Deterioration-related changes in physical properties and mineralogy of limestone monuments

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Abstract: Many public buildings were constructed from limestone in Budapest. The deterioration and the changes in physical and mineralogical properties of two texturally and physically different limestone types have been compared. The first type, that is used on the facades of monuments, is a porous limestone (oolitic) with an effective porosity of up to 36% and having a very low compressive strength (2-11 MPa). The second type is a freshwater limestone (travertine) with effective porosity of generally less than 10% and with a compressive strength of up to 120 MPa.

These textural differences and the variable physical properties are well expressed in the differences of decay phenomena. Schmidt hammer rebound, Duroscope rebound, water absorption and mineralogical composition (XRD, DTA-DTG) were measured and compared.

The porous limestone shows intense black and white crust formation. The strength of the crust is related to the thickness, to the mineral composition, to the crust morphology and to the capacity of water absorption. Calcite-rich, impermeable, thick white crusts have the highest strength, which is followed by gypsum-rich laminar black crusts alongside with thin gypsum-rich white crusts. The crusts are prone to scaling and/or flaking that is followed by rapid material loss and back-weathering. During the removal of the crust not only does the surface with its gypsum-rich crust chip off but also a detachment layer with the substrate. As a consequence, significant loss in volume and decrease in surface strength are attributed to urban air pollution. The freshwater limestone does not show similar decay; only blackened crusts are formed on the limestone surface. These crusts are apparently stable and they do not tend to scale with the carbonate substrate. Decrease in surface strength is minor. Thus, within the polluted urban environment not only aesthetic problems arise when one looks at the blackened limestone facades.

Résumé: Beaucoup de bâtiments publics ont été construits de la pierre à chaux à Budapest. La détérioration et les changements des propriétés physiques et minéralogiques de deux les types texturalement et physiquement différents de pierre à chaux ont été comparés. Le premier type, celui est employé sur les façades des monuments, est une pierre à chaux poreuse (oolitic) avec une porosité efficace jusqu'à de 36% et avoir une résistance à la pression très basse (2-11 MPa). Le deuxième type est une pierre à chaux d'eau douce (travertin) avec la porosité efficace généralement moins de de 10% et avec une résistance à la pression jusqu'à de 120 MPa.

Ces différences de texture et les propriétés physiques variables sont bien exprimées en différences des phénomènes d'affaiblissement. Le rebond de marteau de Schmidt, le rebond de Duroscope, l'absorption de l'eau et la composition minéralogique (XRD, DTA-DTG) ont été mesurés et compares.

La pierre à chaux poreuse montre la formation noire et blanche intense de croûte. La force de la croûte est liée à l'épaisseur, à la composition minérale, à la morphologie de croûte et à la capacité d'absorption de l'eau. les croûtes blanches Calcite-riches, imperméables, épaisses ont la plus haute résistance, qui est suivie des croûtes noires laminaires gypse-riches bord à bord avec les croûtes blanches gypse-riches minces. Les croûtes sont enclines à la graduation et/ou l'écaillement cela est suivi de la perte matérielle rapide et du back-désagrégation. Pendant le déplacement de la croûte non seulement la surface avec son morceau gypse-riche de croûte outre de mais également une couche de détachement avec le substrat. Par conséquent, la perte significative en volume et la diminution de la force extérieure sont attribuées à la pollution atmosphérique urbaine. La pierre à chaux d'eau douce ne montre pas l'affaiblissement semblable ; seulement des croûtes noircies sont formées sur la surface de pierre à chaux. Ces croûtes sont apparemment stables et elles ne tendent pas à mesurer avec le substrat de carbonate. Diminuez dans la force extérieure est mineur. Ainsi, dans l'environnement urbain pollué non seulement les problèmes esthétiques surgissent quand on regarde les façades noircies de pierre à chaux.

Keywords: Durability, limestone, pollution, strength, weathering

INTRODUCTION

Limestone buildings located in polluted urban environments show significant signs of decay. The rate of decay depends on the textural/fabric properties of the carbonates, on the pollution fluxes and on the environmental setting (air pollution, meteorological and micro-climatic conditions). The most common form of weathering on limestone is the development of gypsum crusts. The influence of environmental conditions on gypsum crust formation has been thoroughly studied in the field (Amoroso & Fassina 1983) and under laboratory conditions (Rodriguez-Navarro & Sebastian 1996, Ausset et al. 1999, Primerano et al. 2000, Cultrone et al. 2004). Differences in weathering of different limestones exposed to the same pollution regime have been studied using small test blocks over periods of time (the "scale problem" cf. Smith 1996). Research generally focuses on the description of processes and decay products of one type of limestone. It has been also emphasised that surface properties play a key role in pollution entrapment and various crust formation (Amoroso & Fassina 1983, Zappia et al. 1998, Grossi et al. 2003). Fewer studies are available dealing with the physical changes that are triggered by pollution fluxes. Examples are known for various lithologies, such as limestones (Christaras 1991a, 1991b, Török 2002, 2003, Török et al. 2004) marbles (Christaras 1991a, 1996), granites (Irfan & Dearman 1978, Kahraman 2001) and rhyolite tuffs (Topal & Sözmen 2003, Török et al. 2005).

The aim of this paper is to compare the urban decay processes of two different limestones, both of which have been extensively used as building stones in Budapest in the 19th and early 20th century.

A soft ooidal limestone of Miocene age and a freshwater limestone (travertine) of Pleistocene age were analysed. The paper summarises the results of the study of several selected buildings, giving emphasis on the cause and effect relations using mineralogical analyses under laboratory conditions and strength parameters obtained by non-destructive on site tests, such as Schmidt hammer and Duroscope.

METHODS

On site tests included the description of weathering features by using the nomenclature of Smith *et al.* (1992) and Fitzner *et al.* (1995). Samples were taken form various weathering features for laboratory analyses. At selected ashlars the strength properties of stone surfaces were tested by using Schmidt hammer and Duroscope techniques. On each tested block 10 measurements were made. These tests provide information on mechanical properties of rocks giving a rebound value which correlates approximately with the strength properties of the rock. The water absorption of weathering crust and host rocks were detected by using Karsten tube.

In the laboratory, samples were analysed using X-ray Diffraction (XRD) and differential thermoanalysis (DTA) for the determination of mineralogical composition. The XRD analyses were carried out using a Phillips Diffractometer (PW 1130 generator, PW 1050 goniometer, Cu anode and monochromator). The powdered samples (size fraction less than 63 microns) were analysed at 40kV, 20mA. For the data collection and data evaluation a PCD-APD software package was used. Derivatograph analyses (thermal analyses) were carried out to determine the clay composition, gypsum content and organic matter content of the samples. The samples analysed by XRD were used in this test. The test apparatus was a MOM Derivatograph. Between 400-600 mg of powdered samples were heated at 10°C/min with the analyses carried out between 20-1000°C. The thermic gravimetry sensitivity was 100-200 mg.

Thin sections were also prepared from samples to allow comparison of textural and mineralogical differences between the outer altered surface and subsurface material.

PROPERTIES OF POROUS LIMESTONE AND TRAVERTINE

Lithology

Oolitic limestone

The soft and porous limestone of Miocene age is of marine origin. It has a yellowish-white colour when it is freshly quarried. The main mineral is calcite (92-97%) but minor amounts of quartz and sand-sized lithic clasts are also present. The texture is characterised by the presence of well to moderately rounded micro-oncoids of 0.2-1.0mm in diameter (its commercial name is ooidal limestone of Sóskút). The ooids are surrounded by circumgranular calcite cement. Grain to grain contacts also occur often associated with thin cement rims. Besides ooids, red algae fragments, gastropods, bivalves and foraminifera occur in the rock. Porosity is high (up to 33%), and mainly related to intergranular pores which are between 0.1 to 1mm in diameter (Figure 1). Intragranular pores in the foraminifera or within the ooids also occur. The texture of this limestone shows some variety in the size of ooids and in the amount of other particles, but mainly belongs to ooid grainstone.

Several textural varieties occur but two of them are predominant. The first is characterised by the presence of gastropods (Cerithium type) and usually by coarser ooids (nearly 1mm). The textural inhomogeneity is also reflected in the physical properties. The second type is the ”pure” oolitic type, which has very few other carbonate grains than ooids. The size of ooids is variable but on average is finer than in the first type. The most common sedimentary feature of this limestone is the cross bedding, which is often visible on building blocks. It is a well cemented and relatively compact type whose durability is relatively good.

The quarries of the oolitic limestone still exist but most of them were operated during the second half of the 19th century when most of the public buildings were built in Budapest. The only active quarry is found app. 30 km to the West from Budapest.

Freshwater limestone (travertine)

The travertine was formed in thermal springs during the Pleistocene in a variety of sedimentary environments and as a result various types exist. The strongly recrystallised and cemented types have been used for buildings. Although there is a great variety of lithology most building blocks can be grouped into two broad lithotypes. In all lithologies calcite is the dominant mineral and very often no other minerals are found by using XRD, thus travertine is almost exclusively composed of calcite. Traces of limonite and clay minerals were also found in very few samples. The most common types of travertines are creamy in colour but due to weathering it tends to became white on exposed facades.

The first type is laminated, with large, irregular pores (Figure 1) that are parallel to bedding and often contain large calcified plant fragments. The laminated stromatolithic appearance is related to microbial encrustations. Phytoclast, calcified reeds or shrubs can be quite common. In thin section its microfacies comprise phytoclastic or phytohermal boundstone and stromatolitic phytohermal boundstone. This laminated type of travertine has a complex network of microscopic intergranular and intragranular pores with effective porosities up to 11%.

The second type of freshwater limestone contains smaller but irregularly distributed pores. Gastropods, smaller fragments of calcified reeds or oncoids comprise the main carbonate grains, but peloidal-micritic textures are also common. A massive, very strongly cemented micritic to micro-crystalline freshwater limestone also belongs to this group. Under the microscope a great variety of textures and microfacies types are identifiable including i) peloidal or bioclastic wackestone, ii) peloidal, oncoidal packstone, and iii) intraclast or phytoclast floatstone. These types of freshwater limestones have a lower porosity of between 2-6%. Many of the pores are filled with calcite mosaic cements.

The travertine was quarried in several sites in and near Budapest in the 19th century. It was mostly used in load bearing structures and for early reconstruction works of public buildings in the 19th century. Currently two active quarries are known; one is very close to Budapest (Budakalász), while the other one is located app. 60 km to NW in Gerecse Mts. (Süttő).

Figure 1.Porous oolitic limestone (left) and travertine (right)

Physical properties

Oolitic limestone

The oolitic limestone is lighter than travertine with bulk density of 1800 kg/m^3 to 2000 kg/m^3. The pore system is composed of small but well connected pores. The macro pores are prevailing with pore sizes of 8-10 microns. As a result the effective porosity of coarse limestone is in the order of 20-36 % (w). The rapid water uptake is demonstrated by the water adsorption curve and by the increase in density up to 2300 kg/m^3. The low uniaxial compressive strength (2-11 MPa) is related to the soft “spongy” texture and weak cement between particles.

Travertine

The bulk density of travertine is 2100 to 2400 kg/m^3. By water absorption it can reach 2580kg/m^3. Its effective porosity is mainly related to the elongated bedding parallel pores and is in the order of 2-11 % (w). The micro pores dominant in travertine while macro-pores has a smaller contribution to effective porosity. The compressive strength of travertine are in the range of 35 MPa to 120 MPa with an average of 50 to 70 MPa. This variety is related to the textural inhomogenities. The compressive strength does not show a significant decrease by water saturation (water saturated compressive strength can be as high as 50 MPa).

DETERIORATION RELATED CHANGES IN PROPERTIES

Weathering crust formation

The oolitic limestone and travertine show several types of decay phenomena. Mechanical breakdown, alteration and deposition were identified among others. The most frequent stone decay feature is crust formation. Crusts can be classified according to their colours and morphology. Dark coloured crust such as laminar black crust, globular black crust and dust crust develop on both stone types. White weathering crust is only found on oolitic limestone.

Dark coloured crusts

Two black crust types were documented: framboidal black crusts and laminar black crusts. Framboidal black crust evolves on protected parts of walls, generally below cornices or ornaments. Similar black crust morphology is also known as dendritic black crust (Camuffo 1995; Maravelaki-Kalaitzaki & Biscontin 1999) or as ropey (‘bubble-shaped’) crust (Antill & Viles 1999). Large surfaces are also covered by framboidal crusts especially on sheltered ashlars which are not exposed to direct rain wash. Framboidal black crusts are the thickest dark-coloured decay features with a maximum thickness of approximately 2 cm (Figure 2). These crusts are relatively stable and crust detachment is not common.

Figure 2. Black framboidal weathering crust on travertine

The main mineral of black framboidal crust is gypsum, while calcite and accessory minerals such as clay and quartz have a small contribution to the composition of the crust (Figure 3). Rarely blistering or the loss of the entire crust is observed on oolitic limestone surfaces (Török 2002). It seems that such crusts are firmly adhered to the travertine surfaces.

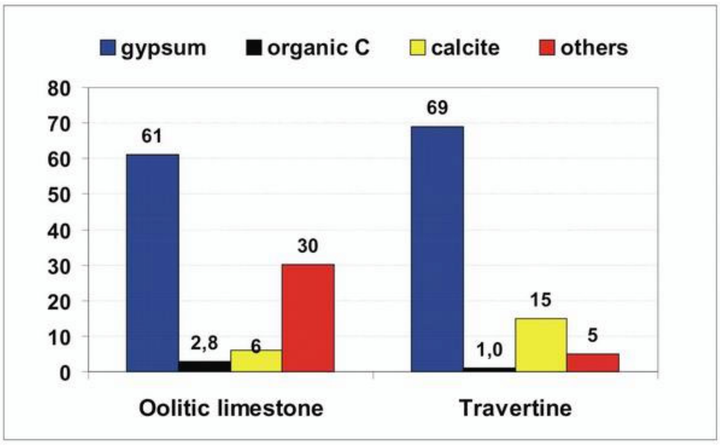

Figure 3. Mineralogical composition of black weathering crust

Thin laminar black crusts are found on vertical walls and surfaces. Similar features are described for Istria stone as "grey black areas with thin deposits" (Maravelaki-Kalaitzaki & Biscontin 1999) or as "dark-coloured crust tracing the surface" (Fitzner et al. 1995). The crusts form parallel to the surface and appear to be stable for long periods of time and they develop mostly on travertine (Török 2004).

Thick laminar black crust is formed on porous oolitic limestone and on travertine. The crust on oolitic limestone is generally less permeable than that of the host rock below (Figure 4). The crust very often incorporates the substrate. It can be considered a thicker variety of thin laminar black crust with the difference that it also incorporates the limestone surface. Gypsum and other salt related mechanical breakdown of such crusts are mostly observed on oolitic limestone. In such cases parts of the crust is uplifted and scaling or blistering takes place (Török 2002) and finally the crust detaches.

Grey dust forms an approximately millimetre-thick, or in some cases a centimetre-thick unconsolidated layer on the stone surface that can be removed without affecting the underlying stone surface. It is found primarily on sheltered and dry stone surfaces in the city centre. The Grey dust layer is very rich in organic carbon (8.1%) and in other minerals (59%; mostly quartz). The average gypsum content is also relatively high (28%) (Török 2002).

Light coloured crusts

The light coloured crusts are found mostly on oolitic limestone. Thick hard white crust and thin white crusts are the most common forms. The thick case hardened crust is found on wind/rain exposed surfaces exclusively on oolitic limestone. The crust ranges in thickness from a few millimetres to a centimetre on the stone surface and partly incorporates the substrate (Figure 5).

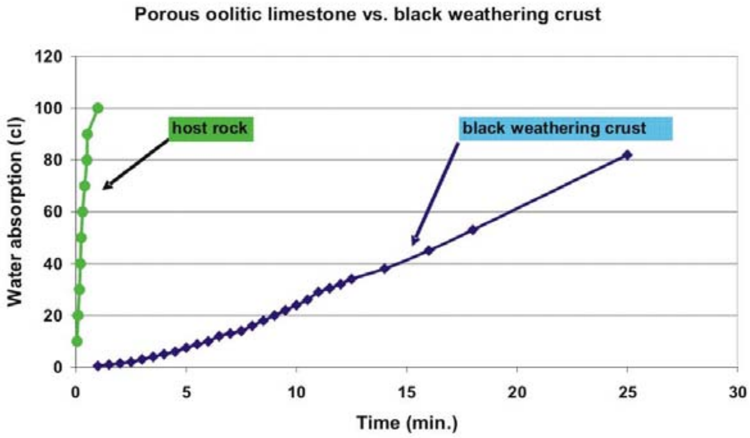

Figure 4. Water absorption difference in host rock and black weathering crust on oolitic limestone

Figure 5. White scaling weathering crust on oolitic limestone

The crust formation leads to an increase in surface strength within the crust zone and a concurrent decrease in strength below the crust at the host rock (Figure 6). The outer surface of the white crust is generally smooth while the underside is often irregular. Case hardened crusts very often show contour scaling and by detachment of the crust an irregular surface is exposed and granular disintegration begins (Török 2002).

Thin white crust is a typical feature on exposed vertical to subvertical ashlars of very fine grained oolitic limestones. It is thin, never thicker than 1-2 mm, and “fragile”. Similarly to the thick white crust the thin variety has a relatively smooth outer surface, but minor surface irregularities have also been observed. It can have a pale greyish colour, which is due to their inclusion of organic carbon. Due to differences in thickness of white crusts the mechanical breakdown of thin crust differs from thick white crust. Blistering and flaking are the typical weathering features of such crusts (Török 2005). After multiple flaking a new crust layer can develop and thus multiple crusts (primary to tertiary) develop (Smith et al. 2003).

Mechanical breakdown

Crust removal features such as contour scaling, blistering and flaking are very common but do not occur equally on all limestone and are mostly limited to oolitic limestone. Thick, white case hardened crusts of oolitic limestone show contour scaling, thinner white crusts are characterised by multiple flaking, while very thin white crusts tend to blister. Surface parallel laminar black crusts also form blisters or scales, while thicker framboidal black crusts scale with the detachment of the entire crust, rather than flaking. When there are no crusts, or after crust removal, granular disintegration begins and crumbling is observed. This process is very similar to the granular disintegration that is observed on many sandstones (Smith et al. 2003). The result is the rounding of edges and corners as well as significant material loss (up to 4 cm). An extreme case of mechanical breakdown is alveolar weathering (honeycombs) but it is restricted to few ashlars of ooilitic limestone. Stone surfaces exposed after crust removal have different surface properties. If we compare the changes in Schmidt hammer and Duroscope rebound values observed on oolitic limestone and travertine it is well documented that the loss in surface strength is more severe for oolitic limestone, than that of the travertine.

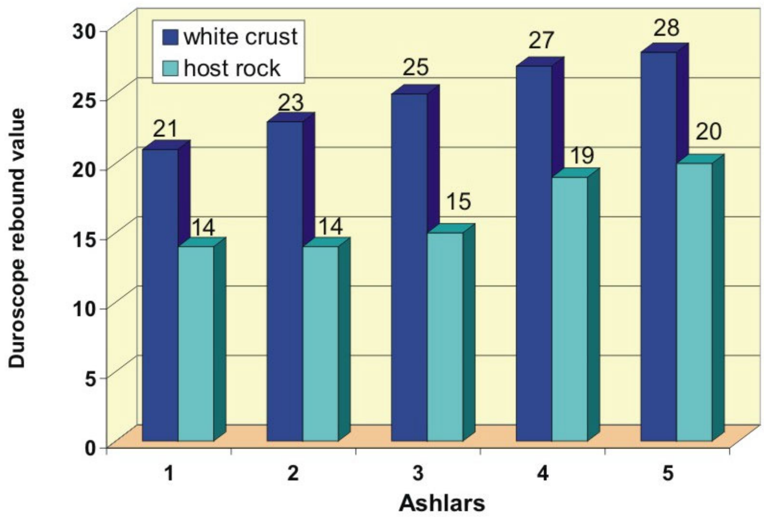

Figure 6. Duroscope rebound value of white weathering crust is greater than that of the host oolitic limestone

DISCUSSIONS

The results presented in this paper document that deterioration is a complex process that changes the mechanical properties and mineralogical composition of limestones. The difference between the surface strength and water absorption of unaltered rock, crusts and host rock varies if we compare various carbonate substrates.

The weathering leads to the formation of secondary gypsum in the near surface zone of the limestone ashlars. The penetration depth of the gypsum crystals depend on the pore structure of the rock. The formation of gypsum can have two various effects. On one hand side it can contribute to the removal of crust by exerting crystallisation pressure if it is formed below the crust. Concurrently when gypsum precipitates in the pores of oolitic limestone it can also serve as a pore occluding cement (Török & Rozgonyi 2004) and thus it helps in the formation of a non-porous surface crust on the oolitic limestone (Török 2003). On-site non-destructive mechanical tests indicated that there is a significant difference in the surface strength of crust and host rock. Thick white crusts have higher Schmidt hammer rebound values than the host oolitic limestone. The difference is in the order of 30%. Duroscope rebound values reflect the same, i.e. the strength of the thick white crust is significantly larger than that of the host rock. Laminar black crusts also strengthen the rock surface of the oolitic limestone according to Schmidt hammer and Duroscope rebound tests. Meanwhile there is a decrease in strength when the travertine and oolitic limestone is compared. Travertine has higher Schmidt hammer rebound values than that of the oolitic limestone when both stones are weathered (Figure 7).

If we compare the Duroscope rebound values of porous oolitic limestone from the quarry with thin white crusts, thick white crusts, laminar black crusts and the weathered host rock below an important trend is observed. Crust formation leads to the weakening of the host rock and concurrently to the formation of rigid crust. The crust itself can have a higher surface strength than the unaltered quarry stone. But this apparent increase in surface strength is not observed on travertine. To the contrary weathering leads to a decrease in Schmidt hammer rebound values and Duroscope values of travertine.

CONCLUSIONS

By comparing the alteration processes of the two limestones it seems that travertine surfaces show much less alteration than that of the porous oolitic limestone. The mineralogical analyses show that gypsum is present in all crusts and as a secondary mineral it is related to calcite/SO_x interaction on the stone surface. The amount of gypsum varies and it depends on the host rock fabric and on crust morphology. The crusts are apparently stabile on travertine while both black and white crusts tend to scale, blister or flake on porous oolitic limestone.

The formation of white weathering crust on oolitic limestone leads to an increase in Schmidt hammer and Duroscope rebound values and also results a decrease in water absorption. Weathering crusts on travertine show decreased Schmidt hammer and Duroscope rebound values compared to quarry stones.

By the removal of the crust from the oolitic limestone a part of the substrate is also removed, leading to surface retreat. To the contrary when framboidal crusts are scraped off from travertine the substrate remains relatively intact and the stone is not damaged. Thin surface parallel laminar black crust are also stable on travertine and can be easily removed without damaging the stone surface. The thin laminar back crust behaves differently on oolitic limestone and it tends to flake or scale. Thick case hardened white crusts are removed by contour scaling while thin white crusts of oolitic limestone tends to blister and flake. These differences of crust stability invoke different methods of surface stabilisation in limestone restoration practice. Namely the black crusts from the travertine can be removed by using abrasive methods (eg. high pressure water spray) without damaging the stone surface.

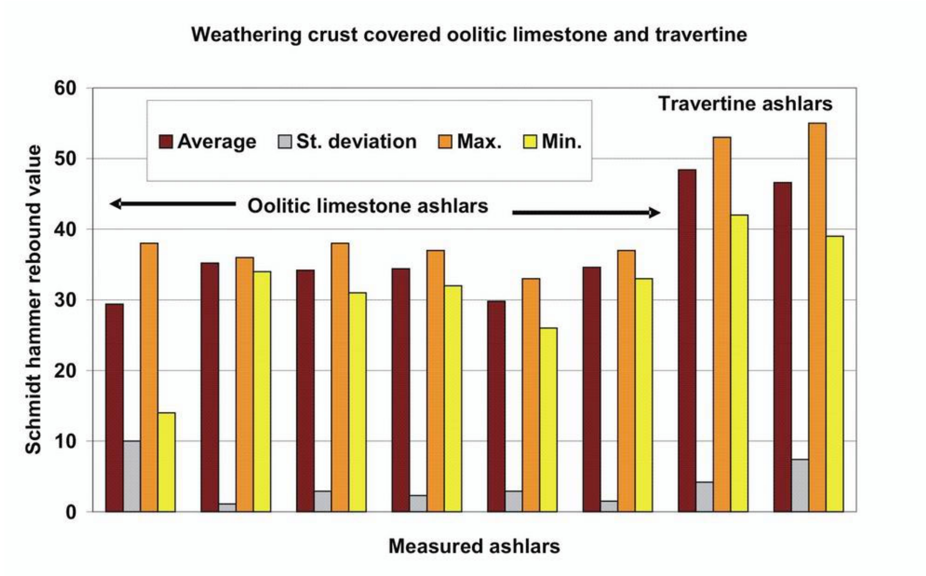

Figure 7. Schmidt hammer rebound values of oolitic limestone and travertine ashlars

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