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# Are there “black holes” in carbonate deposystems?

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## ABSTRACT

The likelihood that extensive dissolution of aragonite (and high magnesian calcite) takes place during very early burial, even in relatively shallow tropical settings, has wide implications for interpreting and modelling ancient limestones. Some low energy environments may constitute net sinks (“black holes”) for carbonates. If this is the case attempts to model sediment budgets and develop depth-productivity profiles for carbonate systems require as yet unavailable quantitative data on the effects of these processes. We query whether the established trends of diminishing carbonate productivity with depth could, for some settings such as ramps, be in part an artifact of selective, offshore dissolution more than simply due to reduced benthic production. Many fossil assemblages from low energy settings may be limited to refractory (residual) skeletal elements, which limits our ability to make palaeoenvironmental determinations: some important carbonate grain producers such as the green algae may have been especially prone to early dissolution. The possibility exists that what is now mud-grade carbonate in low energy deposits may not represent original material deposited from suspension but could represent transformed aragonite from in situ production by the skeletal fauna. The selective removal of labile aragonitic (and high Mg calcite) components could produce sediments that appear to have had the composition of cool water types, and the reduced diagenetic potential of many Palaeozoic and cool water carbonates could be a consequence of the effects of early aragonite dissolution rather than due to differences in primary (depositional) composition.

*KEYWORDS* **Dissolution. Taphonomy. Carbonate ramps. Diagenesis.**

## INTRODUCTION

It is a truism that carbonate deposystems differ from siliciclastic dominated ones in many ways but especially because many carbonate grain types are produced effectively in situ. Local, very shallow water, mainly benthic sediment production is the fundamental process operating in most carbonate settings. However, to see carbonate sedimentation as simply a matter of deposition alone, as in some forward modelling studies, is over simplistic. In reality any carbonate accumulation is product of a series of losses, gains, translocations and transformations, in many ways analogous to the processes that operate in a soil. By comparison with the quartz grains of a sandstone, carbonate components are much more susceptible to a range of processes operating during or immediately after deposition. Gains to the system include input from benthic production, but may also include imported components from suspended and transported bed load, and from local precipitation of carbonate as mud-grade particles or cements. Carbonate grains may be translocated by bioturbation, leading to bio-retexturing (Pedley, 1992). Organisms also play a role in transformations in grain size such as those caused by micritisation. Recrystallization, effectively syn-sedimentary, can occur without the intervention of endoliths (Reid and Macintyre, 1998). In many cases the transformation is not just a physical effect on grain size or shape, but also affects mineralogy. Losses from the system include transportation, bioerosion and dissolution. In tropical, warm water deposystems carbonate undersaturation is normally perceived as a process operating in deeper water settings (below the lysocline), or where meteoric or mixed waters flush carbonate sediments following a fall in sea-level. There is evidence that syn-sedimentary dissolution does take place even in tropical platform interiors; Walter and Bruton (1990) and Ku et al. (1999) estimated that 50% of the annual carbonate produced in the Florida Bay lagoons is lost by dissolution. Sanders (2003) has provided an important review of these syndepositional dissolution processes and their causes. Cool water carbonates are also susceptible to early mineral stabilization and dissolution in marine waters (Kyser et al., 1998; Nelson and James, 2000).

Munnecke and co-workers (Munnecke et al., 2001; Munnecke and Westphal, 2004) have discussed the role of early dissolution in the formation of calcareous rhythmites, and Cherns and Wright (2000) and Wright et al. (2003) have provided detailed examples of the effects of these processes on benthic faunas from Silurian and Jurassic carbonate ramp successions. Sanders (e.g. 2001) has discussed the role of early dissolution on Upper Cretaceous rudist-bearing carbonates. Furthermore, the role of shallow, marine-fluid derived dissolution and its ability to mimic meteoric diagenesis has been discussed by Melim and co-workers (Melim et al., 2002).

The aim of this paper is to explore some of the sedimentological implications of large-scale early carbonate dissolution (excluding bioerosion) in settings above the lysocline. We have few answers but many questions, most of which we believe need to be addressed with some urgency.

## EARLY DISSOLUTION

The long held view that tropical shallow marine sediments, composed of aragonite, high Mg calcite and minor amounts of low Mg calcite were effectively stable during shallow burial in marine fluids is compromised by the growing mass of evidence for the operation of early dissolution. Most studies on the effects of this syndepositional dissolution have invoked increased acidity caused by microbial decay of organic matter, through such processes as the oxidation of organic matter or by the oxidation of products of anaerobic sulphate reduction (see Sanders 2003 for authorative review). Aragonite and high Mg calcite are especially prone to dissolution. To what extent ambient sea water chemistry played a role at various times in the past is unclear, with some authors favouring sea floor dissolution as being a feature of tropical seas in calcite-sea intervals (Palmer et al., 1988). Stanley and Hardie (1998) have suggested that the temporal abundance of some fossil groups was linked to seawater chemistry ("calcite" or "aragonite" seas), relating to controls on actual skeletal secretion. Another possibility is that such trends, at least in part, reflect a preservation bias, particularly during "calcite" sea intervals when aragonitic skeletal grains could have been more susceptible to early dissolution. However, the fact that early dissolution is indicated from studies of modern sediments deposited in today's "aragonite" sea, suggests that the chemistry of the whole shallow water body, while having some role, is not the main factor.

The prime requirement for dissolution is the availability of organic matter for decay. The most likely sites will be relatively low energy settings where fine particulate organic matter can accumulate, such as lagoons and offshore areas. Thus there will be a strong facies control on the process, as demonstrated by Wright et al. (2003) for the Lower Jurassic of South Wales, where all the biogenic aragonite was removed syndepositionally in offshore, mud-dominated settings but in equivalent inshore facies composed of originally clean, skeletal sands, the accumulated aragonite was replaced during later burial. We propose that there are sedimentary environments where selective dissolution is a major process resulting in the net loss of significant amounts of carbonate sediment; low energy settings should not simply be seen as storage sites for sediment but as potential sinks or "black holes" where sediment is effectively destroyed.

This poses the question as to what extent many fossil assemblages are simply refractory ones left after early dissolution has taken its toll? Cherns and Wright (2000) were able to estimate that the aragonitic molluscan component of the Silurian (Wenlock) benthos of Gotland had been depleted at least 100-fold by early dissolution. They were able to do this by comparing a typical fauna (dominated by calcitic forms such as brachiopods) with one from a similar setting in which the whole skeletal fauna had been silicified very early, preserving an abundant and diverse former aragonitic molluscan fauna. The 100-fold figure was derived by comparing the abundance of former aragonitic forms between the two assemblages. Although it was not possible to calculate the actual volume loss from dissolution it must have been very large. In a second study Wright et al. (2003) showed that this "missing molluscs" effect was also a factor in early Jurassic outer ramp facies in South Wales.

## SEDIMENTOLOGICAL IMPLICATIONS OF EARLY DISSOLUTION

### Depth-Productivity Profiles and Modelling

One of the fundamental tenets of carbonate sedimentology is that production is depth related with the highest production of sediment in very shallow water (Ósscher and Schlager, 1993). The depth-productivity profile characteristic of modern low nutrient, tropical settings, dominated by calcareous algae or algal-symbiont-bearing corals is not applicable to all ancient carbonate deposystems (Wright and Faulkner, 1990; Aurell et al., 1997), especially when the carbonate producers were heterozoans. Pomar (2001a) has emphasized the importance of different depth-productivity profiles in influencing platform geometry, such as whether steep margined platforms or ramps develop. Compared to modern reefal settings, carbonate ramp deposystems display a less marked offshore depth-productivity gradient (Wright and Faulkner, 1990), or in some ramps carbonate productivity may have been enhanced in offshore settings relative to inshore sites (Pomar, 2001b; Brandano and Corda, 2002). In most cases the development of a ramp profile requires a gradual decrease in accumulation in an offshore direction.

Devising depth-productivity profiles is a basic element used in forward modelling of ramp stratigraphies, and some workers have identified unique profiles for different ancient successions by this technique (Aurell et al., 1997; Read, 1997). If synsedimentary carbonate dissolution is a major process in low energy settings, such as mid to outer ramp zones, then the decreased production rates modelled for such settings may, at least in part, be a function of the loss of material rather than simply its lack of production. In such a situation to refer to such profiles as "production profiles" is misleading because they are net accumulation profiles which have been affected by dissolution (including bioerosion) of carbonates either produced in situ or transported to those sites.

Is this just semantics or is there a real issue here to be addressed? Cherns and Wright (2000) and Wright et al. (2003), in studies of Silurian and early Jurassic ramp successions discussed above, were able to identify massive skewing of the faunas through the removal of diverse aragonitic components. Sanders (2001) has recorded similar effects in late Cretaceous carbonates. Unless such dissolution takes place in a closed system (see below) significant lowering of the accumulation rates must have resulted. If these case studies are not anomalous, then low energy ramp settings above the lysocline are likely to be sites of early dissolution creating not stores but sinks for carbonate production, especially of the molluscan aragonitic component. Many ancient subtidal ramp successions, especially Palaeozoic ones, preserve a very poor record of aragonitic molluscan benthos, although those dominate the silicified Silurian and Jurassic examples described above. Indeed, these effects may also be important in any low energy setting and Sanders (2001), in discussing the effects of burrow-mediated dissolution on late Cretaceous rudist banks, has also emphasized the implications of such effects when considering sediment budgets.

In conclusion, a major issue to be addressed is whether the reduced accumulation rates characterizing most lower energy, deeper ramp settings evidenced in the stratigraphic record are simply a function of reduced productivity as current modelling theories imply, or whether a significant factor is the loss of carbonate sediment, especially in situ benthic production, by early dissolution.

### Open or closed systems and was mud really mud?

The Silurian and Jurassic examples studied by Cherns and Wright (2000) and Wright et al. (2003) were associated with nodular limestones that represent calcareous, diagenetic rhythmites. Munnecke and co-workers (Munnecke and Samtleben, 1996; Munnecke et al., 1997, 2001) have proposed that the carbonate-rich units in such rhythmites are the products of the transformation and translocation of metastable carbonate mud to diagenetic carbonate "mud" (typically microspar)(Munnecke and Samtleben, 1996; Munnecke et al., 1997; Munnecke and Westphal, 2004). Their model involves microbially mediated dissolution of aragonite mud and reprecipitation as calcite microspar in a closed system. The main source of the diagenetic carbonate in their model is aragonite mud even during periods of "calcite seas" such as the Jurassic and Silurian. If, during these "calcite seas" aragonite was not produced as a precipitate, abiogenic or biogenically-induced, it must have come mainly from the breakdown of aragonitic biota. Even in today's "aragonite sea", aragonite mud appears more likely to be due to the breakdown of skeletal material than of an inorganic origin (Gischler and Zingeler, 2002), but see Yates and Robbins (1998) for another process. In the two studies provided by Cherns and Wright (2000) and Wright et al. (2003) the shallow water equivalent facies lack prominent calcareous algal remains, but a viable source for the diagenetic carbonate in offshore facies comes from dissolution of an abundant former aragonitic molluscan fauna, preserved locally by early silicification.

Thus we have two issues; firstly did the mud grade carbonate in the rhythmites first begin as aragonite mud or was it at least in part originally derived from molluscan aragonite? Secondly, the proposal presented above that early syndepositional dissolution creates sediment sinks or “black holes” in lower energy, organic-rich sediments requires an open or partly open system whereby at least some of the carbonate released by dissolution is lost in solution into the overlying water column, and not into a closed system as Munnecke et al. (2001) propose.

From our observations of rhythmite successions of Palaeozoic and Mesozoic age the remains of former aragonitic benthic faunas are typically sparse to absent even where bottom conditions appear to have been suitable. This is not to claim that hemipelagic input of mud grade aragonite may not have been important, but that it is likely that benthic aragonite sources were significant. Assessing whether carbonate mudstones are the products of hemipelagic fall-out or in situ production by benthic aragonitic faunas is not a simple task, but it is a requirement before the dynamics of ancient deposystems can be elucidated. Many limestones in limestone-marl/shale rhythmite successions contain aragonite relicts or crystal moulds (Munnecke et al., 1997), and more detailed study of such material might reveal their provenance.

The concept of a closed system can be questioned; geochemical studies would lead us to expect dissolution to be most marked in the upper, bioturbated (bio-irrigated) part of the sediment column in an open system where pore waters were readily recycled into the water column (Sanders, 2003, p.118). Evidence that dissolution was more marked in the upper parts of the sediment column in Jurassic rhythmite-bearing successions in South Wales was provided by Wright et al. (2003), who noted that the remains of infaunal molluscs were normally restricted to moulds of deeper burrowing forms (the shallow burrowing aragonitic bivalves had apparently been removed by dissolution), preserved presumably because those lived below the main dissolution levels. If preferential dissolution had been taking place in near-surface levels, where bioturbation was prevalent, the development of a closed diagenetic system becomes improbable. The volume of diagenetic carbonate, of whatever origin, is unlikely to reflect the volume of aragonitic sediment originally deposited making such settings net sinks for sediment. Geochemical analyses are needed to determine whether limestone-marl/shale systems were open or closed. As in many geological systems there is a spectrum of possibilities and it seems more likely that some of these early diagenetic dissolution systems were more or less open than others, as has been demonstrated for Bahamian slope sediments by Melim et al. (2002).

## Diagenetic potentials, missing clues and the cool-versus-warm water issue

The diagenetic potential of carbonate sediments, especially for secondary porosity formation, depends on their composition. Sediments with a higher proportion of aragonite are more susceptible to dissolution relative to those with a lower aragonite content, creating the potential for mouldic porosity and releasing carbonate for cementation. This is clear from comparisons between tropical Palaeozoic and late Cainozoic carbonates (Tucker and Wright, 1990, p. 412) or between cool water and tropical Cainozoic forms (James and Bone, 1989). Many tropical Palaeozoic carbonates appear to have had less aragonite than modern equivalents and Cainozoic cool water carbonates are generally less aragonite-rich than contemporaneous tropical forms. Some authors (e.g Dodd and Nelson, 1998) have even proposed that Cainozoic cool water carbonates are better diagenetic analogues for Palaeozoic limestones than modern tropical ones. The Silurian and Jurassic deeper ramp successions described above were originally dominated by aragonitic forms but that labile component was lost syndepositionally by dissolution. Thus the diagenetic potential was reduced prior to any significant burial such that deeper ramp sediments may have behaved, diagenetically, more like cool water types.

The slow deposition rates of some cool water carbonate deposystems seems to predispose them for early marine diagenesis (Kyser et al., 1998). Long residence times in the taphonomically active dissolution zone could also predispose such sediments to aragonite dissolution. Thus we pose a question: is the apparent reduced diagenetic potential of many cool water carbonates due to their apparent lower primary aragonite content, or because the more labile aragonite was removed during very shallow burial?

In their comparative study of tropical Mississippian and cool water Cainozoic carbonates Dodd and Nelson (1998, p. 121) note: “The reason for the scarcity of originally aragonitic fossil grains in Paleozoic rocks worldwide is unknown. Organisms with aragonitic skeletons such as some molluscan groups and calcareous green algae were present but seldom in much abundance”. However, Cherns and Wright (2000) showed former aragonitic molluscs were a major component of the Silurian shelly faunas they described, but were only revealed where very early silicification preserved them from dissolution. The possibility has to be considered that skeletal benthic faunas throughout the fossil record have been skewed by early dissolution in lower energy settings (Cherns and Wright, 2000; Sanders, 1999, 2001; Wright et al., 2003). Many of the most useful fossils in palaeoenvironmental interpretation in sedimentary facies analysis are the calcareous algae, especially the aragonitic green algae. These should be highly susceptible to early dissolution. The sedimentologist cannot fail to be struck by the abundance of such algae in modern tropical settings, and forms such as *Halimeda* have strikingly similar ancestors back to the Ordovician. Yet such grains are not always seen in such abundance in the fossil record. Is this a consequence of seawater chemistry changes restricting or favouring the calcification in such algae (Stanley and Hardie, 1998), or to the susceptibility of these grains to mechanical breakdown, to some ecological effect or to very early dissolution? Specifically, should the absence of calcareous green algae be simply interpreted as a temperature effect, leading to an interpretation of cool waters, or a depth effect (there are few palaeo-depth indicators more useful than the calcareous green algae to suggest very shallow waters), or is it simply a taphonomic effect? Sanders (2003) illustrates the effects of early dissolution on Palaeozoic green algae. We do not wish to undermine the basis for microfacies analysis but to urge more caution over the significance of the absence of some labile components in a grain assemblage.

Early, chemical dissolution of carbonate is regarded as more important in temperate waters than in tropical ones (see figures presented in Smith and Nelson, 2003) but extreme dissolutional loss may also be as effective in tropical settings. Indeed the higher rates of organic production in the latter may mean that in some settings dissolution through microbially-induced acidity may be more significant.

## CONCLUSIONS

The growing weight of evidence points to very early dissolution being a significant process affecting lower energy, organic rich carbonate deposits. This raises a series of questions for those trying to interpret ancient carbonate successions. These include:

- To what extent are many fossil benthic assemblages from low energy settings refractory, the residues of former skeletal assemblages after the selective dissolution of more labile (largely aragonitic) forms?

- To what extent is the presence or absence of useful palaeoenvironmental indicators in ancient sediments, especially those composed originally of labile aragonite such as the green algae, an ecological or taphonomic effect?

- How facies specific are these dissolution processes?

- Were, what are now, mud-grade carbonates in rhythmic limestone-marl/shale successions originally deposited from suspension as mud-grade material, or was some/much of it originally coarser grade, skeletal, benthic (molluscan) aragonite?

- Was the system in which the transformation and translocation of carbonate took place, especially in low energy settings, an open or closed one? If the former, should such depositional settings be seen as net carbonate sinks (diagenetic “black holes”)?

- To what extent are the depth-productivity profiles determined for carbonate systems, that show diminishing carbonate productivity with depth, more an artifact of selective, offshore dissolution than due to reduced benthic production, especially in deeper ramp settings?

- Could an ancient limestone appear to have the composition of a cool water type simply because the more labile aragonitic components were removed by early dissolution?

- Is the reduced diagenetic potential of many Palaeozoic and cool water carbonates really a function of the effects of early aragonite dissolution rather than due to differences in primary (depositional) composition?

- Is early dissolution just as important in tropical settings as it is in cool water ones?

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