Dolomitization of the Carboniferous Limestone of the Peak District: a Review

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Abstract. Large areas of the Carboniferous Limestone of the southern Peak District have been dolomitized, particularly the more coarse-grained calcarenitic facies. After a summary of the physical features of dolomitized limestone, its stratigraphic distribution and relationship to mineralization, the evidence points to a late Carboniferous date of dolomitization. Possible sources of the magnesium are from buried shales in adjacent basins, effectively as an early flush of hydrothermal fluids, from altered mafic minerals in volcanic rocks or both. It is proposed that a magnesium-rich fluid moved ahead of the hydrothermal mineral fluids and was exhausted before the mineral veins were infilled.

Some 50 km^2 of the Carboniferous Limestone outcrop in the southern half of the Peak District shows evidence of alteration of the original limestone to dolomite, locally known as dunstone from its dull brownish grey colour on weathered surfaces. The dolomitized area (Fig. 1) is less than a tenth of the total White Peak limestone outcrop, but the alteration has produced both distinctive rocks and landforms such as dolomite tors (Ford, 1963). Magnesium was introduced in mobile groundwaters long after sedimentation and resulted in the metasomatic growth of dolomite crystals within the limestone, so progressively obscuring the sedimentary fabric and fossils (Parsons, 1922).

While the character of the alteration and the distribution are well known, there has been little study of the cause of dolomitization or the processes that might have been involved. Furthermore, two schools of thought have emerged concerning the date of dolomitization, broadly late Permian or late Carboniferous: each would imply the operation of a different mechanism for dolomitization.

Dolomites also occur amongst the lowest beds seen in the Wye Valley and were penetrated in the few deep boreholes sunk towards the sub-Carboniferous basement (Cope, 1973; Dunham, 1973; Chisholm & Butcher, 1981; Chisholm et al. 1988). These dolomitic beds are associated with other evidence of sabkha-shoreline conditions to be expected at the start of an early Carboniferous marine transgression and are not regarded as part of the high-level dolomitization discussed here.

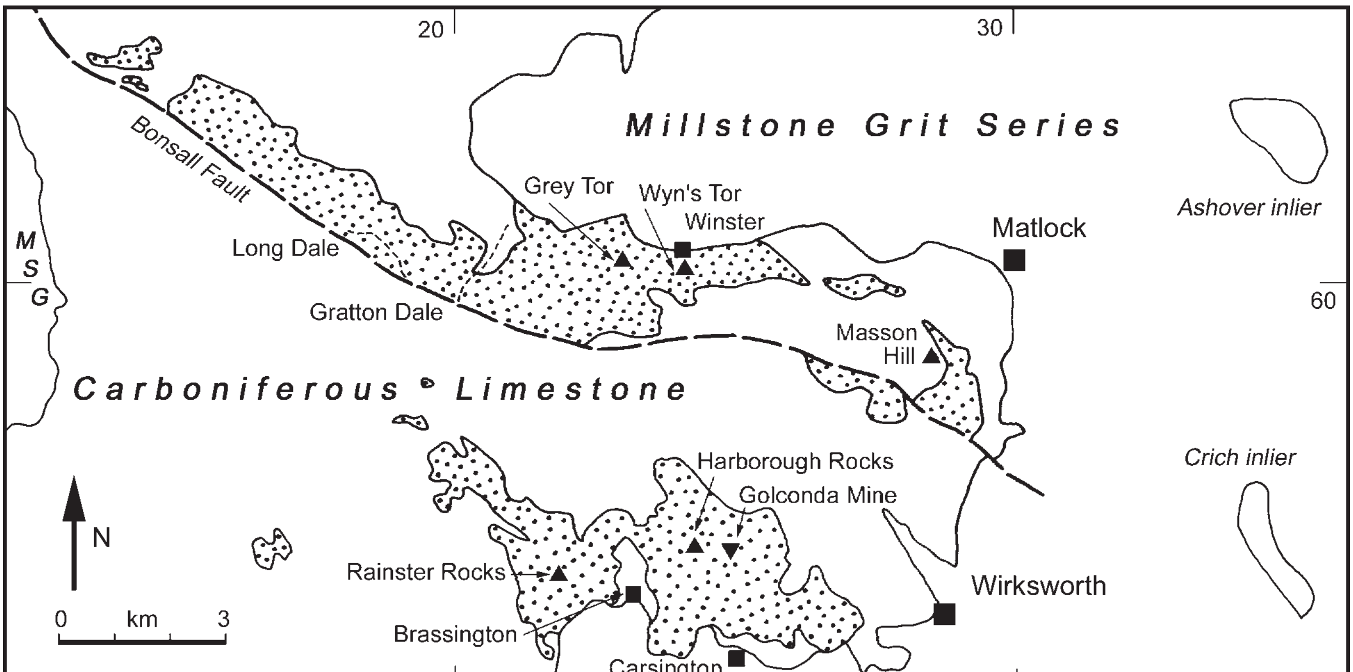

Figure 1. The southern White Peak showing the dolomitized limestone areas (stippled).

The Carboniferous Limestone inliers of Breedon on the Hill in Leicestershire have also been dolomitized (Parsons, 1918) but these are directly overlain by the unconformable Mercia Mudstone of late Triassic age. It seems that a different process has operated there: it is not considered further herein.

The aims of this communication are to review the constraints on the process of dolomitization in the Peak District and to discuss possible mechanisms.

Petrography

Dolomite is the double carbonate of calcium and magnesium, $CaMg(CO_3)_2$ (Deer, Howie and Zussman, 1992). It was named after the 18th century French geologist D. G. de Dolomieu (King, 1995). In the Peak District it has developed as euhedral to subhedral crystals within the coarser-grained limestones of the southern half of the White Peak (Parsons, 1922). The Limestone and Dolomite Resources Reports produced by the British Geological Survey in the 1980s gave many analyses and several photomicrographs (Bridge & Gozzard, 1981; Bridge & Kneebone, 1983; Cox & Bridge, 1977; Cox & Harrison, 1980; Harrison & Adlam, 1985) but most did not discuss the dolomitization process. Indeed, Cox & Bridge (1977) simply commented that the cause was unknown and that magnesian fluids could have moved downwards from the Permian or upwards from below.

Together these reports show that the dolomitized limestone is never pure dolomite (dolostone in American terminology) but that the degree of alteration is generally around 50-80% with the remainder of the rock as interstitial calcite. The content of MgO does not reach the theoretical maximum of 21%, and maxima are generally below 20% (Cox & Harrison, 1980). This degree of alteration, though incomplete, is enough to yield the textures and colour of dunstone. Surface weathering gives a very porous appearance but at depth interstitial calcite reduces the porosity. Boreholes reported in the BGS resources surveys have shown that the degree of dolomitization is highly variable both vertically and horizontally, and that the coarser calcarenites have usually been altered most.

Dolomitization has taken the form of the growth of euhedral to subhedral dolomite crystals in the body of the rock (Fig. 2). Their growth has resulted in partial or almost complete loss of original textures; fossils were destroyed or left as ghost outlines. Some beds contain silicified fossils and chert nodules and these are unaffected, being left surrounded by dolomitized limestone.

With the dolomite molecule being smaller than that of calcite, there is only a little loss of calcium, which has apparently been redeposited elsewhere, such as in mineral veins. The remaining calcite is usually recrystallized in the interstices. However, calcite is more soluble and weathering processes tend to remove it preferentially, eventually leaving a loose dolo-sand residue. Periglacial sludging takes the dolo-sand away leaving upstanding cores of still-solid rock known as dolomite tors: Grey Tor and Wyn's Tor near Winster provide good examples (Ford, 1963).

Distribution

As noted by Parsons (1922) and in the various Geological Survey Memoirs (Frost & Smart, 1979; Aitkenhead et al., 1985; Chisholm et al., 1988; Smith et al., 1967), dolomitized limestone is largely confined to two strips of country in the southern White Peak, one from Long Dale to Winster and the other around Brassington and Carsington on the south flank. Well-known occurrences in the former area are Wynn's Tor and Grey Tor above Winster, and in the latter area are Harborough and Rainster Rocks. Each of these areas totals around 20-25 km^2. A detached area of about 2 km^2 caps Masson Hill above Matlock, and there are a few scattered outliers.

A few old quarry faces and several lead mines show that the dolomitized limestone lies on top of unaltered limestone. The depth to the base of the dolomite is highly variable, but is usually not more than 40 m. The deepest dolomitization base recorded is in Golconda Mine (Ford & King, 1965) at about 120 m. Here it undulates along a NW-SE axis in flat-lying beds, and gives rise to a false concept of an anticlinal disposition of the mineral deposits that lie on the flanks of the "high" of unaltered limestone. In Masson Hill, Matlock, the base of dolomitization is generally along the line of the Masson, Rutland and Wapping Mines, but descends sharply almost to river level in Temple Mine (Ford, 2002).

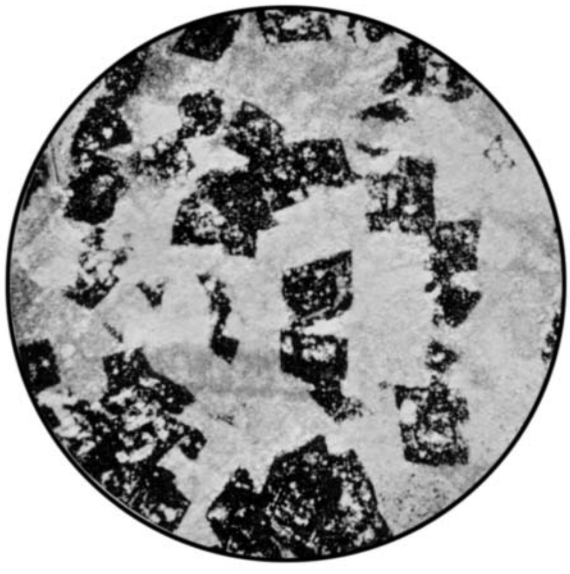

Figure 2. Photomicrograph of dolomitized limestone with dolomite rhombohedra, from Rainster Rocks (photo: G S Sweeting).

Dolomitization does not normally extend beneath the Namurian shale cover to the east, indeed the rather shaly facies of the highest limestone beds (the Eyam Limestones) are generally not altered. Only in the vicinity of Winster is dolomitized limestone known to extend beneath the shale cover for about 1.5 km (Aitkenhead et al., 1985). However, the stripping back of the shale cover to its present position around Winster and Brassington is a Pleistocene feature and the Millstone Grit cover with the thick Edale Shales at its base extended over the whole dolomite outcrop in pre-Pleistocene times.

The stratigraphic relationships of dolomitized limestones are shown in Table 1. In the Long Dale and Gratton Dale area, dolomitization has altered the Bee Low Limestones of Asbian age. Further east, around Winster, it is mainly the Monsal Dale Limestones of Brigantian age which are altered, but the alteration extends upwards into the Eyam Limestones where the more massive limestone beds of reef facies are present. Both Asbian and Brigantian beds have been dolomitized around Brassington. The presence or absence of lavas within the limestones sequence appears to have had some influence, as lavas are few and far between in the Long Dale and Brassington areas, where Asbian beds are affected. On Masson Hill, where there are two thick lavas and several clay wayboards in the Brigantian Monsal Dale Limestones, dolomitization rarely extends downwards into the underlying Bee Low Limestones. The distribution along the two flanks of the western extension of the Masson anticline raises the question as to whether the eroded upper limestones between the two areas might have formed a dolomitization cap to the upwarp but no clear evidence of this has been found.

On Masson Hill, dolomitization is largely confined to the more massive limestone beds between the two main lavas. These beds are about 40 m thick, with several clay wayboards. Dolomitization is confined to the beds above the “Little Toadstone” wayboard, some 6 m above the Matlock Lower Lava (Ixer, 1978). Both the limestones below the Little Toadstone and the dolomitized limestones immediately above have been mineralized with fluorspar replacements, pipe-vein cavities lined with minerals, and minor scrins (Ixer, 1974, 1978). An up-dip fault postulated by Ixer seems to have little effect on dolomitization, but the alteration dies out suddenly down-dip of the Masson Pipe mineral deposit (Ford, 2002).

The boundary of dolomite on limestone is usually sharp, with the transition taking place in a few millimetres. The boundary is often along clay wayboards, but Ixer (1978) noted that the ratio of MgO fell to a minimum immediately above and below wayboards in the Masson opencast, implying that the reduced permeability either side of the wayboards restricted fluid migration. Manystones Quarry, near Brassington (now partly back-filled with mineral processing waste), showed a dolomite/limestone contact undulating across the face with downward prolongations of dolomite on major joints (Fig. 3). Within a short distance to the east the base of dolomitization plunges more than 120 m to the main levels in the Golconda Mine. Together with the dolomitized Harborough Rocks, the total thickness of dolomitization there is around 150 m - rather less than the estimate of 200 m given by Harrison and Adlam (1985). A kilometre further east, an irregular dolomite/limestone contact is visible outside the Gallows Knoll tunnel on the High Peak Trail. Elsewhere a few sections show a gradation from dolomite to limestone, apparently as grain-size decreases. Locally there is an alternation of dolomite and limestone along the contact, where only the coarse-grained limestones have been dolomitized. The fine-grained facies, with much less porosity, remains largely unaltered. Some contacts are nearly vertical and follow joints, demonstrating that the limestones were sufficiently lithified to have developed a joint system before dolomitization. Details of other contacts are to be found in Parsons (1922) and in the BGS resources reports.

stratigraphic unit	dolomitization
Brigantian	
Eyam Limestones	limited
Monsal Dale Limestones (Upper)	extensive
Matlock Upper Lava	
Monsal Dale Limestones (Lower)	extensive
Matlock Lower Lava	
Asbian	
Bee Low Limestones	patchy
Holkerian	
Woo Dale Limestones	contemporary

Table 1. Stratigraphy of the dolomitized limestones.

The strip of dolomitization from Long Dale to Winster is roughly parallel to but not always in contact with the Bonsall Fault. The latter cuts off the dolomitized limestone along part of its length, demonstrating that at least the later fault displacements were post-dolomitization. Neither the Brassington nor Masson Hill areas of dolomitization are juxtaposed to the Bonsall Fault.

The distribution of dolomitized limestone outcrops is similar to but not identical with that of the Neogene (Brassington Formation) silica-sand pocket deposits (Aitkenhead et al., 1985). As these two phenomena are separated by some 270 million years the process of dolomitization is not thought to be associated with the deposition of the Brassington Formation.

Relationship to mineralization

Previous researchers are agreed that dolomitization preceded mineralization. Large and small veins (rakes and scrins) pass from one host rock to the

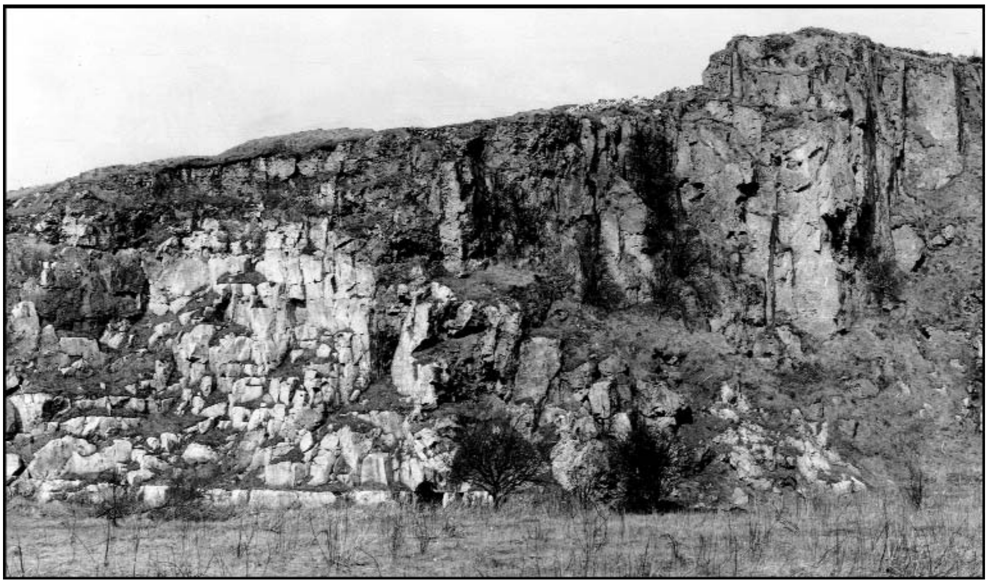

Figure 3. An old face in Manystones Quarry, near Brassington showing the irregular contact of darker dolomitized limestone above and paler unaltered limesone below. The lower part of this section is now buried by waste material.

other without any substantial change. Perhaps the most obvious example is the Great Rake on Masson Hill and High Tor, extending from limestone host rock in Riber Mine to dolomite above Masson Mines (Ford, 2002). Flat, pipe and replacement mineralization is common at or just below the contact of dolomitized and unaltered limestones, particularly along the Little Toadstone wayboard, as in parts of the Masson Mines (Dunham, 1952; Ixer, 1974, 1978; Ixer & Vaughan, 1993; Jones et al. 1994; Ford, 1999; 2002). In the Black Ox workings of the Masson Mines, thin layers of galena partially fill bedding planes and micro-joints in pre-existing dolomite. In the Golconda Mine, near Brassington, the base of dolomitization also follows a clay wayboard in places (Ford & King, 1965). It is notable that the mineral veins do not include dolomite as a primary mineral though it is occasionally present as adventitious lumps or dolo-sand derived from the walls. Dolo-sand in the centre of a baryte-lined vug in Golconda Mine post-dates galena-barytes mineralization (Ford & King, 1965).

Studies of successive generations of cementation of limestones by Hollis (1998) and Hollis & Walkden (1996) have shown that the calcite cement was deposited in four layers which they named Zones, each characterized by different trace element chemistry. Both magnesium and the elements of the ore minerals increase in late Zone 3 and in Zone 4 - which those authors regard as having been deposited during deep burial in late Carboniferous times, at much the same time as the main mineralization phase. While their studies hint at dolomitization being partly contemporary with mineralization, they did not discuss the relationship of their findings to the dolomitized limestones discussed herein.

An attempt to obtain magnesium metal from dolomite near Hopton in the 1960s proved uneconomic, as only about half the anticipated yield could be obtained. One of the factors is thought to have been the minor lead-zinc mineralization on the many joints interfering with the extraction process.

The date of dolomitization

As dolomitization preceded mineralization, the real question is - when did mineralization take place? Also, in turn, did mineralization precede or follow the folding of the Matlock anticline? Dunham (1952) argued, briefly, that dolomitization had occurred as a result of downward percolation of magnesian brines from a late Permian Zechstein marine transgression across the South Pennine region, and several authors have accepted this argument uncritically. However, among others, Frost & Smart (1979) and Cope et al. (1992) thought it unlikely that the Permian transgression had extended over the South Pennines. If dolomitization was late Permian, then the subsequent mineralization must have been very late Permian or Triassic in age. An alternative possibility that dolomitization might have been due to Triassic groundwaters (Aitkenhead et al., 1985) would necessitate mineralization having been post-Triassic, for which no evidence has been presented.

Dolomitization clearly followed lithification of the limestones sufficient for them to have a joint and fracture system. The distribution of dolomitization suggests that it was later than the folding of the Matlock anticline. The latter had been growing since Dinantian times as shown by facies changes in the Eyam Limestones and the thinning of the Edale Shales over the Matlock anticline (Smith et al., 1967; Ford, 2002). Upfolding culminated in the Westphalian, as part of the Variscan orogeny at the end of the Carboniferous, demonstrating a fairly rapid sequence of events at this time – folding and fracturing - dolomitization – renewed fracturing – vein formation. Without reliable means of dating these episodes, it is difficult to constrain the sequence in a definite chronological framework but a sequence of events in late Westphalian to Stephanian times, i.e. part of the Variscan orogeny, is logical.

Burial history curves presented by Hollis (1998) (after Colman et al, 1989) show that maximum depth of burial of the limestones at about 2500 m was reached in late Westphalian times and that mineralization reached a climax then. From Stephanian times (latest Carboniferous) onwards, there was a steady reduction in the cover with waning mineralization. Though Hollis did not discuss dolomitization, there is a clear implication that it preceded mineralization at some stage in the Westphalian.

Fluid inclusion temperatures generally ranging around 100-120°C have been obtained mainly from fluorite crystals in the veins (Atkinson et al., 1982; Colman et al., 1989). They indicate that there was a cover of around 2000 m of Upper Carboniferous strata on top of the limestone at the time of mineralization. Projection of the Permian base from the Mansfield area suggests that most of the Coal Measures and Millstone Grit had been eroded off the limestone by late Permian times, so the depth of burial necessary for the fluid inclusion temperatures was not available then. These stratigraphic arguments confirm the burial histories deduced from cementation generations by Hollis (1998) and Hollis & Walkden (1996). Furthermore Triassic rocks lie unconformably on the limestone around Ashbourne and Snelston, indicating complete removal of the cover over at least that small area. How much cover remained on the dolomitized areas in the late Permian and Triassic may be uncertain, but it is difficult to argue for enough to account for the depth of burial sufficient for the formation of the mineral veins.

An as-yet unsubstantiated report (Willies, pers. comm.) of pebbles of vein-stuff in the Sherwood Conglomerate of the Trent Valley region may indicate that vein minerals were available for erosion from an unknown mineral deposit in the southern Peak District by Triassic times, again indicating that much if not all of the Upper Carboniferous cover had then been eroded away. .

In addition, if there had been a thick cover of Coal Measures and Millstone Grit in late Permian times, their thick shales would have obstructed downward percolation of magnesian brines to the limestone. And why would such percolation have occurred in such limited areas?

On the basis of alteration of wayboard clays adjacent to mineral veins, Ineson & Mitchell (1972) applied the K-Ar isotope dating method and deduced that there had been episodic mineralization from mid-Carboniferous to Triassic, with a climax in the late Carboniferous. However, the dating method has its critics and perhaps not too much significance should depend on these results.

On structural grounds, it seems that mineralization was an episodic process culminating in mid to late Carboniferous times (Quirk, 1986, 1993). He argued that the changing directions of stress fields from late Dinantian to end Westphalian yielded fracture systems with varying orientations as a form of ground preparation. Mineral infill of the fractures could have been contemporaneous or later. Plant & Jones (1989) regarded the main episode of mineralization as being in a short period in the late Westphalian (upper Coal Measures). This is consistent with the timing of the "inversion" of the South Pennine sedimentary basin to an area of uplift as a result of the Variscan orogeny. The hydrothermal fluids responsible for both the dolomitization and the mineral vein infills would have been able to migrate from the basin into the growing upwarp before too much cover had been removed.

The argument that dolomitization was pre-Triassic because the fill of the silica-sand pockets was Triassic (Kent, 1957) has been negated by dating of the latter as Neogene.

Source of the magnesium

Discounting the Upper Permian Zechstein sea brines as a source of magnesium on chronological grounds, as discussed above, a possible source or sources must be sought elsewhere in the South Pennine region. There are several possibilities, all with problems arising.

As with hypotheses concerning the origin of the solutes in hydrothermal fluids resulting in mineralization, a potential source of magnesium

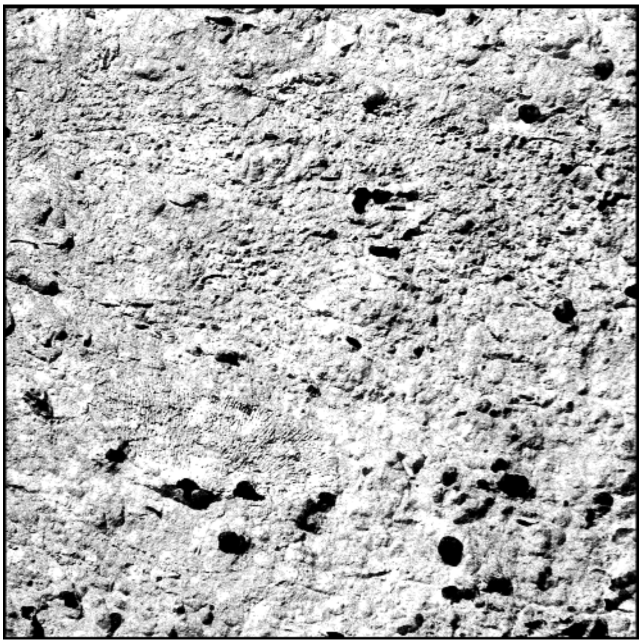

Figure 4. A dolomitized limestone surface on Rainster Rocks. The shadowed hollows are probably where calcite fossils were preferentially dissolved out, though a colonial coral survives in the lower left.

Figure 5. Wyn's Tor – a crag of dolomitized Brigantian limestone crag near Winster.

could be in the Dinantian/Namurian shales of adjacent basins. Few analyses of the clay minerals of the shaly basinal equivalent of the Dinantian limestones and the overlying shales of Namurian age anywhere in the Pennines have been published (Plant & Jones, 1989), and no significant figures for magnesium have been traced for shales in the South Pennine basins. As with the deltaic sandstones of the Millstone Grit, the clay minerals there have been derived from parent rocks in the Caledonian mountain belt containing some mafic minerals, so a small proportion of magnesium could be expected. If this was expelled as an early fluid pulse during burial diagenesis, a magnesium-rich fluid could have migrated into the Peak District limestones ahead of the main hydrothermal fluid. Such a flush of magnesium-rich fluid could have penetrated the coarser-grained limestones and given rise to dolomitization before being exhausted; later pulses of hydrothermal fluid then yielded the vein minerals that filled the fracture systems. It is thus possible to visualize a magnesium-rich front travelling through the limestones and causing dolomitization. Once the magnesium source was exhausted, dolomite was not available to fill fractures and so does not occur in the vein mineral assemblages.

A second possible source of magnesium could be the dolomites of the Woo Dale Limestones (the lowest exposed beds in the Wye Valley) (Aitkenhead et al., 1985) or the deeply buried dolomites found in the few boreholes to the base of the Carboniferous Limestone at Woo Dale (Cope, 1973), Eyam (Dunham, 1973), Via Gellia (Chisholm & Butcher, 1981) and Cauldon Low (Chisholm et al., 1988). The first two boreholes encountered some 50-100 m of dolomites at the base of the Carboniferous sequence resting on a basement of Precambrian or Lower Palaeozoic rocks, and the others were thought to be close to basement before being terminated. However, as these concealed beds near the base of the limestone sequence are still dolomites, it is difficult to see how they could have provided sufficient magnesian fluids while still remaining dolomite. It is probable that the early Carboniferous marine transgression started with a sabkha shore-line phase on the South Pennine block, so any adjacent basins could have had a thicker sequence of dolomites, but these have yet to be proved by deep boreholes.

The third possible source of magnesium is from altered volcanic rocks. The exposed lavas are basaltic, containing a large proportion of mafic minerals, principally augite with subsidiary olivine. Alteration by hydrothermal fluids and by weathering converts these to clay minerals, mainly chlorites, releasing magnesium. But was there enough magnesium released to account for all the dolomitization? From the visible altered portions of the lavas probably not, but there is evidence of considerable amounts of concealed basaltic volcanics further east. At Ashover, some 200 m of basalts and basaltic breccias were found in boreholes without reaching their base (Ramsbottom et al., 1962). There was much alteration in what was thought to be a vent beneath the Ashover anticline, and magnesium could have been released from mafic minerals therein. Much further east, a substantial volume of concealed volcanics was found in the Coal Measures of the N. E. Leicestershire coalfield, but having these as a source would require a mechanism to transfer the magnesian brines, both down to the limestone through the Coal Measures and Millstone Grit with their thick shales, and for a considerable distance to the Peak District. Taken together with other as yet unknown concealed basalts, the volcanics could have been a source of magnesium distinct from the usual hydrothermal mineral source in shales, but no mechanism for transfer of magnesian fluids from the basalts to the limestones has yet been proposed.

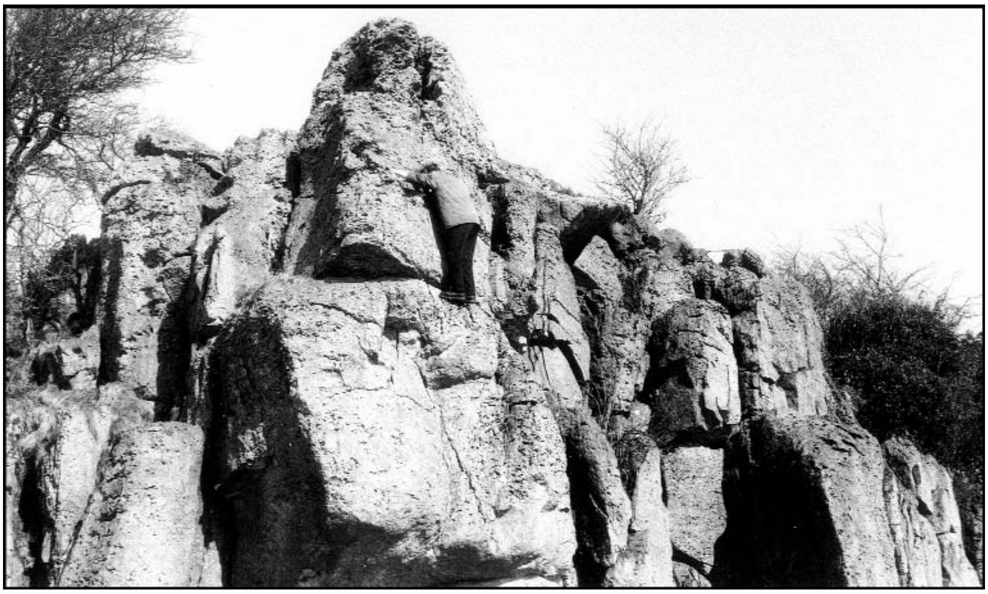

Figure 6. Rainster Rocks – dolomitized limestones of Asbian age.

Quantification of how much magnesium could be obtained from these sources and how much has been deposited in the partially dolomitized limestones is impossible owing to lack of knowledge of the efficiency of extraction and migration and the quantities of the potential source rocks. On balance it seems that the shales must be the preferred source, with a possible contribution from altered volcanics.

The apparent restriction of dolomitization to the present limited areas of outcrop may be misleading, as a dolomitized limestone crest to parts of the Matlock anticline and its western extensions has been eroded away.

Dolomitization in other areas

No comparable large-scale post-depositional dolomitization of the Carboniferous Limestone has been found in other British orefields (Ixer & Vaughan, 1993). Limited areas of contemporary fine-grained dolomite are present in the North Wales and Mendip successions of the Carboniferous Limestone, but no such dolomite has been recorded in the North Pennines. Dolomite is present as a vein mineral in the latter though it is not known as a vein mineral in the South Pennine orefield. Widespread dolomitization of the Waulsortian mud-mound facies of the Carboniferous Limestone in southeast Ireland has been taken to indicate a magnesium-rich fluid front moving upwards from the adjacent Munster Basin (Hitzman et al., 1998). The fine-grained character of the limestone apparently formed no obstacle, as the mud-mounds have abundant stromatactis cavities providing permeability. While the general setting is comparable to the Peak District, the limestone facies and tectonic setting in Ireland are different. A broadly comparable situation to that in Ireland has been described in Cambrian rocks in Missouri (Gregg & Shelton, 1989).

That there is no comparable dolomitization in other British orefields suggests a distinctive source or mechanism in the Peak District, but none has been determined. However, it should be noted that none of the other orefields has lavas interbedded with the limestones. While these may not have been the main source of magnesium they certainly were guiding horizons with limited permeability which could have concentrated magnesian brines at certain limestone horizons, as they did the mineralizing fluids later.

Conclusions

Dolomitization affected some 50 km2 of the Carboniferous Limestone in the southern Peak District. The coarse-grained calcarenite facies seem to have been most altered with generally sharp but transgressive boundaries, often guided by joints. Dolomitization penetrated to depths averaging around 40 m with a maximum of around 150 m, with unaltered limestone below. Dolomitization is rarely complete but has resulted in the distinctive brownish dunstone. Both Asbian and Brigantian beds are affected, with the distribution guided to some extent by lavas and clay wayboards. Dolomitization was post-folding but pre-mineralization and, as the latter is now generally regarded as late Carboniferous, so dolomitization must also have been late Carboniferous in date. Sources of magnesium are likely to have been the Dinantian and Namurian shales of adjacent sedimentary basins, effectively providing an early flush of magnesium-rich hydrothermal fluids, but a contribution from altered basaltic volcanics is possible.

Acknowledgments

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