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Quartzite dissolution: karst or pseudokarst?

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Abstract

A wide range of landforms of great similarity to limestone karst is found on many of the world's quartz sandstones and quartzites. These landforms have often been dismissed as pseudokarst, but recent investigation shows that the dissolutional removal of silica, even quartz, under earth-surface conditions is a critical process in their formation. They must therefore be regarded as true karst features. Recognition of these genetically similar forms on quartzose rocks now demands the worldwide adoption of a less restrictive, process-based, karst definition. Direct evidence for this near-surface dissolutional weathering is not common. Examples of this process are reviewed here, along with further evidence for the dissolution of silica from within the quartz sandstones of the Sydney Basin in temperate south-eastern Australia. Some of the complex processes by which dissolution attacks the rock remain unclear. However the solubility, thermodynamics, fluid throughput and physical removal of detritus are all critical factors in the formation of what can only be termed karst on quartzites and quartz sandstone.

Keywords: pseudokarst, dissolution of quartzite, quartzose karst

Introduction

Detailed study of the calcareous karst of central and eastern Europe began several hundred years ago (reviewed by Shaw, 1992) and has since developed into a highly structured field of research (e.g. Sweeting, 1972, 1981; Jakucs, 1977; Jennings, 1985; Ford and Williams, 1989). It was perhaps unfortunate though that most early definitions of karst, many of which still linger today, were essentially restricted to carbonate bedrock, because it was not long before a range of similar landforms started to be identified on quartz-rich rocks. These non-carbonate morphologies, whilst commonly identical in size, shape and apparent formative process to their limestone analogues, were therefore dismissed as pseudokarst. That is to say, they were considered to be like karst, but only a scientific curiosity and not generally worthy of detailed study. However, even though dissolutional processes have now been demonstrated as a causative agent in the genesis of many of these forms on quartzose rocks, many geomorphologists and geologists following conventional wisdom, have been loath to change their outlook (eg., Marker, 1976; Vitek, 1979; Pouyllau and Seurin, 1985; Osbome and Branagan, 1992; Yanes and Briceno, 1993).

This paradox of dissolutional landforms on some of the world's most insoluble rocks mimicking those on some of the most soluble, both in appearance and scale, has become increasingly difficult to ignore in recent years, yet little attention has been given to the detailed study of the landforms themselves or the dissolutional processes involved. It appears that this may be because of a long-standing and, as it now seems, falsely grounded assumption that quartzose rocks are of extremely low or almost negligible reactivity and, unlike limestone, are *"practically immune to chemical weathering"* (Tricart and Cailleux, 1972, p. 152). Yet, during the last three decades the large scale dissolution of silica, including the least soluble quartz, has been demonstrated clearly and repeatedly as being critical to the development of the great quartzite karst landscapes of Venezuelan Roraima (White et al, 1966; Urbani and Szczerban, 1974; Chalcraft and Pye, 1984; George, 1989; Briceno and Schubert, 1990) and those on the quartz sandstones of Tchad in central Africa (Mainguet, 1972), the Arnhem Land and Kimberley regions of northern Australia

(Jennings, 1979; Young, 1986, 1987, 1988) and the Sydney region in south-eastern Australia (Wray, 1995). See Wray (1997) for a detailed review.

The problems of karst terminology

Considerable terminological difficulties are inherent in the question of what exactly constitutes karst (Twidale, 1984). Many definitions of karstification are restricted to discrete rock or landform types on lithological or morphological grounds (e.g., Dreybrodt, 1988; Ford and Williams, 1989; Self and Mullan, 1996), but such definitions may not always be appropriate. Absence or scarcity of surface drainage, collapse features, caves, grikes, runnels and speleothems are distinctive, but they are not peculiar to karst (Twidale, 1984). Gams (1989) went as far as to state that certain surface landforms are regarded as typical in karst regions but are not essential. He thought that only two phenomena are essential: efficient dissolution of rock and karst drainage.

In recent years several wider-ranging, process-based, definitions that are not restricted by rock type or landscape morphology have been proposed. Sweeting (1972, p. 5) stated that: *"...the sinking of water and its circulation underground is the essence of the karst process...*" but also followed with "*... this process is dominated by a chemical (solutional) activity, and true karst landforms result largely from the action of one erosive process, namely solution".* Grimes (1975) argued that true karst could occur on lithologies other than limestone provided that dissolution was the dominant process, and Gams (1989, p. 169) felt that"*... the processes which control the shape of the landscape can include non-karst processes and are not restricted to solution. However, solution remains the most intensive geomorphic process".*

Ford (1980, p.345) went further, qualifying the importance of dissolution by noting that "*... true karst forms are distinguished from pseudokarst forms by the necessity of rock solution. True forms may be excavated entirely by aqueous solution, or other processes may contribute largely to their dimensions; but where this latter applies, solution plays an essential precursor or 'trigger role' ".* Jennings (1983, p.21) similarly noted the importance of dissolution by stating that karst is the "*... process, solution, which is thought to be critical (though not necessarily dominant) in the development of the landforms and drainage characteristics"* of an area. He later followed with: "*... karst is a terrain with distinctive landforms and drainage arising from greater rock solubility in natural waters than elsewhere... Solution is not always the most prevalent process in karst, nor is it necessarily the dominant one, but it does play a more important role here than in other kinds of landscape"* (1985, p. 1).

More than just the presence of dissolutional weathering is therefore required for sandstone karst development, for dissolved silica is present in most natural waters (Aston, 1983). The dissolution must"*... contribute largely...*" and act in a way that is a "trigger-role" (Ford, 1980) or be "*... critical (but not necessarily dominant"* (Jennings, 1983) in the preparation of the rock leading to the development of landforms that otherwise would not arise. A variety of chemical and physical weathering processes are involved in the development of limestone karst landforms (Jennings, 1985; Ford and Williams, 1989) and, similarly, dissolutional landforms on quartz sandstones or quartzites are also formed by a variety of physical and chemical processes. But the common factor that sets them apart from more general sandstone weathering is the critical action of chemical dissolution.

Wray (1997) presented a wide-ranging and comprehensive analysis of the worldwide distribution and morphology of karst on quartzite and quartz sandstones. This review demonstrates clearly that landforms are found on quartzose rocks that are comparable in type, form and scale to those described from many limestone regions. The present paper examines the evidence for dissolutional weathering within some of these quartz sandstones.

Evidence for karstic dissolution of quartzite and quartz sandstone

Many examples of silica dissolution within deeply buried quartz sandstones are found in literature describing petroleum occurrences, but unquestionable proof of the dissolutional removal of silica from such rocks under near-surface conditions is rarer. However microscopic evidence from several locations shows clearly that slow near-surface dissolution of quartz, 'arenisation' in the terms of Martini (1979), occurs typically along crystal boundaries, freeing individual sand grains that are ideally suited to removal by flowing water.

In the hot, wet, tropical area of central Venezuela, large table-mountains of the Precambrian Roraima Group orthoquartzites display some of the world's most highly developed and striking quartzose karst landforms (see George, 1989). White et al (1966) argued from petrographical evidence that quartz within these rocks was hydrated to much more soluble opal and then removed in solution. Scanning

Electron Microscope (SEM) examination of these quartzites (Chalcraft and Pye, 1984) revealed a widespread and intense crystallographically controlled microscopic attack of detrital quartz grains and overgrowths. Clear evidence was found that widening of grain-to-grain contacts, coupled with etching and corrosion of both quartz grains and cement, eventually leads to the freeing of individual detrital grains. Chalcraft and Pye (1984) rejected the White et al opal hydration mechanism, and demonstrated the direct dissolution of quartz grains and silica cement without an intermediate hydration phase. They also showed that while there is preferential dissolution along joints, beds and lithological contacts, cracks at all scales provide foci for water flow and pathways for dissolution.

Ghosh (1991) found that surface outcrops of weathered Roraima quartzites display an excellent network of lamellar porosity formed by dissolution of quartz cement along overgrowth boundaries. However, samples from deep below the surface show abundant welding of grains by a pervasive syntaxial quartz cement with sutured grain-to-grain contacts. This variation in weathering between surface and buried samples demonstrates that this quartz dissolution is a surface phenomenon, and did not originate at depth in a process similar to that recently recognised by petroleum geologists in the formation of secondary sandstone porosity (Pye and Frinsley, 1985; Shanmugam, 1985; Burley and Kantorowicz, 1986; Hurst and Bjorkum, 1986). However the resultant dissolution features under both circumstances are quite similar.

Wilson (1979) presented SEM images of minute v-shaped chemical etch pits on the quartz grains of the Millstone Grit of South Wales. He attributed these to slow dissolution by fluids of high pH seeping along cleavage and/or fracture planes, and presented evidence for the formation of similar features in the laboratory. Battiau-Queney (1984) also found that the Grit had suffered a long subaerial weathering, possibly under a hot wet tropical climate, resulting in this quartzite being extensively weathered.

Burley and Kantorowicz (1986) analysed microscopic quartz grain surface features (including small pits, notches and larger embayments) produced by dissolutional attack on deeply-buried sandstones. They found a tendency for corrosion to be more intense on those surfaces with a high free-surface energy, such as grain peripheries, along fractures and at crystal boundaries. Two mechanisms of quartz corrosion were proposed. The first, *"transport controlled dissolution"*, is influenced by the rate of transport of fluids to and away from the reactive surface, or the reaction rate at the surface. This results in rapid, non-specific corrosion at all available sites, and is typical of attack by strongly concentrated solutions. *"Surface reaction controlled dissolution"* is generally more specific, tends to produce crystallographically controlled features, such as well defined v-shaped pits, and is typical of slow dissolution.

An SEM investigation of the regional extent and intensity of quartz sandstone etching in the seasonally-arid east Kimberley region of northern Australia (R W Young, 1988) provided graphic evidence of such surface-reaction and transport controlled dissolution features. Young (1988) also noted that Hurst and Bjorkum (1986) had challenged the ideas of Burley and Kantorowicz (1986) by arguing that quartz dissolution rates are too low for transport-controlled etching, emphasising that etching will concentrate at the sites with the highest free-energy, and quartz overgrowth lowers the surface free-energy of a detrital grain. Brady and Walther (1990) and Withe and Peterson (1990) also supported the proposition that silica dissolution occurs preferentially at high-energy surface sites such as defects, and is controlled by the density of such defects. Dissolution should therefore be most rapid at the greatest concentration of detrital grain surfaces, face-corners and edges of overgrowths.

Though R W Young (1988) found widespread dissolution in these high-energy locations, he also found that the intensity of quartz etching in the Kimberley region was variable and more closely related to the primary porosity of the host rock. The more porous sandstones display a higher potential for the penetration of corrosive solutions, with subsequent deep weathering that alters the geotechnical properties of the sandstone.

Intense silica dissolution has also been demonstrated in highly quartzose sandstones of the humid-temperate Sydney region of south-eastern Australia. Tropical conditions have been absent from this area for tens of millions of years (Bird and Chivas, 1989), yet A R M Young (1987) published clear SEM evidence for near-surface dissolution of detrital quartz grains, overgrowths and high free-surface energy grain and overgrowth boundaries. As voids are widened by dissolution detrital sand grains are loosened and removed easily by flowing water.

Wray (1995) studied the sandstone landforms of the Sydney region in more detail, finding widespread microscopic evidence for intense, but spatially variable, dissolutional removal of silica (Figs 1 through 3). Etching within many surface sandstones is much higher than reported from deeply-buried sandstones in this region. In fact, the type, location and intensity of near-surface dissolutional weathering is very similar to that reported from the tropical and seasonally-arid regions noted above. The same small crystallographically controlled pits are seen, along with many larger areas of non-specific dissolution. Grain contacts, overgrowth boundaries and other discontinuities or defects are much more corroded than most overgrowth faces, but even some overgrowth faces are etched, especially in the most weathered sandstones, most commonly on the rhombohedral faces.

The primary porosity of these sandstones is also a major factor governing the intensity of dissolutional weathering and the landforms that later develop (Wray, 1995). Attack of the overgrowths and cement changes the physical properties of the sandstone. Sandstones with little interconnected void provide few pathways for water penetration and are usually only mildly weathered close to the rock surface. Sandstones with a high degree of effective porosity commonly display striking karstic morphology. They are generally very highly etched, their grains are only weakly held, and the rocks have lost much of their physical strength and resistance (Wray, 1995).

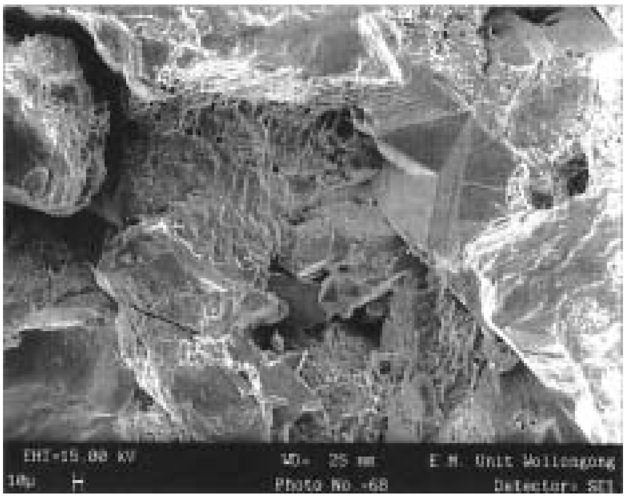

Fig. 1. SEM micrograph showing intense dissolutional attack within the early Permian Snapper Point Formation at Jervis Bay, south of Sydney, Australia. The amount of quartz overgrowth within these sandstones is quite high, but without total elimination of effective porosity. Greatest dissolutional attack is in areas of high free-surface energy (detrital grains, overgrowth faces and edges); however many of the overgrowths, especially prism faces, are only slightly corroded. Cement and overgrowth sutures binding the sandstone have begun to dissolve leaving narrow voids between grains, resulting in a reduction of strength and the production of a sandstone ideally suited to increased physical erosion along joints. Field of view 250 µm.

Fig. 2. SEM micrograph showing intense 'transport controlled' etching within the early Permian Snapper Point Formation at Jervis Bay, south of Sydney, Australia. Much of the overgrowth is highly corroded, though other areas are only slightly etched. Field of view 250 µm.

Fig. 3. SEM micrograph of well sutured but still highly permeable Nowra Sandstone from south of Sydney, Australia. Intense non-selective dissolution is seen on the high free-surface energy grains and overgrowth faces or edges, but lower energy faces display lesser attack. Because of the dissolution this sandstone is very friable in tension and shear, but the interlocking nature of the grains still imparts a high compressive strength. Granular disintegration of this sandstone gives rise to a tower morphology very similar to that illustrated by Young (1986) in the Bungle Bungle Range in the Kimberleys of north-western Australia. Field of view 1OO µm.

Whilst it is undeniable that dissolutional etching and removal of silica from quartz sandstones is found in many locations, it is also necessary to assess the degree to which such dissolution is necessary in the formation of karstic landscapes.

Briceflo et al. (1990, p. 192) discussed the geology and surficial geochemistry of the landscape in the Roraima area, observing that the complex karstic landscapes developed primarily because of the weathering of the quartzites: "... *mainly by solution of siliceous cement in sandstone, which released sand grains"*. Once this cement dissolves, the rocks become very friable and ideally suited to erosion by flowing water channelled preferentially along zones of weakness such as joints and bedding planes. These findings indicate unequivocally that formation of this topography has been dependent upon the effects of "... *chemical weathering, together with the constant removal of detritus"* (Briceflo et al, 1990, p. 179). Briceflo and Schubert (1990, p. 131) examined the geomorphology of Roraima, finding once again that dissolutional weathering of the quartzites, coupled with removal of the detritus produced by flowing water, are the critical landscape forming processes: *"The surface is intricately sculptured, in large measure due to exploitation of fractures by solution of the siliceous cement of the quartzite"*.

In South Africa Martini (1979) provided clear evidence for the integral nature of dissolutional weathering allied with removal of detritus under vadose conditions in the development of karst in the Black Reef Quartzite. Mainguet (1972) and Busche and Erbe (1987) also argued for the pivotal role of intensive dissolutional removal of silica in the formation of the widespread sandstone karst of the south-central Sahara.

R W Young (1986, 1987, 1988) also showed that sandstone etching is widespread in the Kimberley region of northern Australia and illustrated clearly that variations in landform morphology are dependent on the intensity of dissolutional weathering and the resultant geotechnical properties of the weathered sandstones. Where dissolution has been most intense - "... *granular disintegration of sandstone is dominant, as in the Bungle Bungle massif, (and) symmetrical towers and arêtes, often separated by flat-floored embayments, are formed. As granular cohesion increases and the enlarging of fractures becomes the dominant process, the shape of the towers and ridges becomes more irregular until they merge into Mainguet's 'ruiniform 'terrain. On highly indurated sandstone, like those of the Cockburn and Osmond Ranges, cliff and cave assemblages are dominant"* (p.216). It is therefore clear from Young's analysis, like those in Venezuela and Africa, that dissolutional weathering of the Kimberley sandstones has been a critical factor in the development of this region's landscapes, and that variations in the intensity of dissolution have a marked influence on the erosion of the landscape and the resultant landforms.

Like Young, Wray (1999) also found that the range of karstic features on quartz sandstones in the Sydney Basin of temperate southern Australia is reflected in the variable intensity of dissolutional weathering. The most highly weathered sandstones are quite friable, with grikes, runnels, basins, small caves, speleothems and even tower karst not uncommon. Where the dissolution is less intense the sandstones retain much of their cement and sutured overgrowths, and are more robust, less liable to granular disintegration, and only rarely display any karstic features. The variability in dissolutional weathering is therefore clearly a very significant factor in landscape development on these sandstones. A forthcoming paper will discuss these aspects in more detail.

The dissolutional process in quartzose karst

Dissolutional weathering is undoubtedly a highly significant, even critical, stage in the silica karst process. But, though the locus of attack within the rocks is generally clear, some of the detailed mechanisms of this arenisation are complex and still not fully understood.

The equilibrium solubility of silica and especially quartz is low (amorphous silica 100-140 mg/1, quartz 6-14 mg/1 at earth-surface temperatures) (Yariv and Cross, 1979). But solubility is not the only controlling factor. Thermodynamics and reaction kinetics are also critical in the formation of silica karst. Martini (1981) has argued that the rate of reaction is just as important as the total solubility; a faster reaction rate limits the distance solutions can penetrate the rock before saturation, resulting in arenisation close to the surface and a general surface lowering rather than deep karstification. Slower rates allow greater water penetration and joint widening without surface lowering, and still-slower rates lead to crystal boundary dissolution with a deep general weathering of the rock. Voids along crystal boundaries are very narrow and water circulation is very sluggish, with saturation being reached after only a very short distance unless the reaction kinetics are very slow. If the silica dissolution rate was slower still, without changing the total solubility, Martini believes that quartzite karst would be much more common.

Moreover, the bulk removal of silica is dependent not only on its solubility and kinetics, but also on the rate at which water moves through the rock. Douglas (1969) demonstrated that the silica load of rivers is dependent upon runoff, and thus upon rainfall. Douglas (1978, p.230) later commented that: *"the importance/or the rate of solution of relatively rapidly moving water has been demonstrated in limestone terrains... but it is equally significant in silicate areas. ... the rate loss of ions from silicate minerals to waters is controlled by the speed at which dissolved ions are carried away from the surface of the mineral"*. Rimstidt and Bames (1980) also emphasised the importance of this 'flushing rate'.

It therefore appears that the rate or volume of water movement through the region is one of the keys to the problem; the higher the water throughput the higher the expected rate of silica removal. Where water throughput is high, silica remains mobile and may enter streams and be removed from the area, whereas where flushing rates are lower it is not removed as effectively, and may be reincorporated in the neoformation of clays (Young and Young, 1992). However, constant increase in the rate of water throughput will not result in a constant increase in dissolved silica. Flushing accelerates the dissolution of minerals only up to a still unclear limiting rate beyond which additional through flow of water has virtually no effect, and dissolution is controlled by mineral reactivity (Bemer, 1978). This may be seen in the similar degree of quartz etching achieved under vastly different rainfall regimes in the tropical, semi-arid and temperate sandstones discussed above.

But even on the very highly karstified quartzites of Roraima limited field measurements indicate that the chemical conditions under which the karst is forming are at odds with laboratory results, which indicate that highly alkaline conditions are necessary to achieve significant silica dissolution (Krauskopf, 1956; Siever, 1962; Yariv and Cross, 1979). Pouyllau and Seurin (1985) argued for silica dissolution occurring under hyper-acid conditions, but they could provide only limited evidence for the existence of slightly acid conditions. Likewise, Chalcraft and Pye (1984) and Briceno et al (1990) found natural waters at Roraima to be only slightly to moderately acidic.

Field measurements of dissolved silica by Chalcraft and Pye (1984) also showed dissolved silica levels to be quite low (< 1 to 7 mg/1). Similar levels of dissolved silica are also recorded from quartz sandstone terrains in Tchad (Mainguet, 1972), Amhem Land (Noranda Ltd., 1978; Dames and Moore, 1981), and the Sydney region (Johnson and Johnson, 1972; Johnson, 1984; Wray, 1995), even though precipitation regimes differ dramatically between these areas. These results all indicate that silica is being dissolved only slowly, and suggest that these landscapes have formed by slow dissolution over a very long period.

This factor of slow but very prolonged dissolution is one that has all too commonly been ignored, and the low solubility and slow dissolution rate of silica, especially quartz, was believed to preclude formation of karstic landforms on these 'inert' rocks. Though Self and Mullan (1996) have argued for the rapid development of some karstic features on the Fell Sandstone of northern England, in the areas where the most extensive and highly developed quartzose karst are found (notably South America, Australia and Africa) the available evidence indicates that slow rates of dissolution have been offset by very long periods of sub-aerial weathering (Chalcraft and Pye, 1984; Busche and Erbe, 1987; Young, 1986, 1987, 1988; Briceflo et al, 1990; Wray, 1995).

Long periods of arenisation result in a rock that eventually becomes incoherent and is thus highly suited to physical erosion. Martini (1981) notes that karst on quartzite cannot form by dissolution alone, and Jennings (1983) emphasised that while dissolution is critical it actually removes only a small proportion of the rock (about 10 to 20% of rock bulk). The carbonation of limestones leaves behind little residue but the chemical attack of water on silicate rocks produces large volumes of residue, loosened by dissolution, that must be removed by physical transport (Jennings, 1983). Therefore, a plentiful supply of flowing water is necessary, preferably under vadose conditions. Physical erosional processes are thus just as important, if not more important, in the formation of quartzose karst as they are in other quartz sandstone regions.

Conclusions

Distinctive landforms that have developed on many quartzites and quartz sandstones around the world are virtually identical in form and genetic formative process to many landforms developed on limestones (Wray, 1997). It has become ever clearer during the last three decades that use of the term karst can no longer be restricted to describing carbonate terrains. It has also been argued increasingly that karst should not be reserved for the description of specific landscape morphologies, but should be seen more as the process of significant rock dissolution.

Viewing karst more as a process, rather than purely as a morphology or lithology, no longer restricts use of the term artificially to describing particular landforms or rock types. Because it is dissolution that endows karst with its particular characteristics, process-based definitions should now be advocated, thus allowing recognition of the critical genetic process whilst incorporating the essentials and avoiding the limitations of other karst definitions. Such process-based definitions also allow a more natural grouping of landforms that display a similar mode of origin and feature morphology within a range of rock types. This type of definition is thus consistent with geomorphological terminology employed for other landforms (Loffler, 1978).

A significant component of rock dissolution affecting landform development is the key element, critical to karst development. Although pseudokarst features bear a close resemblance to karst forms, they evolve in response to other dominant processes and the action of rock dissolution is not necessarily involved in their formation. Now that widespread karst on quartzites and quartz sandstones has been clearly recognised the blanket application of the term 'pseudokarst' is commonly unjustified. However, as argued by Twidale (1984) for dissolutional landforms in granite, until the formative processes are known the immediate application of the term karst to landforms with karst-like morphology may be over simplistic. Therefore, the use of distinctive names that are available and established in the literature may be preferable (Twidale, 1984).

Although dissolutional processes that act upon quartzose rocks are very different to those affecting carbonates, silica dissolution has been, and will continue to be, clearly demonstrated as having an integral role in the development of sandstone karst. Hopefully, the recognition of increasing numbers of these landforms as true karst, not pseudokarst, should prompt further much-needed critical investigation, especially of the complex chemical interactions and geotechnical effects of dissolution under a wider range of geological, geographical and climatic settings.

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