

## Underlying issues on the selection, use and conservation of building limestone

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**Abstract:** An argument is presented that, despite popular assumptions, many limestones, especially the wide range of clastic and, in general, granular limestones, do not decay in a steady and predictable pattern in response to slow dissolution. Instead these stones, especially when used in construction in polluted environments, invariably decay episodically through physical breakdown. Most commonly this is accomplished through a variety of salt weathering mechanisms that, if unconstrained, can lead to the rapid, catastrophic decay of building blocks and their complete loss – a process that has driven the extensive programmes of stone replacement that are typical of buildings constructed of these stones. In polluted environments, especially those rich in sulphur and particulates, the most common constraint on accelerated decay has been the rapid development of gypsum crusts that, for example, could rapidly 'heal' the scars left by contour scaling. It is ironic, therefore, that any reduction in pollution could conceivably lead to increased erosion by retarding this healing process. Because of this temporal variability of decay and its translation into spatial complexity, it is important that further research is undertaken to understand controls on the decay of these important building stones so that future conservation strategies can be appropriately informed.

### On the predictability of decay and the role of solution

Limestones, especially across Europe, have arguably been the building stone of choice for many centuries, if not millennia, and continue to be used extensively in a wide range of structures in a load-bearing capacity and increasingly as cladding (Calvo & Regueiro 2010; Cassar 2010; Siegesmund *et al.* 2010). In addition, crushed stone and processed limestone forms the basis of many mortars and aggregates (Beck *et al.* 2010; Dotter 2010; Ioannou *et al.* 2010). This widespread use is a reflection of their occurrence worldwide within the geological column, the relative ease with which many can be cut and quarried (especially those that harden only on exposure), the readiness with which many can be shaped to produce ashlar blocks and their perceived durability. This perception is, in the UK context, based largely on the survival of many old limestone buildings in cities such as Oxford, Bath and London, and that are assumed to have resisted previously high levels of atmospheric pollution. It should be noted, however, that such interpretations rarely take into account the many buildings that did not survive nor the possible

extent of earlier stone replacement (Smith *et al.* 2008a).

Limestones are generally homogeneous in their chemical characteristics, being dominated by CaCO<sub>3</sub>, but can be highly variable in terms of physical characteristics such as hardness, fossil content and porosity (Smith & Viles 2006). It is these physical characteristics, especially porosity and other physical properties (e.g. Figueiredo *et al.* 2010; Rescic *et al.* 2010), that are in turn the major determinants of, for example, patterns of moisture movement (Sass & Viles 2010) and, ultimately, the durability of limestones when they are placed in buildings and exposed to often aggressive environmental conditions (Beck & Al-Mukhtar 2010). So that, for example, dense, impermeable Istrian Limestone has traditionally been used not only as a foundation stone within cities such as Venice but also as an effective barrier to rising groundwater (Simunic Bursic *et al.* 2007). Similarly, a significant number of coarse shell fragments tends to increase durability, whereas the presence of numerous very fine pores can result in a stone that is less durable than those with larger pores (Leary 1983).

Despite the structural differences between limestones, their chemical similarity has led to the

belief, at least amongst lay observers, that their decay in buildings is predominantly through gradual dissolution similar to the so-called karstic erosion of natural limestone outcrops. Karst processes are, as identified by Livingston (1992), those where rainfall acidified with CO<sub>2</sub> is the major agent of deterioration producing dissolution. In the case of natural exposures there is also the general assumption that the rock weathers at a near-constant rate to produce overall surface lowering and distinctive solutional relief forms (such as rillenkarren). Livingston acknowledges, however, that in polluted environments the effects of 'natural' dissolution are enhanced where atmospheric moisture acidified through the presence of sulphur and nitrogen oxides. These react with calcium carbonate to produce, for example, the more soluble calcium sulphate, which can then be removed in solution. In addition, dry deposition can also occur where sulphur and nitrogen oxides in the atmosphere react, under moist conditions, with calcium carbonate to form calcium sulphate – which can accumulate as a gypsum crust. This last process is often associated with the surface deposition of complex mixtures of atmospheric particulates derived from the combustion of fossil fuels and a range of environmental dusts, including marine aerosols, that are themselves rich in carbonates, and a variety of salts. These particulates can themselves be modified by surface reactions with and between acidified atmospheres and the underlying stone to produce secondary salts or simply be included within gypsum crusts as they form to produce their characteristic grey–black tone (McAlister *et al.* 2006, 2008).

As well as the physico-chemical processes described above, limestones are, like any exposed stone, subject to a wide range of additional weathering regimes. This includes those associated with deteriorative biofilms found under many environmental conditions (Miller *et al.* 2010), freeze–thaw processes that are important in, for example, many central European locations, and salt weathering (Espinosa-Marzal & Scherer 2010) that is common not just in urban areas, but also in maritime and arid environments characterized by alternate wetting and drying cycles and an adequate supply of salts (Smith & Viles 2006). From this Smith & Viles (2006, p. 192) concluded that:

limestone deterioration can be more accurately conceived as the result of often complex interactions between many different process regimes. These regimes may be spatially patchy – for example, acid rain processes may dominate one part of a building, whilst other areas may be more sheltered and prone to dry depositional effects. Another complexity is introduced by the fact that, as pollution and climatic

conditions have changed, many building stones have been exposed to changing process regimes over their lifetime.

### Alternative patterns of decay

The idea that environmental conditions, pollution regimes, and, hence, patterns and rates of weathering and erosion may change over the lifetime of a structure is fundamental to understanding present and future rates of decay. Not just because new processes may come to bear, but because stone carries with it a stress history or 'memory' of past conditions that can make it more or less susceptible to new processes and process combinations. This means that, in general, complex histories are likely to elicit complex future responses, each of them unique not just to a building, but to the micro-environments found on it and even down to the way that previous conditions have been factored through the specific properties of individual stone blocks (McCabe *et al.* 2007). In the case of limestones, however, it would appear that in many instances there is a belief, translated largely from studies of natural limestone outcrops, that the rate of surface erosion of limestones is gradual and uniform – both spatially and temporally. This has had a major influence on perceptions regarding the nature of the threat that decay poses to buildings, and strongly influenced assumptions underpinning many early scientific investigations into the nature of limestone decay and calculations of erosion rates. Because of this 'uniformitarian' approach it was, for example, assumed that long-term erosion could be readily extrapolated either from the results of downscaled laboratory simulations or from short-term measurements of erosion loss from a limited number of key sites on a building.

Subsequent understanding of the temporal and spatial variability of decay systems has called into question these assumptions, such that Trudgill & Viles (1998) asserted that whilst chemical weathering relevant laboratory rates may be used to predict the order of magnitude of field rates, they cannot be used to ascertain the precise rate. They were even more concerned for physical and biological weathering, and were of the view that 'we are very far from having representative, replicable rate measurements from either field or laboratory studies' (p. 339). Central to their arguments were a series of field measurements and trials, especially repeat micro-erosion measurements of the Portland Limestone of St Paul's Cathedral in London that indicated considerable temporal and spatial variability (Trudgill *et al.* 1989). These showed that erosion rates can differ significantly from those predicted from a theoretical analysis of potential solution related to rainfall patterns and

chemistry. The key to this variability is, they consider, the particular importance of the residence time of water on, and within, the pores of stone in controlling solution rates that is, in turn, controlled by a range of micro-environmental conditions and localized material properties.

This latter point is extremely important, in that when seeking to transfer theories and observations on karstic weathering processes and characteristics to limestone buildings it must be remembered that karstic limestones are always dense, hard and, apart from a well-developed macro-joint system, effectively impermeable. Apart from exceptions such as the aforementioned Istrian Limestone, the majority of building limestones do not fall within this category. Instead, the most widely used limestones are likely to be those that are more easily worked and have not experienced intense deep diagenesis processes. As Leary (1983) observed, these are also likely to be less crystalline, less dense and, therefore, less resilient to many decay processes than many geologically older limestones. Numbered amongst these commonly used stones are a wide range of largely granular limestones that include numerous subvarieties of bioclastic limestones and, in particular, oolitic limestones. Of all the limestones commonly used in construction within the British Isles (and widely across Europe, e.g. Török 2002) it is oolitic limestones that exhibit the greatest variability. By definition they consist primarily of small rounded grains coated with  $\text{CaCO}_3$  and embedded in calcitic cement, but their durability varies hugely in response to differences in characteristics such as porosity and bioclasts content (Smith & Viles 2006). These characteristics determine a range of secondary properties that include permeability, saturation coefficient and water absorption, that in turn influence the operation of a range of stone decay mechanisms – especially salt weathering (Goudie & Viles 1997).

It is the general susceptibility of oolitic and other granular limestones to physical weathering, especially salt weathering, that sets them apart from dense limestones and from the general preconceptions regarding the pre-eminence of solution loss. Because of this it is more common to see these limestones affected by patterns of decay that one normally associates with sandstones rather than with the surface dissolution of limestone. Although such decay may begin with limited surface loss through granular disaggregation, decay eventually begins to concentrate spatially leading to localized accelerated retreat forming characteristic alveolar weathering or ‘honeycombing’ of the stone (Fig. 1). Once retreat has been initiated small hollows can amalgamate to form larger cavernous hollows or Tafoni (Fig. 2). Once



**Fig. 1.** Honeycomb weathering of *Globigerina* limestone, Malta, controlled by patterns of fossil bioturbation (approximate image width 70 cm).



**Fig. 2.** Cavernous hollows developing on a Quaternary calcarenite, Mallorca (approximate image width 40 cm).



**Fig. 3.** Catastrophic decay of an individual block of *Globigerina* limestone, Malta (approximate image width 2 m).



**Fig. 4.** Catastrophic decay of a section of wall constructed of Jurassic oolitic limestone, Oxford (approximate image width 4 m).

small-scale cavernous hollows are initiated in oolitic limestones, like those used in cities such as Oxford, there is ample visual evidence that individual blocks (Fig. 3) and, ultimately, whole sections of wall (Fig. 4) can become prone to rapid and, eventually, catastrophic decay. Invariably this is associated with the near-surface presence of salts (e.g. Espinoza-Marzal & Scherer 2010), such as gypsum within the cavernous hollows, and is achieved by a combination of multiple flaking and, especially, granular disaggregation.

### Understanding catastrophic decay

Understanding the causes of catastrophic decay is clearly central to understanding the long-term performance of many granular limestones. Likewise, it is apparent that catastrophic decay can only be controlled if the reasons why rapid retreat is triggered in the first place and its continuation is facilitated are understood, and the causes avoided or removed. This is particularly true where inappropriate conservation can accelerate decay, and where choices have to be made regarding stone selection for new build and stone replacement on older structures. To achieve this understanding four questions need to be asked.

- What processes or combination of mechanisms are responsible for rapid retreat?
- What physical, chemical and mineralogical characteristics determine stone susceptibility to rapid retreat, and how do these properties change during decay?
- How do microclimatic conditions at and beneath the stone surface change as stone retreats, and how do these influence decay mechanisms?
- What permits continued weathering despite rapid loss of weathered material in which, for example, salts are concentrated?

Typically, limestones prone to catastrophic decay are clastic or granular and often oolitic in character, but differ from other clastic stone types, such as sandstones, in that they are initially prone to both chemical attack and salt-induced decay as well as severe and/or prolonged freezing. The balance of these processes inevitably changes as decay continues and the retreat of individual limestone blocks progressively shelters them from direct rainfall, but leaves them subject to wetting from dew and frost. Such limestones have been widely used as a building material in England, for example, in many of the large cathedrals (e.g. Wells and Lincoln), historic buildings in Bath and Oxford, and are found in many ecclesiastical ruins. They are diverse in nature, exhibiting a range of petrological fabrics, geochemical characteristics and durabilities (Leary 1983), and often decay through interlinked chemical, physical and biological processes often exacerbated by the presence of atmospheric pollutants.

The first step towards understanding long-term performance is knowledge of what controls the initiation of surface irregularities within which, for example, salts can accumulate. These may originate through slight variations in surface topography produced during the initial working of the stone, but a more general source of spatially concentrated decay may lie in structural and textural variations. In laboratory simulations of salt weathering carried out on oolitic limestones Smith *et al.* (2008*b*) identified meso-scale structural diversity, such as that associated with distinctive bedding (Fig. 5), fossil bioturbation (Fig. 1) and texturally different inclusions such as clay lenses, as potential focal points for localized decay. In highly polluted environments, however, sulphation (the chemical reaction of  $\text{CaCO}_3$  with water acidified by  $\text{SO}_2$ ) can be so rapid that it can quickly mask any surface heterogeneity through the development of a surface gypsum crust, especially in areas sheltered from large amounts of surface runoff. In these areas gypsum crusts can act as a hard, relatively impermeable surface, but once breached they may fail



**Fig. 5.** Decay of a Jurassic oolitic limestone initiated along clay partings within the stone, Oxford (approximate image width 1 m).

catastrophically leading to the production of blisters and scales.

A large body of work has focused on identifying the nature and origin of blackened gypsum crusts on limestone and other calcareous building materials (Sabbioni 1995; Maravelaki-Kalaitzaki & Biscontin 1999; Jimenez de Haro *et al.* 2004; Sabbioni *et al.* 2004). Work by Rodriguez-Navarro & Sebastian (1996), Cultrone *et al.* (2004) and Gomez-Heras *et al.* (2008) has illustrated experimentally the role of particulate matter in facilitating gypsum formation on limestone. Biofilms and other rock-dwelling organisms can also encourage decay (Schiavon 2002), although some communities may have a more bioprotective role (Carter & Viles 2004). Once initially protective crusts and/or biofilms are breached, salts arising from air pollution, groundwater contamination, road de-icing activities and other sources contribute significantly to limestone decay, producing flaking, disintegration and alveolar forms (Goudie & Viles 1997; Fitzner *et al.* 2002; Török 2004).

A possible explanation for rapid decay is that once weathering initiates a hollow on the stone surface, accumulated salts are less likely to be washed away by rainfall and reduced moisture availability might enhance salt concentration near the stone surface. However, this does not explain why, with reduced wash-in and near-surface concentration, any salts present are not rapidly lost together with the debris. Neither does such a micro-environmental model explain why only certain stones on an otherwise uniform façade experience rapid, catastrophic retreat. To explain these requires not just an examination of microclimatic conditions, but also subtle variations in the physical characteristics of the stone and the operation of specific

decay mechanisms controlled by, for example, available salt types and the balance between salt input, output and storage. One possibility identified by Smith *et al.* (2002) working on the rapid decay of quartz sandstones in a polluted maritime environment is that the presence of a store of 'deep salts' within stone blocks accumulated over a long period could maintain decay as blocks retreated and tapped into it. Initially this was aided by the presence of the core softening of the stone behind a surface accumulation of gypsum. This phenomenon has been widely observed on oolitic limestones in cities such as Oxford, where rapid retreat has also been associated with areas where dry deposition or mixed process regimes dominate, and where salt deposition and surface formation can continue even in cavernous hollows sheltered from wet deposition of salts and pollutants (Smith & Viles 2006).

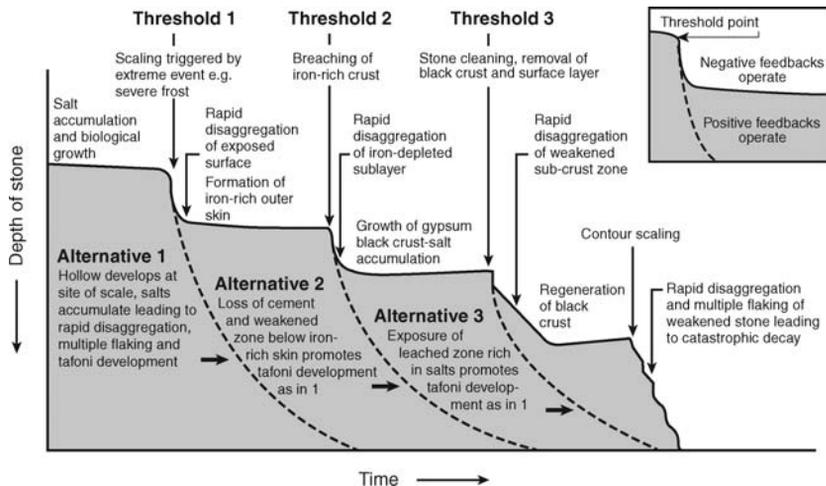
Similar studies to those in Oxford have been carried out by Török *et al.* (2007) in Budapest. Here, they studied ashlar blocks composed of three porous Miocene limestones (a fine-grained limestone, a medium-grained oolitic limestone and a coarse-grained biogranular limestone) from buildings in the city and compared them with quarry blocks of the same lithologies. On the urban stones the most common weathering forms are white and black crusts. Apart from their impact on the visual and thermal properties of stone (Searle & Mitchell 2010; Thornbush 2010), the presence of the surface crusts, together with calcite recrystallization, is also marked by pore occlusion and reduction in microporosity, which was documented using a combination of microdrilling resistance and ultrasonic pulse velocities. These tests also showed degradation of the underlying fine- and medium-grained limestones, with drilling resistances less than those of equivalent quarry stones. Where crusts were detached from this underlying weakened zone the detachment was initiated by the opening up of microfissures that develop below the cemented crust zones. The fine-grained limestone appeared to be less durable than the coarse-grained variety, and more prone to rapid crust formation and detachment. In this particular case, microbiological activity did not appear to play a significant role in crust formation and removal, whereas the latter is probably influenced by freeze-thaw weathering and strongly controlled by the texture and porosity of the limestone substrate.

### The 'switching off' of rapid decay

Studies in the city of Oxford have confirmed the episodic nature of blistering on historic limestone

walls, its patchiness over wall surfaces, and its complex relation to salt, climatic conditions and pollutant content (Viles 1993; Antill & Viles 1999). Archival investigations of blister development, climate and air pollution over the period 1864–1937 in central Oxford have also indicated particular ‘spasms’ of blistering (which cannot easily be related to any specific environmental variable) followed by relative stability. Such episodic and complex production of catastrophic decay features poses a severe problem for the conservation and management of historic buildings and monuments, as well as presenting difficult challenges when choosing potential stabilization procedures or replacement stone types. Studies of the rapid decay of non-calcareous stones may again provide some insight into the nature of this decay and what controls its operation. This is because, in the absence of slow and gradual decay in response to surface dissolution, a more appropriate paradigm for the decay of stones such as quartz sandstones is one based on periods of relative surface stability followed by short-lived episodes of rapid surface loss (Smith *et al.* 1994, 2003). The first episode of rapid loss may very well be associated with the sudden loss of a surface gypsum crust and the rapid erosion of a weakened subsurface layer. After this is removed, the stone appears to be faced by one of two choices, either negative feedbacks come into play or, as described above, the initiation of a localized hollow can trigger a number of positive feedbacks that accelerate decay leading to catastrophic loss (Fig. 6).

The processes by which a scaled surface could be stabilized are numerous. In the case of iron-rich sandstone, for example, surface induration could result from the outward migration of iron to form a surface crust, whereas in a limestone it could take the form of a case-hardened layer formed by the precipitation of calcium carbonate which either migrated outwards or flowed in solution over the surface. The most obvious source, however, of surface stabilization is, as indicated above, the growth of a new gypsum crust. In Oxford, for example, there is widespread observational evidence of the multiple ‘healing over’ of oolitic limestones following contour scaling through the apparently rapid regrowth of black gypsum crusts (Fig. 7). Similar patterns of multiple black crusts have also been observed on the oolitic limestone of St Matthias Church, which sits above the still polluted city of Budapest (Smith *et al.* 2003). Regrowth of gypsum crusts on the church is associated with the rapid deposition of gypsum, dust and surface biological colonization by fungal hyphae that help to trap and bind the dust to the surface. As indicated earlier, the presence of the dust appears to catalyse the further precipitation of gypsum in the form of a new, stabilizing surface crust. The importance of dust deposition in a dry environment was highlighted by a comparative analysis of the Hungarian Parliament building that sits below the church on the banks of the Danube. Here, the same stone is exposed to a considerably moister atmosphere and contour scaling appears to be followed more often than not by rapid retreat rather than recrusting.



**Fig. 6.** Hypothetical decay pathways for quartz sandstone used as a building material in a polluted environment. The inset identifies feedback options at critical thresholds of decay associated with rapid surface loss of material. Adapted from Smith *et al.* (1994) and Smith (2003).



**Fig. 7.** Multiple gypsum crusts on Jurassic oolitic limestone, Oxford (approximate image width 50 cm).

### Summary and conclusions

There is a general preconception, at least amongst lay observers, that limestones used in construction typically decay in a slow, uniform and largely predictable fashion primarily in response to surface dissolution. It is accepted that the rate of dissolution can be accelerated by, for example, an increase in rainfall acidity associated with atmospheric pollution, but the underlying assumption of uniformitarian change still persists. This assumption draws further support from the behaviour in natural outcrops of dense hard limestones that exhibit a distinctive karstic behaviour associated with the formation of persistent surface solutional forms. It can be readily demonstrated, however, that the decay of many limestones used in construction is far from being uniform and predictable. Instead, it has been demonstrated that in many cases decay is characterized by marked temporal and spatial variability, both within individual blocks and often across complete facades. This particularly applies to granular limestones (e.g. bioclastic and oolitic limestones) that are frequently characterized by effective physical breakdown similar to that normally associated with, for example, sandstones. This includes the presence of phenomena such as initial surface roughening, leading to pitting and honeycombing,

leading to cavernous weathering, leading in some cases to the rapid, catastrophic decay and complete loss of individual blocks. This is commonly accomplished by mechanisms such as granular disintegration, contour scaling and multiple flaking, but in polluted environments may also be linked to the rapid formation of surface crusts composed primarily of gypsum. The formation of such crusts indicates the significance of dry deposition in the weathering of urban limestones, including surface sulphation and particulate deposition, but also emphasizes the role of salts such as gypsum in driving the various decay mechanisms.

Spatial differentiation of decay at the level of individual blocks may initially be controlled by small-scale variability in key rock properties, most notably the degree of induration, texture and associated porosity/permeability. At the facade scale, however, localized environmental controls on processes such as surface soiling and crust formation may assume a greater significance (Turkington & Smith 2004). In temporal terms, initial gypsum crust formation can stabilize a surface, but over their lifetime such crusts can be associated with subsurface weakening and act as a sources of salts that can eventually penetrate deeply into the underlying stone. As a consequence, if the crust is breached in any way this is often followed by rapid erosion as pre-weathered subsurface material is lost and 'deep salts' exploited. That all such stones (or, indeed, buildings) do not quickly disappear is testimony in many cases to previous and ongoing campaigns of stone replacement by those with a duty of care (Fig. 8), but also to the fact that it is possible for stones to stabilize through the 'switching off' of the positive feedbacks that accelerate.

In the case of cities like Oxford, which were once heavily polluted and in the still polluted city of



**Fig. 8.** Replacement of a section of wall in Oxford previously destroyed by catastrophic decay of Jurassic oolitic limestone (approximate image width 6 m).

Budapest, it has been shown that stabilization was and is commonly associated with rapid surface sulphation and growth of a new gypsum crust that effectively ‘heals’ the erosion scars left by surface delamination. During the lifetime of a block this process might be repeated several times, and across a facade one might expect to see a patchwork of stones at different stages of the cycle. This includes the final stages of decay whereby, for whatever combination of circumstances, the rate of regrowth cannot keep pace with the accelerating rate of surface loss and the block (or by this time possibly a collection of adjacent blocks) rapidly disappears. This could be part of a natural progression, in that repeated cycles of scaling and crust development may eventually create a hollow that is deep enough to create a micro-environment that is particularly conducive to, for example, salt weathering. Conversely, there is scope for the potentially ironic situation in which high levels of atmospheric pollution, especially dust in the atmosphere that can catalyse crust formation, may have contributed significantly to the long-term stability of stones that are intrinsically susceptible to processes such as salt weathering, and which can perform relatively badly in durability assessments, such as the standard sodium sulphate crystallization test, used to specify stone for construction.

It is doubly ironic, therefore, that strategies to reduce atmospheric pollution, especially the reduction of sulphur and particulates, could in future curtail the ‘healing’ process, and that historic stone – often pre-weathered and salt infused over many centuries – might disappear at an accelerated rate following contour scaling. Furthermore, it is possible to envisage that the delicate balance that prevents the triggering of rapid decay could be upset by a number of other interventions and changes. Aggressive removal of black crusts, especially in environments that no longer facilitate their rapid regrowth, could, for example, trigger rapid surface loss. Alternatively, any change in environmental conditions that affected patterns of heating–cooling and wetting–drying, perhaps linked to wider climatic change, might be expected to influence crust stability and/or growth as well as the operation of existing decay processes such as salt weathering, and may even promote new ones.

It is in the face of such complexity and uncertainty, and the demonstrable need for conservation (e.g. Buj *et al.* 2010; Figueiredo *et al.* 2010; Pápay & Török 2010; Ruiz-Agudo & Rodriguez-Navarro 2010; Stefanidou 2010; Vazquez-Calvo *et al.* 2010), that the idea for the original workshop on granular limestones, on which this volume is based, was conceived. This was on the basis that not only do we need to understand the nature and causes of their decay more fully, but also that we

must begin to explore how this improved understanding can be used to inform future conservation strategies.

Support for this paper was provided by EPSRC research grant EP/D008603/1.

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