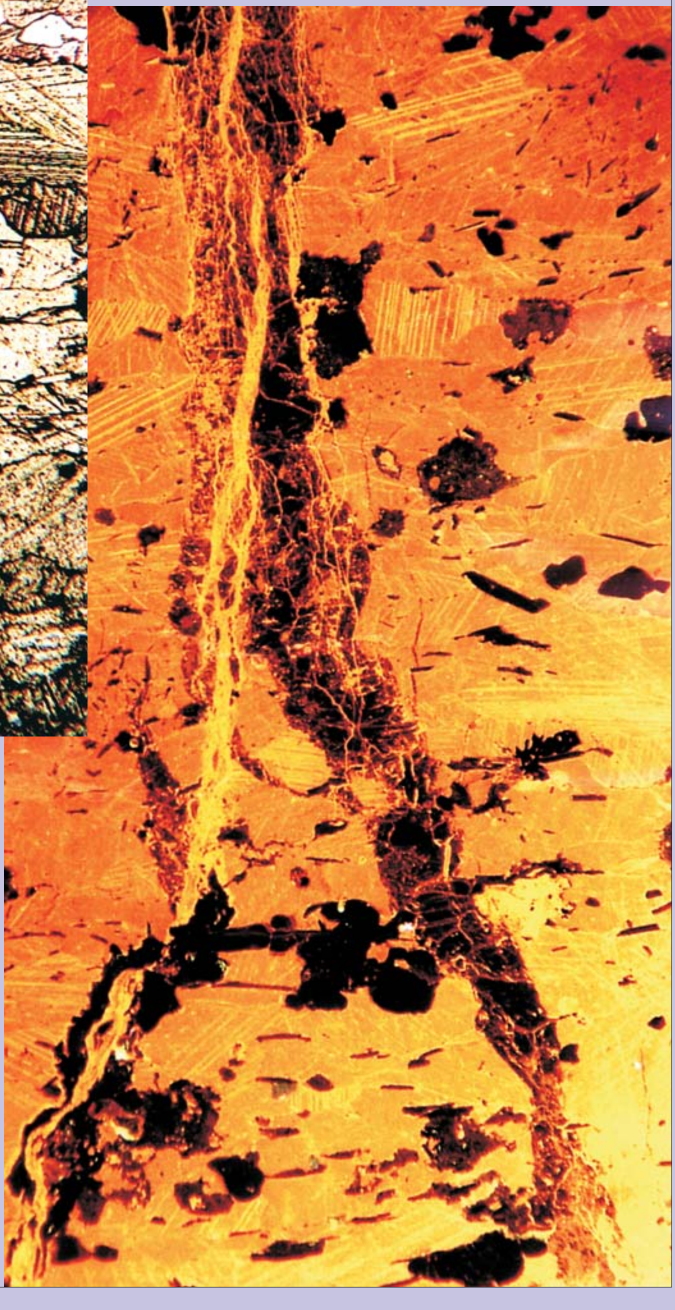
Petrography of metalimestones and metadolostones from the Dalradian of Northern Ireland



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Petrography of metalimestones and metadolostones from the Argyll and Southern Highland groups, Dalradian, Northern Ireland

Christopher W Thomas

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Contributors: M R Cooper and T P Johnston
(Geological Survey of Northern Ireland)

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Parent Body
Natural Environment Research Council
Polaris House, North Star Avenue,
Swindon, Wiltshire, SN2 I EU.
Telephone 01793 411500
Fax 01793 411501

Kingsley Dunham Centre
Keyworth, Nottingham, NG 12 5GG.
Telephone 0115 936 3100
Fax 0115 936 3200

Murchison House, West Mains Road,
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Telephone 0131 667 1000
Fax 0131 668 2683

London Information Office at the Natural History Museum, Earth Galleries, Exhibition Road,
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Telephone 0171 589 4090
Fax 0171 584 8270

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Telephone 01392 78312
Fax 01392 437505

Llandinam Building,University of Wales,
Aberystwyth, Ceredigion, Wales, SY23 3DB
Telephone 01970 622541
Fax 01970 622542

Geological Survey of Northern Ireland,
20 College Gardens, Belfast, BT9 6BS.
Telephone 028 90666595
Fax 028 90662835

Maclean Building, Crowmarsh Gifford,
Telephone 01491 38800
Fax 01491 25338

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Summary

Thirteen samples of metacarbonate rocks from the Argyll and Southern Highland Groups within the Dalradian of the Sperrin Mountains, central Northern Ireland, have been studied by optical petrography and cathodoluminescence (CL) imaging. Three of the metacarbonate samples are metadolostones and the remaining ten are calcitic metalimestones. Mineral assemblages are dominated by calcite or dolomite, but subordinate quartz, feldspar and white mica are ubiquitous. Graphite appears common in some samples as dusty inclusions. Grain boundaries are only moderately well equilibrated texturally and the CL imaging reveals considerable chemical heterogeneity on the grain-scale. Mineral assemblages and CL images indicate that infiltration of metamorphic fluid was very unlikely to have been significant in most samples and it is probable that many samples have retained pre-metamorphic (post-diagenetic) geochemical signatures, particularly of strontium and stable isotopes. Of the thirteen samples, six calcitic metalimestones have best potential for further work on their major oxide, trace element and isotope geochemistry. Geochemical data resulting from analysis of these samples could have significant potential for geochemical correlation of lithostratigraphical units and aiding interpretation of the geological structure. This, in turn, would aid mineral exploration and other geological programmes of work in Northern Ireland. In addition, the data elucidate further the significant changes which occurred in marine $^{87}Sr/^{86}Sr$ and $\delta^{13}C$ during the late Neoproterozoic.

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John Craven (Department of Geology and Geophysics, University of Edinburgh) is thanked for use of cathodoluminescence equipment

Terminology

For brevity, ‘metalimestones’ and ‘metadolostones’ are referred to simply as limestones and dolostones in the main body of this report.

1. INTRODUCTION

Metacarbonate rocks are distinctive components of the heterolithic metasedimentary and metavolcanic successions of the uppermost Argyll and lower Southern Highland Groups within the Dalradian of Northern Ireland. They are mostly calcitic limestones, but, as shown below, also includes some dolostones. Work on the geochemistry of carbonate rocks from the Dalradian of Scotland has shown that many lithostratigraphical units have distinctive geochemical signatures which have been of value in aiding and elucidating regional lithostratigraphical correlation (Hickman and Wright, 1983; Rock, 1985; Rock, 1986; Thomas, 1989; Thomas, 1993; Thomas, 1995; Thomas, 1999; Thomas and Aitchison, 1998; Thomas et al., 1997). Recent work by Thomas (1999) shows that the distinctive geochemistry of Dalradian metacarbonate rocks extends to the strontium and stable isotopes: in many cases, the signatures are primary or near-primary in origin and have not been disturbed significantly by metamorphism via metamorphic fluid infiltration and accompanying deformation.

This report discusses the petrology of thirteen samples of metacarbonate rocks from the Dungiven Formation (Tayvallich Subgroup, Argyll Group) and the Claudy Formation (Southern Highland Group), within the Dalradian of the Sperrin Mountains, Northern Ireland. Optical petrography and cathodoluminescence imaging have been used to establish the mineralogy, mineral textures, the degree of chemical heterogeneity, potential fluid infiltration pathways and the extent of metamorphic fluid-rock interaction. The observations have been used to identify a subset of samples which are most likely to have retained primary or near-primary strontium and stable isotope signatures and which could be used for further geochemical study.

2. GEOLOGY

The samples discussed in this report come from metacarbonate rock units within the Dungiven and Claudy Formations, at the top and base of the Argyll and Southern Highland Groups, respectively (Figure 1, Table 1). The limestones are exposed intermittently in Counties Londonderry and Tyrone (Figure 2; Appendix A). Outcrops are disposed around the hinge of the southeast-facing F_2 Sperrin Nappe on the northwest side of the Sperrin Mountains. The lithostratigraphy across this structure and the Sperrin Mountains is a subject of much debate (Alsop and Hutton, 1993; Alsop and Hutton, 1994; Arthurs, 1976; Arthurs, 1994; Smith and Johnston, 1994). Staff in the Geological Survey of Northern Ireland interpret the structure and lithostratigraphy to show that the core of the Sperrin Nappe is occupied by Argyll Group strata (the Newtonstewart Formation) fringed by the Dungiven Formation and the succeeding Claudy Formation. Alsop and Hutton (1993) take an alternative view, interpreting all the strata in the Sperrin Mountains as belonging to the Argyll Group and placing the core of the Sperrin Nappe in Glenelly, southeast of the main Sperrins ridge. I use the existing GSNI interpretation of the lithostratigraphy for the purposes of this report.

Metamorphic grade locally reached lower amphibolite facies, but is generally restricted to greenschist facies over most of the area from which the samples were collected. At present, there are no geothermobarometric estimates of metamorphic conditions. In discussion of the phase relations in the limestones, I consider temperatures in the range 300–550°C and pressures of 4 to 5 kbars.

3. METACARBONATE ROCK LITHOLOGIES

The metacarbonate rocks in the Dungiven and Claudy formations comprise limestones and dolostones, with the former predominating. Three of the thirteen samples provided for this study are dolostones (Table 1). Descriptions of the hand specimens provided by M R Cooper and T P Johnston show that the rocks range in colour from pale grey to nearly black. Most are medium- to coarse-grained, but sample **GSNILS4** from Rallagh Quarry Eeast is of a very coarse grained and black metalimestone, with calcite crystal plates up to 1cm across. This distinctive lithology appears to be a particular characteristic of Tayvallich Subgroup limestones, being observed in outcrops of ‘Tayvallich’ limestones in both the Southwest Scottish Highlands and Northern Ireland. The dolostones have much the same appearance in the field as the limestones, the main difference being the presence of brown surface weathering. **GSNILS1** is recorded as having a brownish organic coating, but this may be the result of weathering of iron-bearing dolomite.

The carbonate rocks are commonly interbedded with pelitic and psammitic lithologies; intrusive metabasic rocks, quartzites, greenbeds and pillow lavas also occur locally. More site-specific details are given in Appendix A.

4. PETROLOGY

4.1 Optical petrology

4.1.1 Limestones

Almost all the limestones have very similar petrographical characteristics. Calcite dominates the mineralogy and, although ubiquitous, variably abundant quartz, feldspar (chiefly plagioclase) and white mica are subordinate.

Calcite crystals are generally elongate and range in grain size from 0.1 to about 1 mm, with a texture similar to that shown in Figure 8a. Calcite is ubiquitously strain-twinned (generally type II twins of Burkhard, 1993). Grain boundaries are uneven and may be sutured locally and good triple-point contacts are rarely developed. The form of the grain boundaries indicates that the grain boundary architecture has not reached equilibrium during metamorphism and deformation. Dusty inclusions, probably mainly of graphite, are common, but inclusions are less common in some samples (e.g. **GSNILS 2**, **6**, **8**) than in others (e.g. **GSNILS 3**, **4**, **10**).

Calcite in samples from Rallagh Quarry (**GSNILS 4**,**5**) is different texturally to the other limestones. Calcite in **GSNILS 4** is equant and coarse-grained: large 10–12 mm calcite plates are dusted with aligned inclusions, presumed to be graphite. The large calcite plates are wrapped in a mesh of clear calcite crystals which effectively form a plate boundary network. The large calcite plates themselves are optically continuous, but internal grain boundaries are developed locally. The origin of the texture is unclear. The large plates appear to result from dynamic recrystallisation of the dark calcite matrix. In places, the outer parts of plates can be seen to be recrystallising to form finer grained calcite. Twinning in the large plates is truncated and locally bent in the vicinity of the clear calcite, indicating that the latter is later. The texture is a common feature of ‘Tayvallich’ equivalent limestones in the Southwest Highlands of Scotland and Northern Ireland, but has not been observed in other Dalradian limestones (Thomas, 1999).

In **GSNILS 5**, large calcite crystals are dusted with silicate inclusions, possibly of an iron-poor epidote such as clinozoisite (*electron microprobe analysis would be required to* *confirm their composition*). The calcite crystals commonly have rims of clearer calcite which only rarely contain inclusions.

Silicates are ubiquitous, though subordinate to calcite. Quartz and feldspar dominate and the latter is probably plagioclase, based on experience with other Dalradian limestones. Both have anhedral rounded to ragged form. The quartz and feldspar are difficult to tell apart in these rocks because of their fine grain size and shape and the general lack of twinning in the plagioclase. Cathodoluminescence serves generally to distinguish them (see below).

White mica forms laths and locally stubby crystals. The laths are generally aligned with the direction of elongation of the calcite crystals where the latter is more strongly developed.

The silicates tend to be isolated at grain boundaries, rather than as inclusions in plates (**GSNILS 5** excepted), although the silicates are more clustered in some samples (e.g. **GSNILS 8**).

4.1.2 Dolostones

Three samples, **GSNILS 1**, **7** and **10**, are dolostones. Although calcite and dolomite are generally difficult to distinguish petrographically, in these samples, the dolomite has a distinctive appearance. The grain shape is equant and ‘blocky’, being defined by rhombohedral crystal terminations. The rhombohedral cleavage is commonly prominent and the dolomite is dusted with inclusions. In addition, the dolomite lacks the twinning observed in the calcite, although it may display undulose extinction. Grain size is similar to that recorded in the limestones and, likewise, the grain boundaries are generally uneven or locally sutured.

Quartz and feldspar are locally abundant and concentrated in strings, as are phyllosilicates in **GSNILS 7**, which define a weak fabric. In **GSNILS 10**, the white mica occurs in clots which are locally deformed by a crenulation cleavage oblique to the earlier penetrative mica fabric.

4.2 Cathodoluminescence imaging

Cathodoluminescence (CL) imaging effectively provides a geochemical map of luminescent carbonates. In this study, it has been used to distinguish dolostones (non-luminescent) from limestones and has elucidated grain-scale chemical variation in calcite crystal interiors and at grain boundaries in the luminescent calcitic limestones. It has also been used to elucidate fluid infiltration pathways. The CL imaging aids assessment of the potential for samples for stable and Sr isotope work and the interpretation of such data.

Cathodoluminescence in calcite is activated by Mn and quenched by Fe. Thus calcites with high absolute Mn contents may not luminesce significantly if Fe is also appropriately enriched. However, more luminescent metamorphic calcites are generally interpreted as being relatively Mn-rich and this is generally borne out by electron or ion microprobe analysis (Graham et al., 1997; Lewis, 1999; Thomas, 1999). In recent studies of metamorphic carbonate rocks, (Cole, 1997; Graham et al., 1997; Lewis, 1999; Thomas, 1999), cathodoluminescence has proved invaluable in elucidating fluid infiltration pathways, these generally being domains of enhanced Mn and reduced Sr and $\delta^{18}O$ within carbonate matrices. This is because most metamorphic fluids are derived from silicate-rich sources characterised by low $\delta^{18}O$ and relatively high Mn.

The calcites in this study luminesce from dark orange brown to bright orange and bright yellow. The value of cathodoluminescence is revealed dramatically in the frontispiece, where a cryptic vein, undetectable in plane-polarised light, is revealed in the CL image by dark brown luminescence.

4.3 CL petrography of the dolostones and limestones

4.3.1 Dolostones

Dolomite is generally non-luminescent and this characteristic serves to distinguish dolomite from calcite. This phenomenon is best illustrated by sample **GSNILS 7**, in which small relict clots of pale orange-luminescent calcite occur within the non-luminescent dolomite matrix (Figure 3). This is the only dolostone of the three identified in the study (**GSNILS 1**, **7**, **10**) to show this phenomenon.

4.3.2 Limestones

The CL images show that most of the calcite crystals in the limestones are not chemically well-equilibrated. A particularly common feature is that the interiors of many calcite crystals are more darkly luminescent, whilst the outer parts are more luminescent. The darker luminescent domains commonly have sharp boundaries where they abut paler luminescent domains, but gradual variation from dark to more pale luminescent calcite is also observed. These features are well illustrated by the CL image of **GSNILS 12** (Figure 4). Although the darker domains are mainly wholly within a calcite crystal, the limits to these domains and the paler surrounding domains are not concentric with the current metamorphic grain boundary architecture. Thus the zoning predates the current grain boundaries and is likely to be mainly diagenetic in origin.

In one or two samples (e.g. **GSNILS 12**, Figure 4) the grain boundaries are picked out by bright orange/yellow luminescent calcite. This is a common feature of metamorphosed calcitic limestones and is interpreted as elucidating fluid infiltration pathways (e.g. Graham et al., 1998; Lewis et al., 1998). Most samples in this study do not have prominently luminescent grain-boundary calcite, suggesting that infiltration of metamorphic fluid was limited. An exception to this is **GSNILS 3**. The CL image of this limestone reveals irregularly shaped domains of darkly luminescent calcite isolated within wider domains of more luminescent calcite and that the calcite gets more luminescent towards the grain boundaries (Figure 5). This sample bears all the hallmarks of a limestone infiltrated extensively by metamorphic fluid, with extensive alteration of the chemistry of calcite crystals.

The CL image of **GSNILS 4** (Rallagh Quarry East) shows that dark, inclusion-rich calcite plates have very even orange-brown luminescence (Figure 6). Clearer calcite at the margins of the plates is much more luminescent. Similarly luminescent calcite is also present very locally within the calcite plates. The CL image of **GSNILS 5** is somewhat similar (Figure 7). The inclusion-rich, darker orange luminescent calcite plates are surrounded by more luminescent clear calcite in which grain boundaries are picked out by more yellow-luminescent calcite. This texture may be a thermally-enhanced version of the texture seen in **GSNILS 4**, given the presence of metadolerite sills in the vicinity, but it is difficult to be certain of the origin of the chemical differences between the two different luminescent calcites without further work.

Special mention must be made of the CL image of **GSNILS 6**. The CL reveals a darkly luminescent calcite vein not visible in the plane-polarised light image (Figure 8a, b) (*note, however, that it is just visible in thin section*). The dark vein is overgrown by the metamorphic grain boundaries and thus predates the fabric: the vein calcite has been 'absorbed' texturally into the metamorphic calcite fabric. The pathway of the dark vein has also been reused by a vein network of bright yellow luminescent calcite. Although this latter vein is more visible as a zone of alteration in the ppl image, it is also clear that metamorphic grain boundaries are crossing this vein, at least in part. Thus this vein also predates the metamorphic grain-boundary fabric. The important point to be made here is that although the textural identity of the veins is being destroyed by metamorphic recrystallisation of the calcite, the chemical identity is not and the chemical discontinuities across the veins are as sharp as when the veins were formed. The corollary of this is that although the crystal shapes and boundaries may have changed, there has been no homogenisation of at least the cationic components of the chemistry. This is likely to extend to isotopic components of $\delta^{18}O$, $\delta^{13}C$ and $^{87}Sr/^{86}Sr$. The implication is that the reorganisation of crystal shapes and boundaries has occurred without the presence of an infiltrating fluid phase and was probably anhydrous and that the limestone has retained its pre-metamorphic chemistry.

5. PHASE RELATIONS AND CONDITIONS OF METAMORPHISM

5.1 Introduction

There are no phases in these metacarbonate rocks which are suitable for geothermobarometric work. Although dolomite and calcite occur together in **GSNILS 7**, the fact that the latter is relict and being replaced indicates that the calcite and dolomite are not in thermodynamic equilibrium. However, it is possible to use the limited number of phases present in these rocks to place broad limits on temperatures of metamorphism and to make general statements about the likely fluid-rock interaction history during metamorphism.

5.2 Thermodynamic composition space and limiting phases

The mineral assemblage in the dolostones is dolomite + quartz + plagioclase + white mica, and, in the limestones, calcite + quartz + plagioclase + white mica. These phases, when modelled as end-members (*i.e. no solid solution, for example, celadonite – muscovite solid solution in white mica*) in a system free of iron and sodium, lie in the composition space K_2O – CaO – MgO – Al_2O_3 – SiO_2 – H_2O – CO_2 (KCMASHC). The assemblages can then be used with this composition space to generate a net of mineral - fluid reactions in temperature and fluid composition (T-X_{CO_2}) space which places temperature and fluid compositional constraints on assemblage stabilities at a range of pressures.

Although phases such as zoisite, tremolite, phlogopite, K-Feldspar and diopside are absent from the assemblages, they are included in the analysis because they place limits on the stability limits of the observed assemblages. Thus the phases included in the analysis are: dolomite (dol), calcite (cc), quartz (qz), plagioclase (pl), white mica, treated as muscovite (mu), clinochlore (clin), tremolite, (tr), diopside (di), zoisite (zo), phlogopite, (phl), K-feldspar (ksp) and a binary fluid phase H_2O – CO_2.

5.3 Pressure and temperature boundary conditions

Phase relations amongst the mineral assemblages in T - X_{CO_2} space have been calculated using the Perple-X thermodynamic software suite of Connolly (1990). The complimentary program VertexView (Castelli et al., 1997) has been used to generate the T - X_{CO_2} sections from the results calculated by Perple-X. I have generated two T - X_{CO_2} sections at pressures of 4 and 5 kbars respectively for temperatures in the range 300° to 550°C, covering typical lower greenschist to lower amphibolite facies conditions. Because all the limestones and dolostones contain at least some quartz, quartz is treated as being in excess and SiO_2 as a saturated component. The sections are shown in Figure 9.

5.4 The importance of plagioclase composition

Although most phases can justifiably be dealt with as end-members with unit activities, it is very unlikely that the plagioclase is pure anorthite (An, $CaAl_2Si_2O_8$). The topology of phase equilibria in T - X_{CO_2} sections for assemblages including a plagioclase feldspar is very sensitive to the plagioclase composition: more calcic plagioclase is stable only in more CO_2 – rich fluids. Thus an estimate of plagioclase composition is important in any attempt to estimate the limiting fluid compositions in equilibrium with the observed assemblages. Work on limestones from the Dalradian of Northeast Scotland shows that plagioclases generally have andesine compositions of about An_{40}. In lieu of any compositional data on the plagioclase, I have used An_{40} as an estimate of plagioclase composition in the calculations. Electron probe analysis would be required to verify the plagioclase composition.

5.5 Results and interpretation

The T - X_{CO_2} sections are shown in Figure 9a, b. The net of reactions has been simplified to show only the key reactions. Reactions involving dolomite as a reactant are shown in red, whilst those involving calcite as a reactant are shown in blue.

5.5.1 Dolostones

For the dolostones the key limiting equilibria are those involving mu and dol and pl. These equilibria extend through a series of invariant points, limiting the mu + dol + qz assemblage to regions of relatively higher X_{CO_2} and lower temperature. The maximum temperature stability of this assemblage is about 500°C at 4 kbars, and about 525°C at 5 kbars. However, these peak thermal stabilities only occur at high X_{CO_2}. If the equilibrium fluid phase is significantly less or more than about 0.6 to 0.8, the temperatures at which this assemblage is stable are much lower, particularly at rather low X_{CO_2} (< ~ 0.3). The assemblage dol + pl + qz constrains X_{CO_2} to be higher than for the assemblage dol + mu + qz below the invariant point through which these two reactions pass. The dol + mu + pl + qz assemblage is consistent with the dolostones having not been infiltrated by hydrous metamorphic fluids as one would expect to see reaction products if infiltration had occurred. This is also true with regard to temperatures of metamorphism, which must be less than thermal stability limits for the assemblages, as discussed above.

5.5.2 Limestones

For the limestones, maximum thermal stability limits for the assemblage cc + mu + qz range from about 485°C at 4 kbars to about 510°C at 5 kbars. At high and low X_{CO_2}, the thermal stability of this assemblage declines and the stability is increasingly limited by X_{CO_2}. The zoisite breakdown reaction zo = cc + pl is very strongly a function of X_{CO_2} under greenschist facies conditions. The reaction constrains the stability of cc + pl to significantly higher X_{CO_2}

at low temperatures under a pressure of 5 kbars. However, at typical greenschist facies temperatures of around 400°C, pl + cc is stable over a very wide range of X_{CO_2}. This wide-ranging stability is limited value in constraining equilibrium fluid compositions. Nevertheless, the absence of zoisite is consistent with the lack of extensive infiltration of typically very hydrous metamorphic fluids.

5.6 Summary

The thermal stability of mineral assemblages in the dolostones and limestones is limited to temperatures of less than about 525°C at 5 kbars and 500°C at 4 kbars. If equilibrium fluid compositions were H_2O or CO_2 – rich, then the assemblage stabilities are strongly a function of the fluid phase composition. The assemblages in both the dolostones and limestones are consistent with the rocks having not been infiltrated extensively by hydrous metamorphic fluids. The phase equilibria data are consistent with the CL evidence for the lack of fluid infiltration, although **GSNILS 3** may be an exception.

6. STRONTIUM AND STABLE ISOTOPE GEOCHEMISTRY OF UPPERMOST ARGYLL GROUP AND SOUTHERN HIGHLAND GROUP LIMESTONES

A particular aim of the work presented in this report was to establish the potential for elucidating primary or near-primary $^{87}Sr/^{86}Sr$ and stable isotope signatures in the carbonate in the Dungiven and Claudy Formation carbonate rocks. Work to date by Thomas (1999) on a small number of samples from Tayvallich-equivalent limestones from Scotland and Northern Ireland, including limestones from the Dungiven Formation and the Torr Head Limestone Formation, suggests that pre-metamorphic $^{87}Sr/^{86}Sr$ and $\delta^{13}C$ are preserved, at least in some samples. Samples from Banagher Glen, Butterlope Glen, Torr Head, Tayvallich and Boyne Bay (Northeast Scotland) have $^{87}Sr/^{86}Sr$ values in the range ~0.7082 to ~0.7096 (Table 2). Undoubtedly, some of these values are altered in part, but they are typical of limestones of ~600 Ma in age in other non-metamorphosed, carbonate rock bearing successions from around the world (*note that the age of the Tayvallich Limestone itself is constrained by the Tayvallich keratophyre age of 595 ±4 Ma; Halliday, et al., 1989)*.

Values of about 0.7082 – 0.7088 are probably near primary, on the basis of certain geochemical criteria, including Sr and Mn contents and $\delta^{13}C$ and $\delta^{18}O$ values. These $^{87}Sr/^{86}Sr$ values are much higher than those for Appin and Grampian Group limestones and the higher values reflect a significant shift in marine $^{87}Sr/^{86}Sr$ in the latter part of the Neoproterozoic, after about 650 Ma. The $^{87}Sr/^{86}Sr$ are *consistent* with the correlation of the Dungiven and Torr Head limestones with the Tayvallich limestones of Scotland, at least at Subgroup level, although the resolution of the data for variation in late Neoproterozoic marine $^{87}Sr/^{86}Sr$ is too coarse to be confirmatory. I would anticipate that suitable limestone samples from the suite discussed in this report would also yield primary to near-primary $^{87}Sr/^{86}Sr$ data and that these would also have relatively high values of about 0.7088 – 0.7090.

The presence of limestones in the Southern Highland Group Claudy Formation in Northern Ireland contrasts with their general absence in the Southern Highland Group of Scotland, the Leny Limestone and some thin Southern Highland Group limestones in Tayvallich excepted. The Claudy Formation limestones offer the opportunity to extend the Dalradian limestone $^{87}Sr/^{86}Sr$ and stable isotope database into the lower part of the Southern Highland Group.

The work presented in this report represents the first stage in screening of samples for possible future isotope work. Ideally, petrographic screening needs to be accompanied by geochemical analysis to obtain the concentrations of Sr, Rb and Mn. Bulk-carbonate $\delta^{18}O$ and $\delta^{13}C$ data are also required to permit informed interpretation of bulk carbonate $^{87}Sr/^{86}Sr$ data.

From the thirteen samples examined in this work, six appear to have the best potential for isotope work. The dolostones need not be considered further because work to date on other Dalradian dolostones shows that their $^{87}Sr/^{86}Sr$ isotope compositions are strongly disturbed by dolomitisation, most likely arising from the fact that dolomitising fluids are largely non-marine and have radiogenic $^{87}Sr/^{86}Sr$ signatures typical of 'continental' fluids. Of the limestones, four suitable samples come from the Claudy Formation (**GSNILS 2, 6, 8, 13**) and two from the Dungiven Formation (**GSNILS 4, 9**). The latter samples would add to the isotope data which already exist for the Dungiven and equivalent Torr Head limestones (Table 2).

7. SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

- 13 samples from the Claudy and Dungiven Formations of the Dalradian of central Northern Ireland have been studied petrographically. Emphasis has been placed on establishing their potential for strontium and stable isotope work.

- The work shows that three samples are dolostones and ten are calcitic limestones.

- Texturally, metamorphic grain boundary architectures are only moderately well-equilibrated texturally and CL work shows that the limestones are chemically heterogeneous on the grain scale.

- CL imaging and consideration of the stability limits of the mineral assemblages indicates that infiltration of a metamorphic fluid phase was insignificant in most samples. It is likely that most samples have retained their pre-metamorphic strontium and stable isotope geochemistry.

- Based on the petrography and CL imaging, six limestone samples are considered suitable for strontium and stable isotope work and, with careful subsampling and analysis, would be anticipated to yield primary or near-primary isotope signatures.

- Determination of such signatures would extend the current strontium and stable isotope database for Dalradian limestones into the lower parts of the Southern Highland Group and provide additional constraints on the correlation of the Dalradian between Scotland and Northern Ireland.

- The data would serve to elucidate further the variation in marine $^{87}Sr/^{86}Sr$ and $\delta^{13}C$ during the late Neoproterozoic and, depending on the geochemical characteristics of the samples, may allow distinction between limestones of different formations.

- The geochemical data may provide extra constraints on correlation of lithostratigraphical units through poorly exposed ground in the Sperrin Mountains and with the rest of the Dalradian through the wider part of Northern Ireland.

- Improved lithostratigraphical correlation will contribute to the testing of the current conflicting structural models for the Sperrin Mountains.

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APPENDIX A
Sample location and outcrop details from M R Cooper and T P Johnston (GSNI).

GSNILS1 Glenconway, stream section, [26353 41479]
Site located ~7.5km NW of Dungiven. Grey, medium to coarsely crystalline metalimestone (*sample is actually dolostone*) in beds up to 30cm thick (sampled). Beds are often separated by associated schistose (sheared) metalimestone. There is evidence for faulting in close vicinity to the sample site. Mineralised veins containing quartz, calcite and barytes cut the rock mass. Metalimestones are attributed to the very top of the Claudy Formation (Southern Highland Group).

GSNILS2 Ballyholly Quarry, [25742 41082]
Site located ~5km WNW of Foreglen. Grey, medium to coarsely crystalline metalimestone in beds up to 30cm thick (sampled). Beds are often separated by associated schistose (sheared) metalimestone. Mineralised veins (sweats) containing quartz and calcite with minor galena and pyrite cut the rock mass. Metalimestones are attributed to the top of the Claudy Formation (Southern Highland Group).

GSNILS3 Tirglassan Quarry, [25975 40708]
Site located ~3km SW of Foreglen. Grey, medium to coarsely crystalline metalimestone in beds up to 30cm thick (sampled). Beds are often separated by associated schistose (sheared) metalimestone. Mineralised veins (sweats) containing quartz and calcite with minor pyrite cut the rock mass. Metalimestones are attributed to the middle of the Claudy Formation (Southern Highland Group).

GSNILS4 Rallagh Quarry East, [26588 40567] and
GSNILS5 Rallagh Quarry West [26574 40570]
Sites located ~4km SW of Dungiven. Dark grey to black, medium to very coarsely crystalline metalimestone in beds up to 1m thick (sampled). Siliciclastic laminations are present cutting the rock mass at cm to dm intervals. Mineralised veins (sweats) containing quartz and calcite cut the rock mass. Patchy recrystallisation fabric noted. Late stage haematite bearing veins also present. Metabasite sills are present in close proximity to sample site. Metalimestones are attributed to the top of the Dungiven Formation (Tayvallich Subgroup, Argyll Group).

GSNILS6 Ashlamaduff stream section, [27920 41169]
Site located ~10km ENE of Dungiven. Grey, medium to coarsely crystalline metalimestone, with significant siliciclastic content, in beds up to 30cm thick (sampled). Beds are often separated by associated calcareous schist (sheared). Mineralised veins containing calcite with significant pyrite cut the rock mass. Patchy recrystallisation fabric noted. Metalimestones are attributed to the middle of the Claudy Formation (Southern Highland Group).

GSNILS7 Butterlope Glen, [2491 3952]
Located ~4km N of Plumbridge, Co Tyrone. Sample from small partially filled quarry pit. Limestones (*sample is actually dolostone*), medium to coarse grained, massively bedded and locally schistose. Weathered surfaces indicate localised graphite. Part of the Dungiven Formation (Tayvallich Subgroup, Argyll Group).
Associated lithologies include quartzite, semipelite, pelite, pillow lava and greenbeds.

GSNILS8 Longlands Road Quarry, [2522 4040]

Active limestone quarry on Longlands Road, ~4km SW of Claudy, Co Londonderry.
Medium grained limestone, massive units up to 1m with thin cm scale pelite interbeds. Outcrops throughout quarry contain disseminated sulphides (pyrite). Part of the Alla Limestone Member, Claudy Formation (Southern Highland Group).
Associated lithologies include quartzite, greenbeds, psammite and semipelite.

GSNILS9 Balix Hill [2480 3958]

Abandoned lime pit on slopes of Balix Hill. Located on the Crockrow Road, ~4.5km N of Plumbridge, Co Tyrone. Thinly bedded (2-10cm) deformed limestone beds, grey and bluish grey to black weathering limestones interbedded with thinner grey and silvery grey phyllite. Part of the Dungiven Formation Limestone (Tayvallich Subgroup, Argyll Group)
Associated lithologies in the surrounding area include quartzites, pillow lava and greenbeds.

GSNILS10 Drain Quarry [24580 40185]

Disused and partially flooded limestone quarry located ~2km SE of Donemana, Co Tyrone. Light grey to greyish blue locally “sugary” medium to coarse-grained limestone (*sample is actually dolostone*) units up to 1m thick with thin <1cm chloritic and locally slightly graphitic phyllite and pelite interbeds. Tight folding in places associated with the regionally dominant cleavage (F_2, S_2) Part of the Alla Limestone Member, an important marker horizon in the Claudy Formation (Southern Highland Group) north of the axis of the Sperrin overfold.

GSNILS11 Feeny Quarry [26340 40490]

Disused limestone quarry on left bank of Altcattan Burn, ~1km SE of Feeny, Co Londonderry. Thin bedded greyish blue crystalline limestone. Part of the Dungiven Formation Limestone (Tayvallich Subgroup, Argyll Group).
Associated lithologies include amphibolite metabasite, quartzite, and quartz biotite psammite, semipelite pelite.

GSNILS12 Kennedy Quarry, Bonds Glen [24985 40700]

Working limestone quarry on Bonds Glen Road, 4.7km W of Claudy, Co Londonderry. Massive beds (up to 1m thick) grey micaceous limestones interbedded with grey and minor black chloritic phyllite. Part of the Bonds Glen Limestone Member, an important marker horizon in the Claudy Formation (Southern Highland Group) north of the axis of the Sperrin overfold.
Associated lithologies include grits, quartz biotite psammite, minor greenbeds and grey chloritic phyllite.

GSNILS13 Lough Ash Road [24945 40125]

Minor roadside outcrop recently exposed on Lough Ash Road, 5.8km ESE of Donemana, Co Tyrone. Pale grey to white sugary limestone interbedded with quartz psammite and grey green chloritic phyllite. Part of an unnamed limestone member within the Claudy Formation (Southern Highland Group) north of the axis of the Sperrin overfold.
Associated lithologies in the formation above and below include grits, quartz biotite psammite, grey chloritic phyllite and minor greenbeds and metabasite.

APPENDIX B

Sample lithology details provided by M R Cooper and T P Johnston (GSNI).

GSNILS1

Limestone (*actually dolostone*), medium grained, medium grey, crystalline. Cut by fine veinlets of cc. Covered by brown organic material, but fresh internally.
Several pieces
Thin sections from largest piece, including a veinlet

GSNILS2

Limestone, pale medium grey, medium/coarse grained, cystalline. Contains more micaceous partings locally and is weakly foliated and faintly banded in appearance.
Fresh and clean.
One large piece
Thin sections cut to include banded material

GSNILS3

Limestone, medium grey, medium/coarse grained, crystalline and weakly foliated. With thin micaceous films/partings. Sample shape suggests thinly bedded in outcrop.
Generally fresh, though some micaceous films a little weathered.
Several pieces
Thin sections cut from one piece

GSNILS4

Limestone, black, coarse to very coarse grained, crystalline, not obviously foliated, though some larger crystals may be aligned weakly; typical ‘Tayvallich’ lithology.
Fresh and clean
Several pieces
Thin sections

GSNILS5

Limestone, dark grey, fine to coarse grained, crystalline. Developing coarser black crystals locally across laminae, though not as coarse grained as GSNILS4. Weakly foliated with aligned streaky white calcite strings in places.
Reasonably fresh.
Several pieces.
Thin sections from largest piece.

GSNILS6

Limestone, medium grey, medium to coarse grained, crystalline and foliated, with strings and blebs of coarse white calcite in the grey matrix; white calcite is less obviously foliated in the larger blebs. Micaceous partings on sample surface, but not obvious internally.
Fresh and clean.
One large piece.
Thin sections cut to include strings.

GSNILS7

Limestone (*actually dolostone*), medium to dark medium grey, crystalline, fine to medium grained, faintly banded appearance in part, not obviously foliated.
Fresh, clean.
Several pieces.
Sections to cut to include banded material.

GSNILS8

Limestone, medium/pale medium grey, medium to coarse grained, crystalline. Locally spotted with coarse black cc crystals (?same as the large black crystals in other samples). Faintly banded, very weakly folded.
Fresh and clean.
One piece.
Sections cut to include banding.

GSNILS9

Limestone, pale medium grey, crystalline, with uniform to very weakly banded appearance. Not obviously foliated.
Fresh and clean.
Three good pieces.
Thins sections cut from largest piece.

GSNILS10

Limestone (*actually dolostone*), medium grey, fine to coarse grained, crystalline, with more micaceous limestone locally defining minor fold hinge and poor spaced cleavage.
Reasonably fresh.
One large piece.
Thin sections cut to include micaceous parts.

GSNILS11

Limestone, dark grey, fine to medium grained, crystalline and foliated. Foliation picked out by micas – looks like a stretching lineation in HS on fresh surfaces.
Fresh and clean
Two pieces.
Thins sections cut from larger, flatter piece.

GSNILS12

Limestone, pale medium grey, uniform, medium grained, crystalline.
Fresh and clean.
One large piece.

GSNILS13

Limestone, pale medium grey, coarse, crystalline, with blebs/laminae of white coarse calcite.
Fresh and clean.
One piece.
Thin sections cut to include white calcite.

Sample	Formation	Location	Grid Reference (Irish Grid)	Lithology	Lithostratigraphical position or member
GSNILS 8	Claudy Formation	*Longlands Road Quarry*	2522 4040	metalimestone	Alla Limestone Member
GSNILS 10	Claudy Formation	*Drain Quarry*	24580 40185	metadolostone	Alla Limestone Member
GSNILS 12	Claudy Formation	*Kennedy Quarry, Bonds Glen*	24985 40700	metalimestone	Glen Bonds Limestone Member
GSNILS 13	Claudy Formation	*Lough Ash Road*	24945 40125	metalimestone	not given
GSNILS 1	Claudy Formation	*Glenconway, stream section*	26353 41479	metadolostone	top
GSNILS 2	Claudy Formation	*Ballyholly Quarry*	25742 41082	metalimestone	top
GSNILS 3	Claudy Formation	*Tirglassan Quarry*	25975 40708	metalimestone	middle
GSNILS 6	Claudy Formation	*Ashlamaduff stream section*	27920 41169	metalimestone	middle
GSNILS 4	Dungiven Formation	*Rallagh Quarry East*	26588 40567	metalimestone	top
GSNILS 5	Dungiven Formation	*Rallagh Quarry West*	26574 40570	metalimestone	top
GSNILS 7	Dungiven Formation	*Butterlope Glen*	2491 3952	metadolostone	not given
GSNILS 9	Dungiven Formation	*Balix Hill*	2480 3958	metalimestone	not given
GSNILS 11	Dungiven Formation	*Feeny Quarry*	26340 40490	metalimestone	not given

Table 1. Location, lithological and lithostratigraphical details of the samples of limestone and dolostone discussed in the report

Location	Sample	$^{87}Sr/^{86}Sr$	error	Sr	Mg/Ca	1/Sr	Mn*	Mn/Sr	Rb	Rb/Sr	$\delta^{18}O_{SMOW}$	$\delta^{13}C_{PDB}$
Southern Highland Group												
Leny Limestone												
Callander, Perthshire	HY1362	0.711859	0.000030	796		0.00126	n.a.		0.03	0.00004	15.7	-4.6
	HY1364	0.712781	0.000017	815		0.00123	n.a.		0.04	0.00005	18.3	-5.0
	Median :	**0.712320**		**806**					**0.04**		**17.0**	**-4.8**
Argyll Group												
Boyne Limestone												
Boyne Bay, NE Scotland	HY147	0.709018	0.000016	4607	0.0296	0.00022	310	0.07	13	0.0028	19.0	1.9
	HY148	0.709451	0.000016	2695	0.0620	0.00037	697	0.26	43	0.0160	19.8	0.5
	HY149	0.709305	0.000014	3534	0.0334	0.00028	697	0.20	23	0.0065	18.2	-1.9
	HY150	0.709297	0.000016	3225	0.0635	0.00031	542	0.17	52	0.0161	18.2	1.2
	Median :	**0.709301**		**3380**			**620**		**33**		**18.6**	**0.9**
Tayvallich Limestone												
Port an Sgadain, Tayvallich	T1	0.708851	0.000020								20.7	-2.3
	T2	0.709203	0.000020								15.4	-0.7
	T3	0.708960	0.000017								16.6	1.1
	T4	0.709096	0.000018					No elemental data available			16.3	1.1
	T5	0.709467	0.000020								17.5	1.5
	T6a	0.709637	0.000017								17.7	1.6
	T6b	0.708969	0.000016								17.3	2.0
	Median :	**0.709096**										
Torr Head Limestone												
Torr Head, Co. Antrim	HY1348	0.708857	0.000039	1865	0.0109	0.00054	144	0.08	26	0.0139	14.8	7.4
	HY1349	0.708810	0.000032	2115	0.0080	0.00047	144	0.07	20	0.0095	17.9	7.5
	Median :	**0.708834**		**1990**			**144**		**23**		**16.4**	**7.4**
Dungiven Limestone												
Banagher Glen	HY1350	0.708220	0.000033	265	0.0838	0.00377	3890	14.68	3	0.0113	13.4	7.0
Butterlope Glen	HY1351	0.708745	0.000043	2299	0.0070	0.00043	216	0.09	12	0.0052	23.4	7.3
	Median :	**0.708483**		**1282**			**2053**		**8**		**18.4**	**7.2**

Data from Thomas (1999)
* data converted from wt% oxide to ppm element

Table 2. $^{87}Sr/^{86}Sr$, $\delta^{18}O$ and $\delta^{13}C$ & Sr, Mn and Rb data for 'Tayvallich-equivalent' and Southern Highland Group limestones

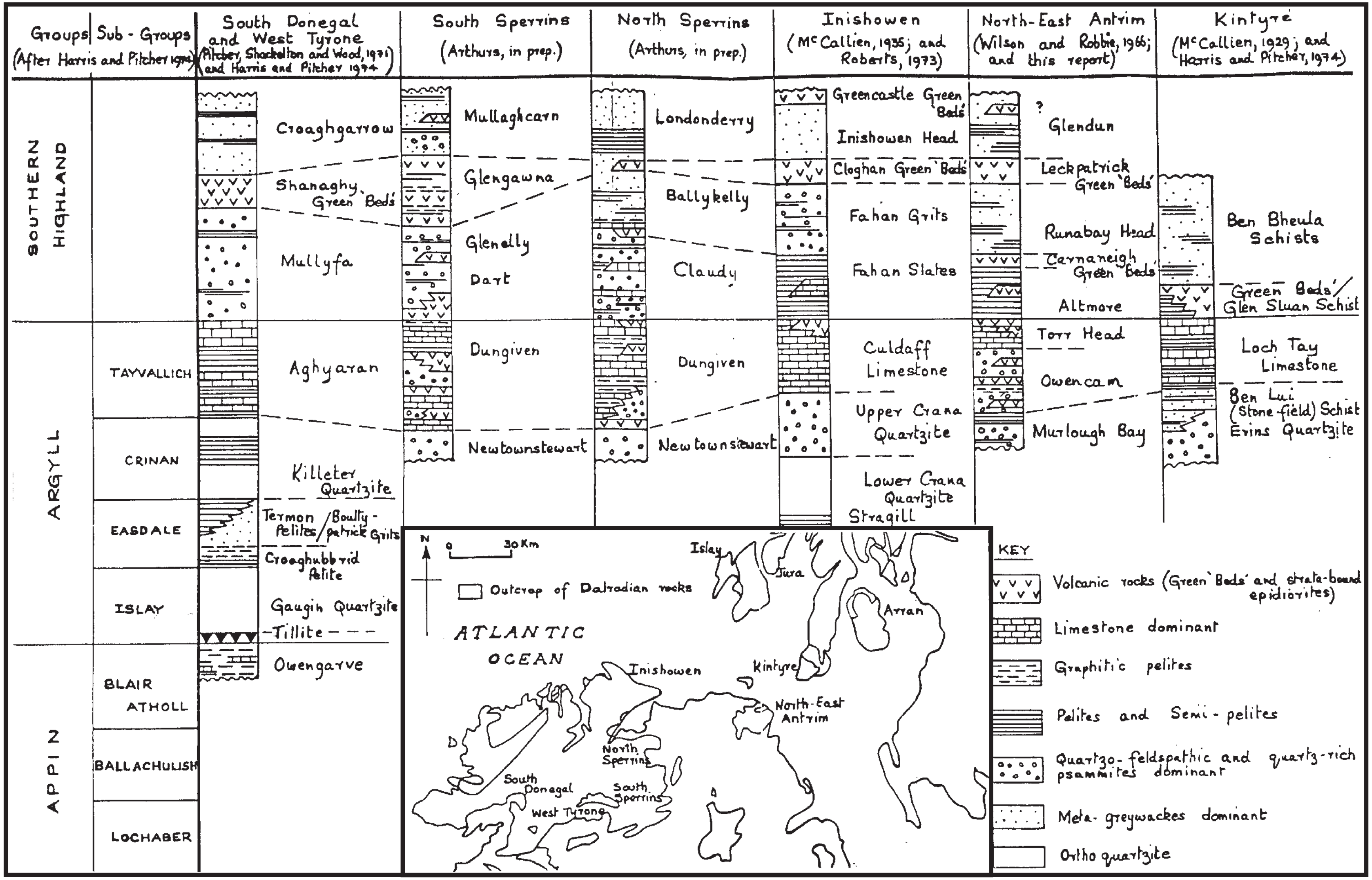


Figure 1. The lithostratigraphy of Dalradian rocks in Northern Ireland and Southwest Scotland, after a compilation by Arthurs (1976). The metacarbonate rock samples come from the Claudy and Dungiven Formation, North Sperrin Mountains.

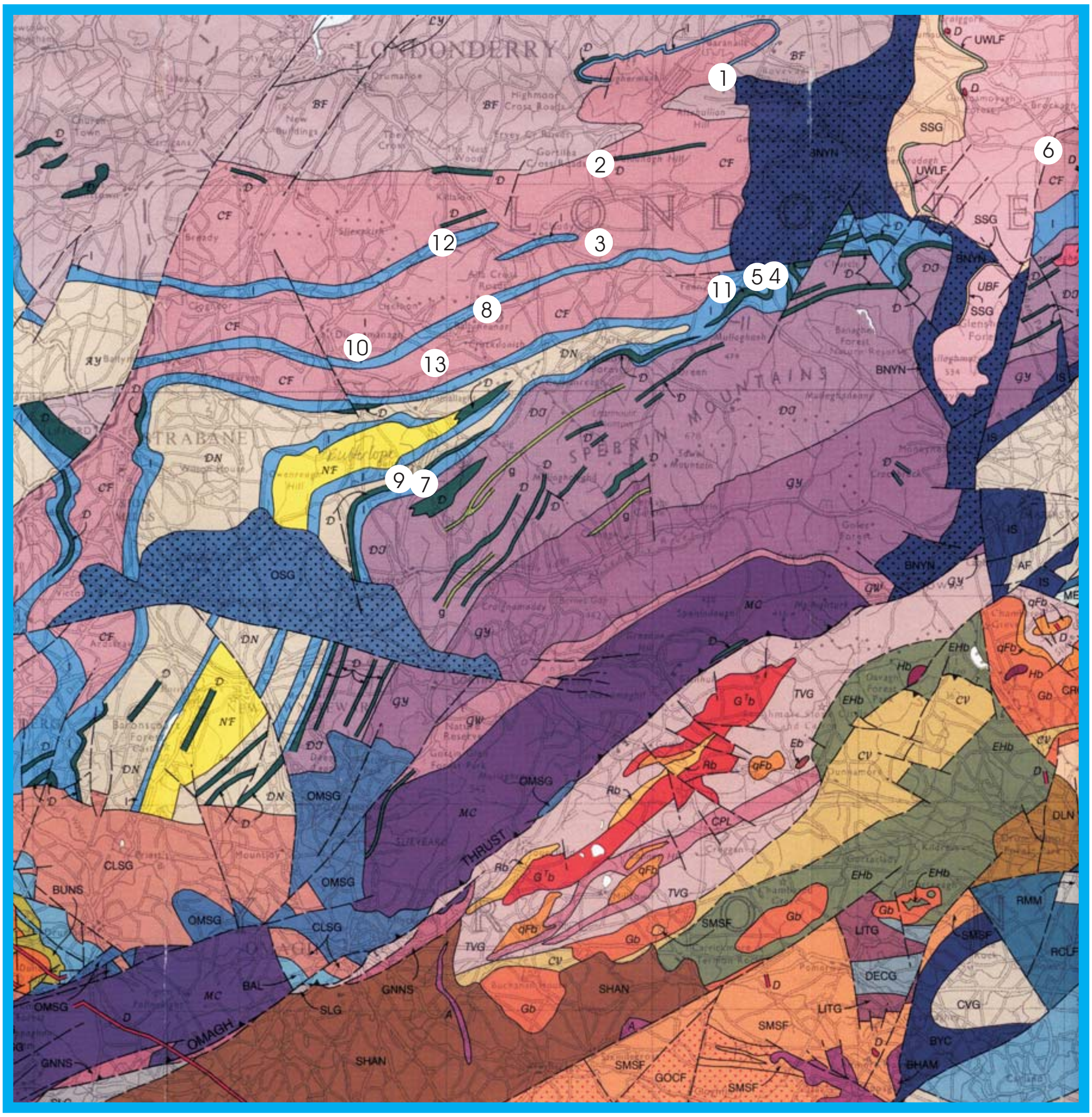


Figure 2. Geology of central Ulster and the location of samples of Dalradian metacarbonate rocks in the Dungiven and Claudy Formations (Argyll and Southern Highland Group respectively). Taken from 1:250 000 Series Solid Geology map of Northern Ireland, 2nd edition, 1997.

CF : Claudy Formation — Southern Highland Group

DN : Dungiven Formation
NF : Newtonstewart Formation
} Argyll Group

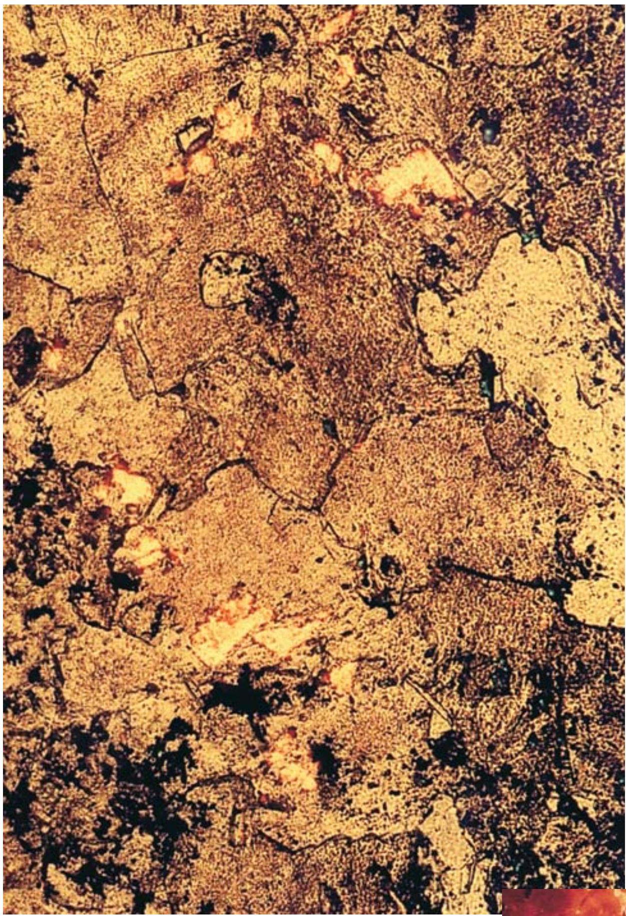


Figure 3. Pale orange luminescent relict calcite in dolomite.

Dolostone, Dungiven Formation
Buttelop Glen

GSNILS 7

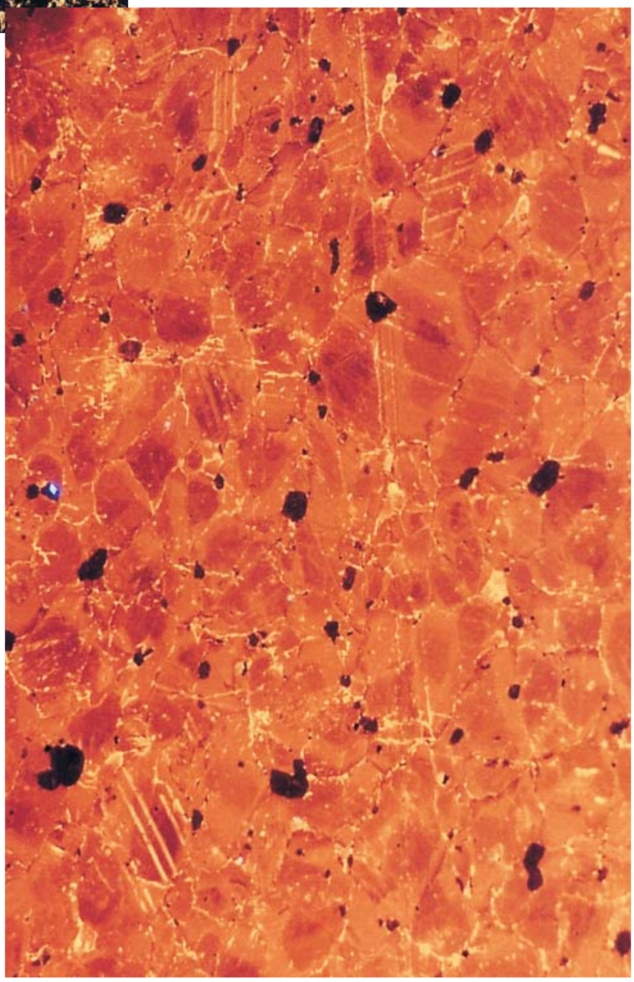


Figure 4. Calcite crystals with interiors of dark orange brown luminescent calcite. Note the locally sharp to more diffuse margins to the darker interiors. Note also how bright yellow luminescent calcite picks out the grain boundaries locally. This bright yellow calcite is taken to record the passage of infiltrating metamorphic fluid. See text for discussion.

Limestone, Kennedy Quarry,
Glen Bonds

Claudy Formation

GSNILS 12

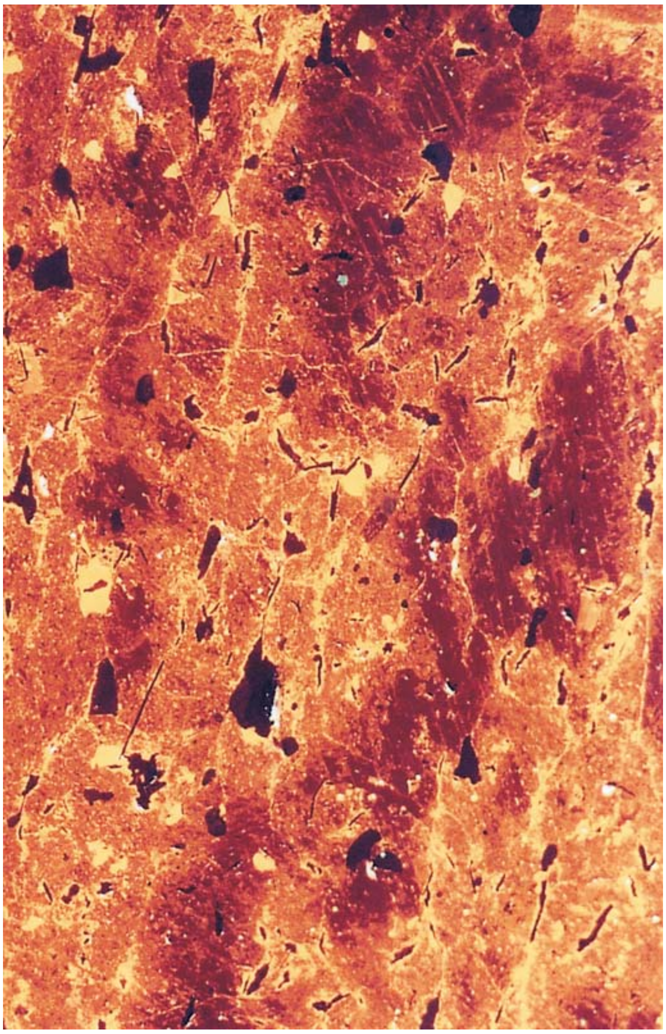


Figure 5. Relict dark-luminescent calcite cores in domains of orange to bright yellow luminescent calcite. This image is indicates extensive alteration of the original, pre-metamorphic calcite chemistry. The textural character of the alteration suggests that it resulted from metamorphic fluid-rock interaction.

Limestone, Tirglassan Quarry, Claudy Formation
GSNILS 3

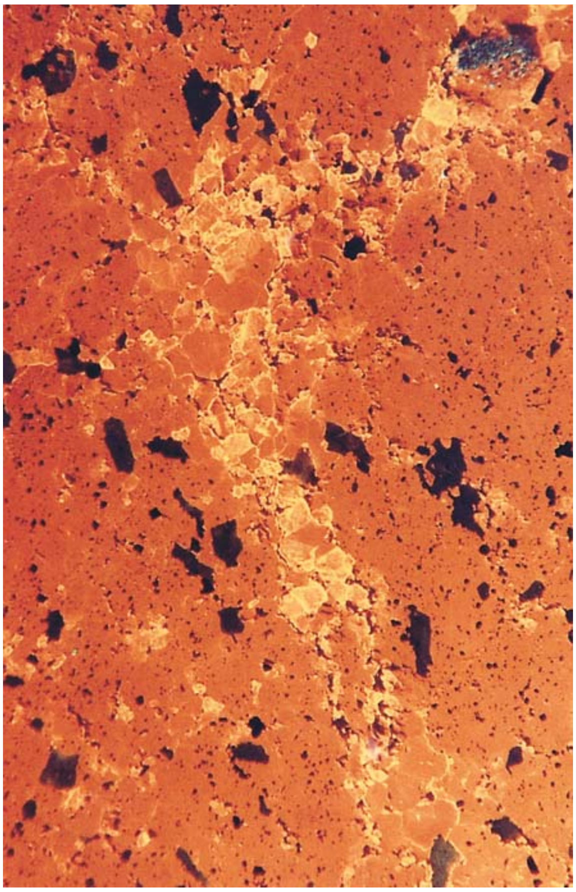


Figure 6. Large calcite plates with even orange luminescence finged by ‘granular’ finer grained and more yellow-orange luminescent calcite. The large calcite plates are optically continuous and contain aligned trains of dusty ?graphite inclusions.

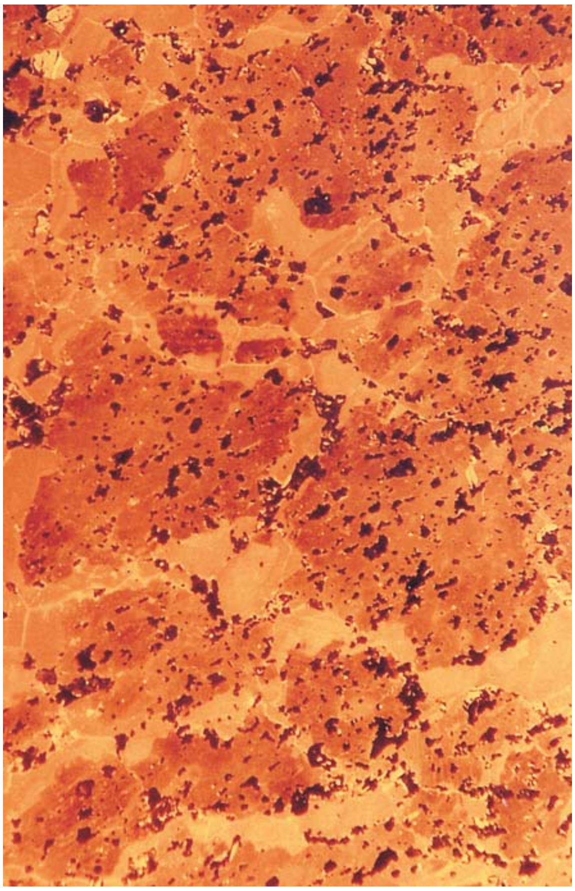
Limestone, Rallagh Quarry, east
Dungiven Formation

GSNILS 4

Figure 7. Sample of limestone from Rallagh Quarry, west, showing discrete, dark orange-luminescent, inclusion-rich calcite plates fringed by more orange- and yellow-luminescent calcite. cf. Figure 6. It is thought this may be an annealed version of GSNILS 4.

Dungiven Formation

GSNILS 5



8a

8b

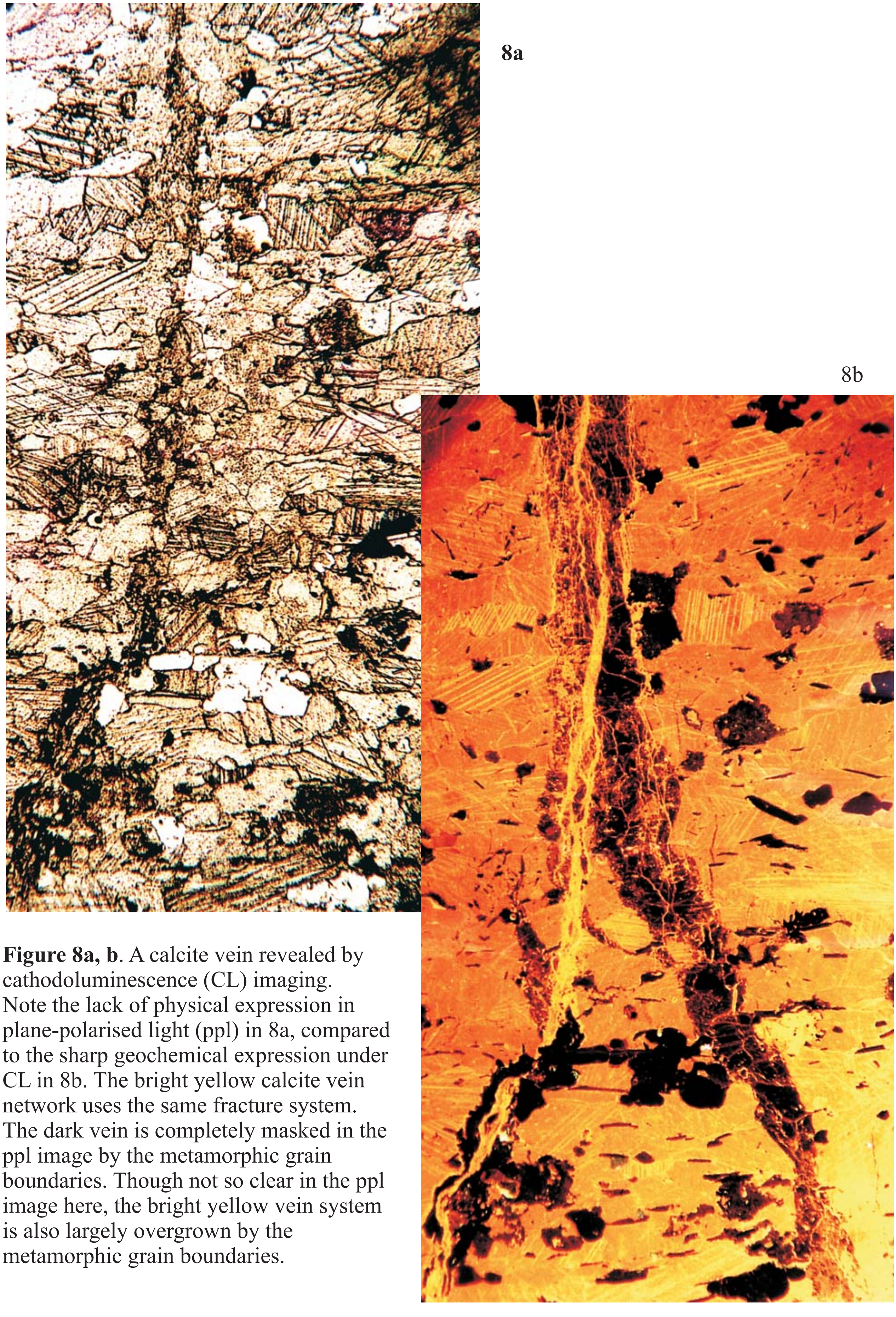


Figure 8a, b. A calcite vein revealed by cathodoluminescence (CL) imaging. Note the lack of physical expression in plane-polarised light (ppl) in 8a, compared to the sharp geochemical expression under CL in 8b. The bright yellow calcite vein network uses the same fracture system. The dark vein is completely masked in the ppl image by the metamorphic grain boundaries. Though not so clear in the ppl image here, the bright yellow vein system is also largely overgrown by the metamorphic grain boundaries.

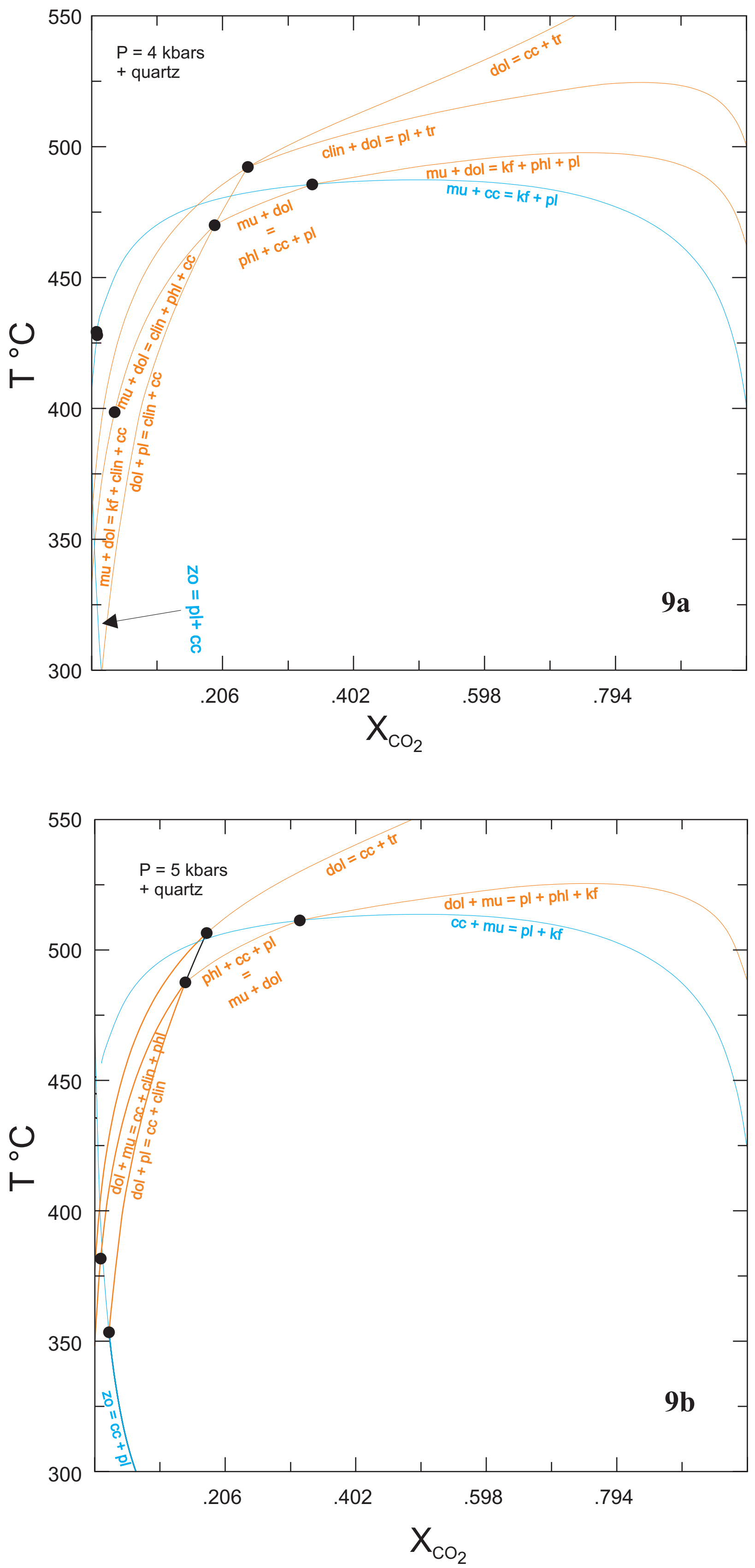


Figure 9a,b. T - X_{CO_2} sections for mineral assemblages in the dolostones and limestones. Phase equilibria shown in blue are for the limestones, those in red for the dolostones.

Sample	Formation	Location	Grid Reference (Irish Grid)	Lithology	Detailed lithostratigraphical position or member
GSNILS 1	Claudy Formation	*Glenconway, stream section*	26353 41479	Metadolostone	top
GSNILS 2	Claudy Formation	*Ballyholly Quarry*	25742 41082	Metalimestone	top
GSNILS 3	Claudy Formation	*Tirglassan Quarry*	25975 40708	Metalimestone	middle
GSNILS 6	Claudy Formation	*Ashlamaduff stream section*	27920 41169	Metalimestone	middle
GSNILS 8	Claudy Formation	*Longlands Road Quarry*	2522 4040	Metalimestone	Alla Limestone Member
GSNILS 10	Claudy Formation	*Drain Quarry*	24580 40185	Metadolostone	Alla Limestone Member
GSNILS 12	Claudy Formation	*Kennedy Quarry, Bonds Glen*	24985 40700	Metalimestone	Bonds Glen Limestone Member
GSNILS 13	Claudy Formation	*Lough Ash Road*	24945 40125	Metalimestone	not given
GSNILS 4	Dungiven Formation	*Rallagh Quarry East*	26588 40567	Metalimestone	top
GSNILS 5	Dungiven Formation	*Rallagh Quarry West*	26574 40570	Metalimestone	top
GSNILS 7	Dungiven Formation	*Butterlope Glen*	2491 3952	Metadolostone	not given
GSNILS 9	Dungiven Formation	*Balix Hill*	2480 3958	Metalimestone	not given
MRC 95	Dungiven Formation	*Oldmill Bridge*	23766 38400	Dolomitised metasandstone	not given
MRC 102	Dungiven Formation	*Bessy Bell*	23866 38173	Dolomitised metasandstone	not given
MRC 107	Dungiven Formation	*Mary Gray*	24297 38541	Metalimestone	not given
MRC 108	Dungiven Formation	*Victoria Bridge*	2355 3906	Metalimestone	not given
MRC 121	Torr Head Formation			Metalimestone	not given
MRC 122	Torr Head Formation	*Torr Head*	32342 44047	Metalimestone	not given
MRC 123	Torr Head Formation			Metalimestone	not given

Table 1. Location, lithological and lithostratigraphical details of the samples of limestone and dolostone discussed in the report

Location	Sample	$^{87}Sr/^{86}Sr$	error	Sr	Mg/Ca	1/Sr	Mn*	Mn/Sr	Rb	Rb/Sr	$\delta^{18}O_{SMOW}$	$\delta^{13}C_{PDB}$
Southern Highland Group												
Leny Limestone												
Callander, Perthshire	HY1362	0.711859	0.000030	796		0.00126	n.a.		0.03	0.00004	15.7	-4.6
	HY1364	0.712781	0.000017	815		0.00123	n.a.		0.04	0.00005	18.3	-5.0
	Median :	**0.712320**		**806**					**0.04**		**17.0**	**-4.8**
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Boyne Bay, NE Scotland	HY147	0.709018	0.000016	4607	0.0296	0.00022	310	0.07	13	0.0028	19.0	1.9
	HY148	0.709451	0.000016	2695	0.0620	0.00037	697	0.26	43	0.0160	19.8	0.5
	HY149	0.709305	0.000014	3534	0.0334	0.00028	697	0.20	23	0.0065	18.2	-1.9
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	Median :	**0.709301**		**3380**			**620**		**33**		**18.6**	**0.9**
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	T3	0.708960	0.000017								16.6	1.1
	T4	0.709096	0.000018					No elemental data available			16.3	1.1
	T5	0.709467	0.000020								17.5	1.5
	T6a	0.709637	0.000017								17.7	1.6
	T6b	0.708969	0.000016								17.3	2.0
	Median :	**0.709096**										
Torr Head Limestone												
Torr Head, Co. Antrim	HY1348	0.708857	0.000039	1865	0.0109	0.00054	144	0.08	26	0.0139	14.8	7.4
	HY1349	0.708810	0.000032	2115	0.0080	0.00047	144	0.07	20	0.0095	17.9	7.5
	Median :	**0.708834**		**1990**			**144**		**23**		**16.4**	**7.4**
Dungiven Limestone												
Banagher Glen	HY1350	0.708220	0.000033	265	0.0838	0.00377	3890	**14.68**	3	0.0113	13.4	7.0
Butterlope Glen	HY1351	0.708745	0.000043	2299	0.0070	0.00043	216	0.09	12	0.0052	23.4	7.3
	Median :	**0.708483**		**1282**			**2053**		**8**		**18.4**	**7.2**

Data from Thomas (1999)

* data converted from wt% oxide to ppm element

Table 2. $^{87}Sr/^{86}Sr$, $\delta^{18}O$ and $\delta^{13}C$ & Sr, Mn and Rb data for 'Tayvallich-equivalent' and Southern Highland Group limestones