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Influence of environmental (external) factors on durability of freshwater limestones *tenelija, miljevina* and *muljika*

Krešimir Šaravanja
HNC Ministry of Construction and Physical Planning and Faculty of Civil Engineering, Architecture and Geodesy, University of Mostar, Ph.D., kresimir.saravanja@fgag.sum.ba

Abstract: In order to avoid complex processes of changes, or weathering (decay) of freshwater limestones embedded in numerous buildings and/or monuments due to interaction with various environmental conditions (external factors), builders have always tried to avoid construction (internal) factors, such as the selection of stone, the method and (often too small) depth of its extraction from the deposit for the construction of significant buildings, insufficient aging (storage) after extraction, improper installation (stacking) of stone in relation to its position in the deposit, as well as mistakes in stone processing and execution, in addition to inadequate maintenance. This paper presents the complex processes of changes, or weathering (decay) of freshwater limestones embedded in buildings and monuments as a result of its interaction with various environmental conditions (external factors), such as: the effects of temperature, humidity and quality of air, winds, precipitation, acidic rain, exhaust gases, micro-particles and organisms in the air and water, harmful salts, radiation, soil, biological colonization, along with the destructive action of man. In this process, the aesthetic deterioration of the stone surface is only a secondary problem compared to the destruction of its structure. The consideration must certainly include the pollution flows and the environment, and meteorological and microclimatic conditions.

Keywords: limestones, freshwater (limestones), lacustrine (limestones), oolitic (limestones), *tenelija*, *miljevina*, *muljika*, durability, external factors

Utjecaj okolišnih (vanjskih) faktora na trajnost slatkovodnih vapnenaca *tenelije, miljevine* i *muljike*

Sažetak: Da bi izbjegli složene procese promjena, odnosno trošenja (propadanja) ugrađenih slatkovodnih vapnenaca u mnogobrojnim građevinama i/ili spomenicima uslijed interakcije s raznovrsnim uvjetima okoliša (vanjskim faktorima), graditelji su oduvijek pokušavali izbjeći ugrađene (unutarnje) faktore, kao što su izbor kamena, načina i (često premale) dubine njegovog vađenja iz ležišta za gradnju značajnih građevina, nedovoljno dugog odležavanja (skladištenja) nakon vađenja, neispravne ugradbe (slaganja) kamena u odnosu na njegov položaj u ležištu, kao i greške u obradi kamena i u izvedbi, uz neodgovarajuće održavanje.
U radu su prikazani složeni procesi promjena, odnosno trošenja (propadanja) ugrađenih slatkovodnih vapnenaca u građevinama i spomenicima uslijed njegove interakcije s raznovrsnim uvjetima okoliša (vanjskim faktorima), kao što su: utjecaj temperature, vlažnosti i kvalitete zraka, vjetrova, padalina, kiselih kiša, ispušnih plinova, mikro- čestica i organizama u zraku i vodi, štetnih soli, radijacije, zemljišta, biološke kolonizacije, uz destruktivno djelovanje čovjeka. Pri tome, estetsko nagrđivanje površine kamena je tek sekundaran problem u odnosu na uništavanje njegove strukture. U razmatranje trebamo obvezno uključiti tokove onečišćenja i okruženje, odnosno meteorološke i mikroklimatske uvjete.

Ključne riječi: vapnenci, slatkovodni, jezerski (vapnenci), oolitični (vapnenci), *tenelija, miljevina, muljika*, trajnost, vanjski faktori

1. INTRODUCTION TO THE DURABILITY/ RESISTANCE/ STABILITY OF STONE

Natural stone has been used in construction since ancient times, where it is exposed to the phenomenon of aging, which is more or less accelerated by some factors, so we are talking about changes in stone, or its weathering, and in more severe cases even deterioration of stone (gradual destruction). Increasing air pollution, starting with the industrial revolution in the middle of the 19th century, accelerates these processes more and more.

According to prof. Crnković, the durability of built-in natural stone includes the "*behavior of built-in stone in a wide range of effects of various factors*", [1]. The literature also uses the terms: temporal permanence or durability (prof. Bilbija and Tamara Plastić), resistance, and stability.

The durability of stone is primarily influenced by the processes to which the rock mass in the stone deposit was exposed, or its textural and structural properties (exceptionally coarse-grained stone is weathered faster), and porosity (the smaller the pores, the greater the possibility of capillary moisture spreading, and thus also the possibility of damage or changes in the porous stone whose minerals are less resistant to degradation), [2]. The mineral composition (and its possible transformations) has a particular influence on the durability of the built-in stone, [3].

The issue of stone durability should be placed in the context of the stone's utility value, i.e. whether the stone is incorporated into a valuable building of cultural heritage or a commercial building of lesser importance.

2. ENVIRONMENTAL (EXTERNAL) FACTORS, AS CAUSES OF CHANGES IN STONE BUILDINGS MADE OF FRESHWATER LIMESTONES *TENELIJA, MILJEVINA* AND *MULJIKA*

As cultural and historical buildings are usually built using locally available, easier to process types of stone, the portion of sedimentary (carbonate) stone is high (in Italy the portion of limestone and marble is as high as 90%) due to its aesthetic properties, availability and workability, even though its chemical composition makes it more exposed to environmental effects and deterioration, compared to silicate stone, which is much more widely used in Germany, Spain and other countries, [4].

On natural stone built into numerous stone structures and/or monuments (hereinafter: buildings), complex processes of changes occur on the stone due to its interaction with various physical and chemical environmental influences (external factors), such as: influence of temperature, humidity and quality of air, winds, precipitation, acidic rain, exhaust gases, micro-particles and organisms in air and water, harmful salts, radiation, land, biological colonization, use of inappropriate materials in previous construction (iron, cement,...), with destructive effects of humans (war effects, additions that damage old buildings, faulty repairs, soiling with stone glue, mortar, grease, stains from iron (oxide), copper and bronze (copper oxide, carbonate and bicarbonate), oils, paraffins, various types of acrylic and synthetic paints for drawings, signs and graffiti (sprays, markers, felt-tip pens, etc.), chewing gum,... Climatic influences are best manifested on prominent parts of the building (effect of rain, wind, ice).

Thus, the durability of stone is a resultant of all environmental actions and the stone's ability to resist these actions and to continuously keep its aesthetic and functional properties, [1].

2.1 Effect of temperature/temperature changes

Regarding the effect of temperature or temperature change (in the environment), the intensity and dynamics of changes on stone/deterioration depend on the absolute value of temperature and the rate of its change, but also on the type of stone, the size of the pores, and the quantity of water in the stone. The phenomenon is most common in heated stone, so chemical deterioration is more pronounced in warmer (and wetter) climate areas, [2]. Insolated parts heat up more intensely, and shaded parts cool down faster, so moisture from the atmosphere can condense on those colder parts.

Alternating heating and cooling of the stone or temperature changes (daily and seasonal) in the environment causes changes to the stone/deterioration in the long term. In the process, seasonal temperature variations of the surface of the built-in stone are less significant than daily variations, which in the summer period in the continental climate area can be as much as 40-50°C, depending on the position (exposure), where damage to the stone from thermal stress is usually increased on the south and west sides, [5].

A couple of examples of the impact of pronounced temperature changes in summer on stone in buildings will be mentioned.

According to prof. Radnić et al. (2011), "*in addition to the effect of vertical loads, temperature effects are considered to be the main cause of cracks in the original and reconstructed arch of the Old Bridge in Mostar*", [6].

According to the conservation and restoration studies of the facade of the bell-tower and the western facade of the church of St. Peter and Paul on Gorica in Livno (2011), "*due to significant daily temperature changes in the summer months on the facade of the church of St. Peter and Paul on Gorica in Livno, the conducted tests established that there was damage to the lime joints and stresses of microcracks in the stone, which in winter allowed the penetration of water, leading to damage due to the daily freezing and thawing cycles*.", [7].

However, the greatest damage is not caused by the influence of temperature/temperature changes on the stone, but in conjunction with other environmental factors, primarily water. At very low temperatures, as a result of freezing, the water trapped in the pores of the stone increases its volume, breaking it, while in the heated stone, at very high temperatures, there are internal stresses due to the pressure of the generated water vapor, which lead to the formation of micro-cracks, which gradually turn into cracks and fissures, leading to damage of the stone (structure). Therefore, temperature is an indirect cause of material deterioration here, [5].

2.2 Influence of wind(s)

Very strong winds can create large horizontal loads in structures, and cause some deterioration, [5].

Different exposure to wind/s (and rain), which often carries particles of pollutants, dust and earth, can lead to the process of formation of various types of stone deterioration on various walls of the building, including the deposition of pollutants.

2.3 Inorganic pollution (impact of polluted atmosphere and salt crystallization)

According to Gobić-Bravar, "*every monument in an urban environment that is exposed to atmospheric effects will eventually be covered with various impurities that will more or less adhere to the stone surface. These deposits are mostly black in color and are usually found in places that are protected from rain*", [8].

According to the *ICOMOS-ISCS Glossary*, [9] crusts (Cro. "*kora*" sing.) are "*coherent accumulations of material on the surface of a stone of visible thickness which, when uneven, make it difficult to read the surface relief; they are often dark in color, although they can also be light, include exogenous material in combination with material from the stone itself, and can adhere more weakly or firmly to the substrate, so removing them can also remove part of the stone material*", while black crusts (Cro. *crna kora*) are "*calcified accumulations (deposits) of stone elements and small particles dispersed in the air (soot, from which the black color usually originates, and organic and inorganic particles of different origins), generally caused by urban or industrial pollution, on surfaces protected from direct impact of rain and of its runoff (under cornices, windows, parts of statues and tops).*" They mainly consist of particles from the polluted atmosphere "caught" in the plaster matrix and usually firmly adhere to the substrate, [10]. The black color comes from soot particles produced by the burning of coal (and wood) in domestic fireplaces and industrial plants. They are visibly thick, in some places thicker than 1 cm.

According to Grbelja, "*dark deposits of crust* (Cro. krasta, skrama) *accumulate on the surface of the stone and do not represent its integral part. They are formed by the action of sulfuric acid from an excessively polluted atmosphere on calcium carbonate in the stone, and turn it into calcium sulfate (gypsum). In the process of formation, they take on different ingredients from the atmosphere (soot, iron and manganese compounds, dust and organic substances).*", [11].

In the Croatian conservation and restoration literature, the names "skrama" (crust), "crna skrama" (black crust) and "tamna skrama" (dark crust) are also used for deposits formed in protected places, which contain calcium sulfate (gypsum), the main cause of damage to stone plastic. They are formed due to the action of atmospheric agents; they are insoluble, cohesive and adhere firmly to the substrate, [12]. In addition to these names, the names "tamna kora" (dark crust) and "crna inkrustacija" (black incrustation) are also used.

The names "skrama" and "crna skrama" appear in very serious professional publications and are used more widely than the names "kora" and "crna kora", which, according to Hraste, are still linguistically clearer as translations of the standardized names *crust* and *black crust*, [10].

Black crusts are the most visible and the most studied. They represent the most extreme form of impurities on stone buildings. They often contain harmful (water)soluble salts (sulfates, nitrates, chlorides), which visually impair the stone, concealing its deterioration (up to breaking of the stone structure). Chemical and physical damage disintegrate the stone surface. Salts driven by moisture travel and reach the stone substrate, where they crystallize by drying, breaking the surface structure of the stone, [13], [14], [15]. Surface change and depth of damage are defined by the porosity of the stone, [16]. Stone deterioration shows significant variations depending on the stone, pollution and meteorological factors.

From the changes in the names for the phenomenon of thin black deposits on Peristyle, it follows that the name "tamne presvlake" (dark coatings), used in publications from 2007 and 2009, did not survive and was replaced by the better known name "patina", which, according to the *Glossary*, means "*a micron thin layer in which discoloration of the stone occurred, but in most cases a noticeable drop in the quality of the material did not occur*". In the literature on stone in the English language, we often find the name thin black crust for the same type of deposit, which Pagona-Noni Maravelaki-Kalaitzaki defines as "*a black crust up to 0.2 mm thick that develops on the surface of the stone in a continuous, tightly adhering layer*", [17]. In Italian literature, the name "*patina nera*" is common. According to Grbelja, [11] this name is used as a higher order name in relation to the name "crna kora". Matijaca, [18] uses the phrase "sivkasta patina" (grayish patina) in the meaning of the very thin black deposits that were discussed, while Gobić-Bravar uses the term "patinas" for "*dried lichen colonies*", [19]. Wavering in the naming of the thin dark deposits of exogenous atmospheric particles of almost invisible thickness, which give the stone the appearance of dirtiness, according to Hraste, shows the indecisiveness of the profession in classification of this phenomenon. The issue is not a terminological one, since there are two assumptions about how these deposits are formed. According to one, it is the first step in the development of "black crusts", and according to the other, it is not always so, since environmental conditions do not always support their development on a particular surface, [17].

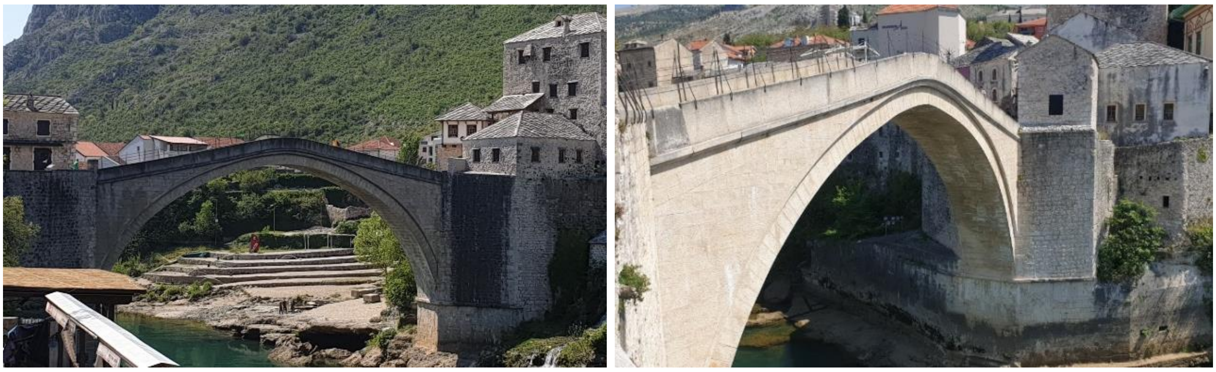
According to Nikšić and Sunara, three types of surface deposits are present on the surface of the Peristyle stone: carbonate crusts, grayish-brown crusts and thick sulfate crusts rich in carbon particles, black in color. Under the black crusts there was mainly a well-preserved yellowish oxalate patina, [13]. According to them, "*patina is a thin recrystallized surface layer of stone, or a layer that goes from the stone surface towards its interior. The minerals that form patinas are calcium oxalates and they protect the surface of stone*". The sedimented dark deposits on the surface contain harmful calcium sulfate, so they should be removed with lasers, while preserving the oxalate films - patinas that are located between the stone substrate and the deposits, and whose yellowish-brownish tint is desirable on the stone surface, [13], [14].

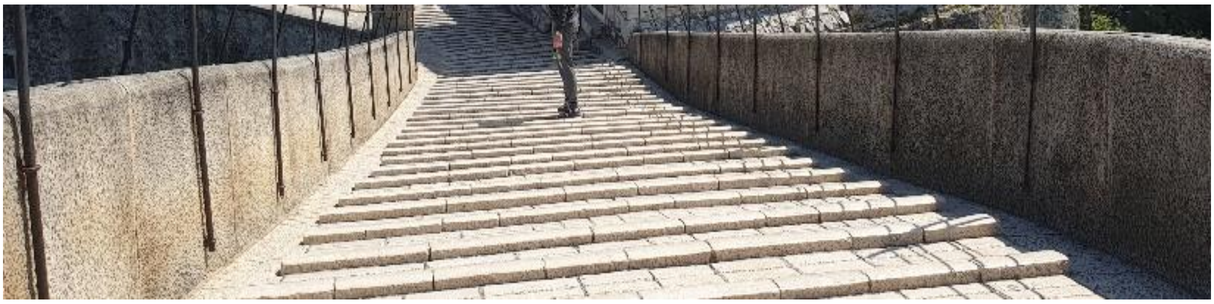
When removing impurities, it is important to distinguish between crusts on the stone, which are a foreign body on the stone surface, and considering their composition and contamination with salts, they can be the cause of damage, and the natural patina, chromatic modification, which mainly results from natural or artificial aging and it is its natural protection against the atmospheric effects, [20] that is, in most cases it does not involve visible deterioration of the surface. They are found on all exposed parts, not only on parts sheltered from rainfall.

Bearing in mind the above, the terms "kora" (crust), "crna kora" (black crust) and "patina" are used in this paper.

Tests of the chemical composition of samples of built-in freshwater limestones in Mostar, Posušje and Tomislavgrad are performed, as well as tests of the chemical composition of patina on the same samples ("Cerberus" d.o.o. Tuzla, 2020). The values of $CaCO_3$ in the patinas on the *Mostar* and *Posušje miljevina* were in a relatively narrow range of 94.96%-96.42%, while in the patina samples on the *Tomislavgrad muljika* they were in the interval of 87.07%-88.18%, or slightly lower, due to a significant proportion of SiO_2, [21].

In order to separate it from "crust" and "patina", the authors of the *Glossary* chose the name *soiling* (Engl.) for "*a very thin deposit of exogenous particles (like soot) that can more or less firmly adhere to the substrate*". Its source is atmospheric pollution (industry, households, traffic) etc. It gives the stone a dirty appearance, but it does not corrode it as much as the "black crust", into which it can transform with an increase in the forces of adhesion and cohesion. The meaning of the noun *soil*, which is "impure" (or "earth"), was later much more widely accepted in the forms "impurity" (nečistoća) and "pollution" (onečišćenje), which in professional texts have a slightly modified meaning of deposit in general, for which the *Glossary* suggests the English name *deposit*.





Figures 1-3. The Old Bridge in Mostar, a monument on the UNESCO list of protected world heritage: the upstream, northern side with a more pronounced patina (above, left), compared to the downstream, southern side (above, right); In addition to patina on the arch intrados, patina is visible on the parapets, cornices and vertical walls made of tenelija freshwater limestone; View from the west side of the bridge cobblestone with the inner sides of the parapet showing patina, especially the right, north side of the downstream (southern) parapet (left); damage is also visible on the joints of individual parapet blocks, and on some of them the "lime binder" is washed out, which atmospheric water has partially dissolved over a long period of time so that the joints between the blocks remain empty (below), (photo: K. Šaravanja), [21]

Figures 4-5. The facade of the Shopping Center "Mercator" in Mostar (left), with a detail (right), made of (Mostar) miljevina, which shows a tendency to patina the surface and a tendency to soiling when the stone is exposed to the action of seepage water from horizontal surfaces and gutters, on which there is a deposition of dust and polluting particles from the atmosphere that can stain the surface layer of vertically installed elements made of this stone, (photo: K. Šaravanja), [21]

Figures 6-7. Various forms of changes to the stone in the retaining wall of the Bishop's Palace in Mostar (physical impact of atmospheric water, damage from war activities, biocolonization, damage caused by corrosion of the iron fence, anchored in the stone, which corrode in contact with moisture, and the change in volume causes cracking of stone, as well as multi-meter vertical stains on the stone, which are very difficult to remove; The stone in the building of the Bishop's Palace was repaired after the war, (photo: K. Šaravanja), [21]

Tombstones made of freshwater limestone in the area of Mostar and Herzegovina (and beyond), some of which are hundreds of years old, have different surface patination and soiling, from partial to very high. The stone of the horizontal surfaces of the tombs has acquired black deposits due to the deposition of soiling particles from the atmosphere and various impurities (earth, leaves,...), dust and microorganisms, which in periods of wetting have stuck to the surface that remains moist for longer, while the accumulated impurities are washed away by rain from the vertical part of the tombs, so only black particles of plant origin bind to it. In addition, there is also the capillary action of water from soil, salt efflorescence. The surface of the stone scales and falls off, so the stone loses its original aesthetic properties, and to a certain extent, its bearing capacity and durability. Due to the draining of water with these impurities, the surface layer of the lower vertical stone elements is also soiled.

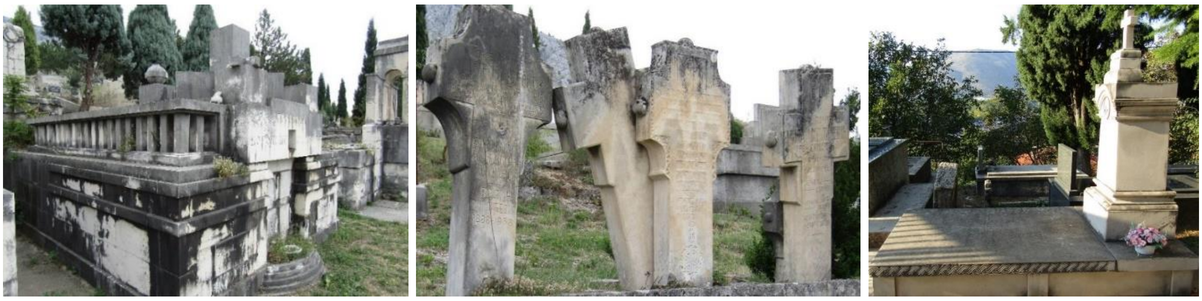


Figure 8-10. Different forms of patination and soiling of surfaces on tombs made of freshwater limestone in Mostar: Bjelušine cemetery (left and center); Although the grave crosses are almost of the same height and shape, they are patinated differently, which indicates not only their different age, but also probably the different quality of the stone, or different layers in the quarry from which they were extracted. The inscriptions on some of them have completely disappeared due to the action of rainwater or are difficult to read (middle); The contrast between the dark (horizontal) and light (vertical) stone surfaces of the tomb in the Šoinovac cemetery in Mostar, along with the patinated inclined and smaller horizontal surfaces of the tombstone, create the impression that the tomb made of (Mostar) miljevina was made of several types of carbonate lithotypes (right), (photo: K. Šaravanja), [21]

In the area of the municipality of Posušje, to reduce construction costs, walls of houses together with facade were built with *(Posušje) miljevina*, which was quarried in several locations 80-160 years ago.

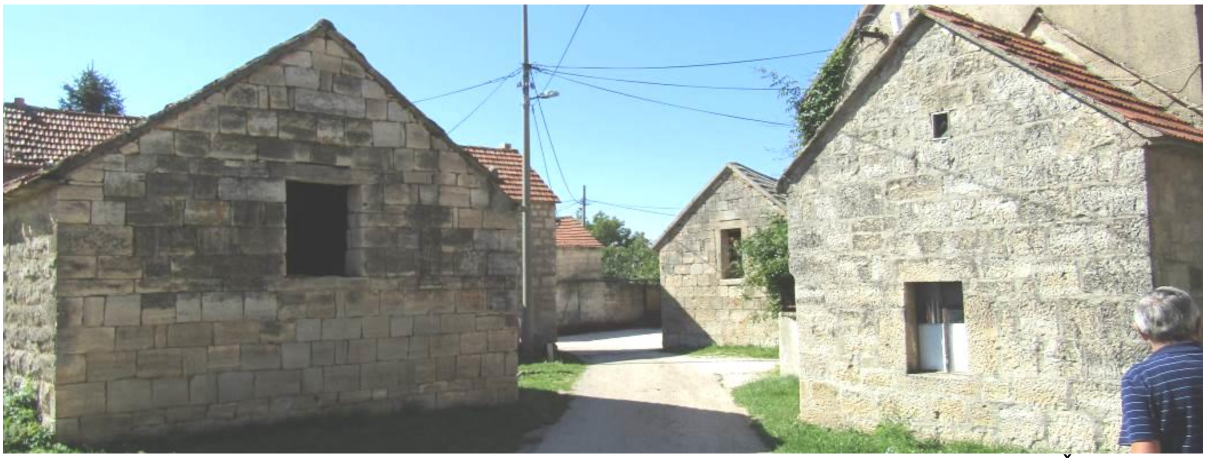
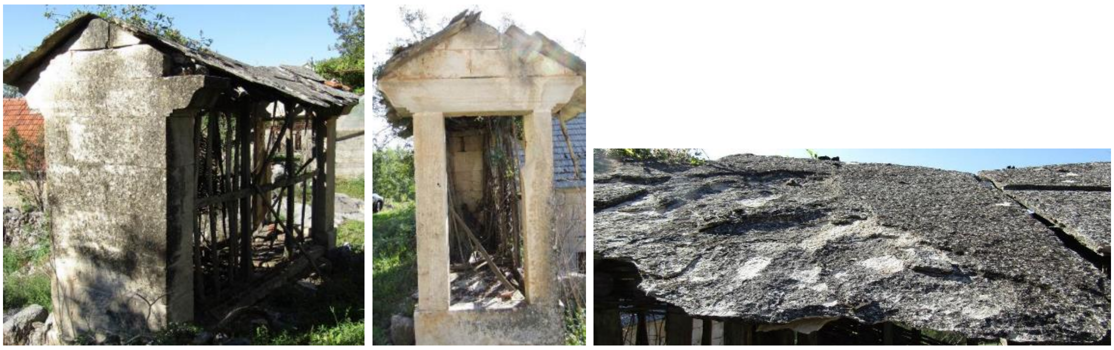


Figure 11. Relatively little patination of facades made of (Posušje) miljevina in Ramljaci-Čitluk near Posušje; Orientation of the building and its position in relation to the adjacent buildings are important, so there are certain differences when it comes to a standalone building, or a building that is the first/last in a row, (photo: K. Šaravanja), [21]

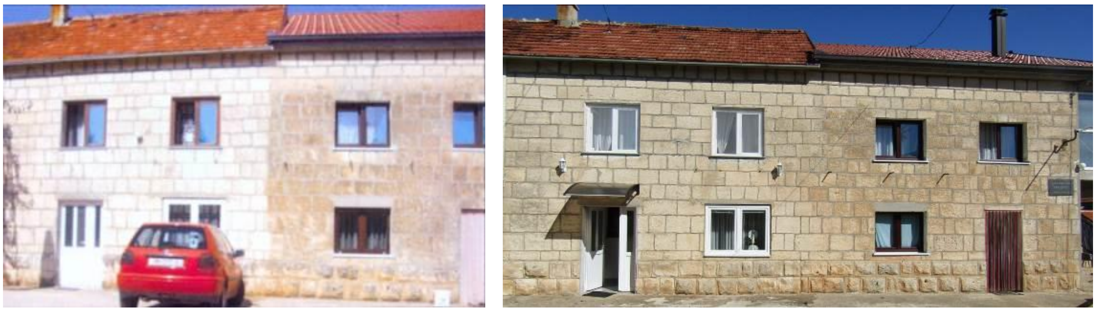


Figures 12-14. Corn storage (corn crib) in Tomić Brig near Posušje, about 120 years old, with dimensions 3x1.5 m; one can see the excellent preservation of the (Posušje) miljevina stone, especially the entrance part, which has less patina (middle); A cover made of stone slabs 0.2-0.3 m^2 in area, about 3 cm thick, covered with patina, with some lichen (right) deserves special attention, (photo: K. Šaravanja), [21]

Buildings made of *miljevina* (and *tenelija*) have an advantage over buildings built with hard limestone due to the easy refreshing of the facade by removing patina from the stone by hand or by machine.

Figure 15. Removal of patina from the facade of the "Sivrića" building in Mostar made of tenelija (end of 2016), [22]

Frano Oreč found different thicknesses of patina on buildings in the Municipality of Posušje, built from stone quarried in a large number of places, 90-170 years old, and concluded that it is not determined by the age of the building, but by the size and density of the *miljevina* grains, [23].



Figures 16-17. An example of removal of a layer of stone with patina from the left part of the facade made of (Posušje) miljevina on the house in Vinjani near Posušje with a special tool (pick with 3 teeth). An 8-10 mm thick layer was removed from an area of about 50 m^2 in 3 days, (left, photo: F. Oreč, 2011). Eight years later, a certain return of the patina is visible on that part of the house, (right, photo: K. Šaravanja, 2019), [21]

2.4 Physical impact of (atmospheric and ground) water and moisture

Buildings/monuments are directly exposed to the effects of moisture in three ways: in the form of liquid water (rain, floods, groundwater), in the form of hydroscopic moisture that is formed when salts accumulated in the building absorb water vapor from the air, while condensation moisture is created from high humidity in the air and hydroscopic salts, but it is extracted on the surface of the building in the form of dew when warm air comes into contact with cold air.

Water is not only one of the fundamental causes, but often a companion of almost all degradation processes on stone, especially to porous freshwater limestones, whose porosity (shape, size and distribution of pores) significantly affects the amount of water (and water vapor) received in the stone structure, its retention and draining. Increased moisture penetration can activate passively accumulated and stored salts in the stone, which, once activated, cause damage by migration to the point where the moisture evaporates. Frequent alternations of wetting and drying of stone cause the dissolution, accumulation and crystallization of salts contained in the stone, which forms strong crystallization and hydration pressures that cause damage in the form of "floury" falling and flaking of the material, [24].

Therefore, the stone should be installed so that it is well ventilated in order to reduce the amount of moisture in it as much as possible.

Built-in freshwater limestones in the open space are directly exposed to the influence of water and moisture, which is mostly chemical in character, although the physical influence is not negligible either. Rainwater drives out loosely bound particles and lowers the relief (erosion), and constant wetting and drying, especially on rough surfaces, leads to deterioration of physical and mechanical properties (softening, reduction of strength, as well as of resistance to wear and frost). This is pronounced especially on the (northern and western) sides exposed to the impact of raindrops carried by the wind. In cities where squares and streets are washed, more frequent wetting leads to increased deterioration of built-in stone. Different sides of the building/monument may have different exposure to rain (and wind), and the duration and intensity of rain increase with the increase in extreme wind and storm speeds (impact of raindrops carried by the wind). In addition to the increase in the penetration of rainwater into the building, there may also be an increase in the amount and locations of outflow caused by rain.

In addition to mechanical (erosive) action, water also plays an indirect role in its deterioration, participating in chemical reactions with the mineral components of stone, dissolving and transferring soluble salts (corrosion) and migrating to the point where moisture evaporates, [13], [14], [15]. This effect of rainwater is most pronounced when the air temperature is slightly above zero, because then the water contains the most dissolved CO_2, or it has the lowest pH-value.

In addition, water enables the growth and development of microorganisms. It should be emphasized that the damage caused to the stone is directly related to the duration of moistness, [5], [9].

The more water present, the faster the process of degradation of freshwater limestone.

Regarding the mechanism of water movement in the stone, it goes on from the beginning of the genesis of the stone. After extraction ("picking of stone"), freshwater limestones, due to their high porosity, contain a large amount of natural (quarry or mine) moisture, which may contain dissolved salts. As the stone dries, salts together with moisture migrate towards the surface of the stone, where they crystallize, forming patina. By losing moisture, they acquire greater hardness and strength due to the formation of a solid carbon lattice on the surface.

Rain carried by the wind, as well as wind gusts and air pressure can lead to an increased amount of absorbed water, or an increased rate and depth of rainwater penetration into the stone. Fluctuations in air pressure associated with gusts of wind can cause pressure differences between the interior and exterior of the building/monument, which can force water to flow over the surface to enter voids and be trapped in the pores. In the following successive steps, water can get inside the porous stone.

The reception, retention and draining of water, or the amount of water and water vapor that enters and moves through freshwater limestone, is most influenced by its porosity and pore structure (shape, size and mutual distribution of pores), which is defined as the pore space content. Stone with increased open (relative) porosity, and thus greater absorption, is more sensitive and less durable.

When rainwater is relatively clean, changes to the stone are not a serious problem, but when it contains dissolved CO2, a weak carbonic acid is formed, which converts the carbonate into a very soluble bicarbonate and thus destroys the limestone. Consequently, an increase in porosity changes the hydrological properties of stone. The loss of material also weakens the structure of the stone and decreases its mechanical resistance, [25].

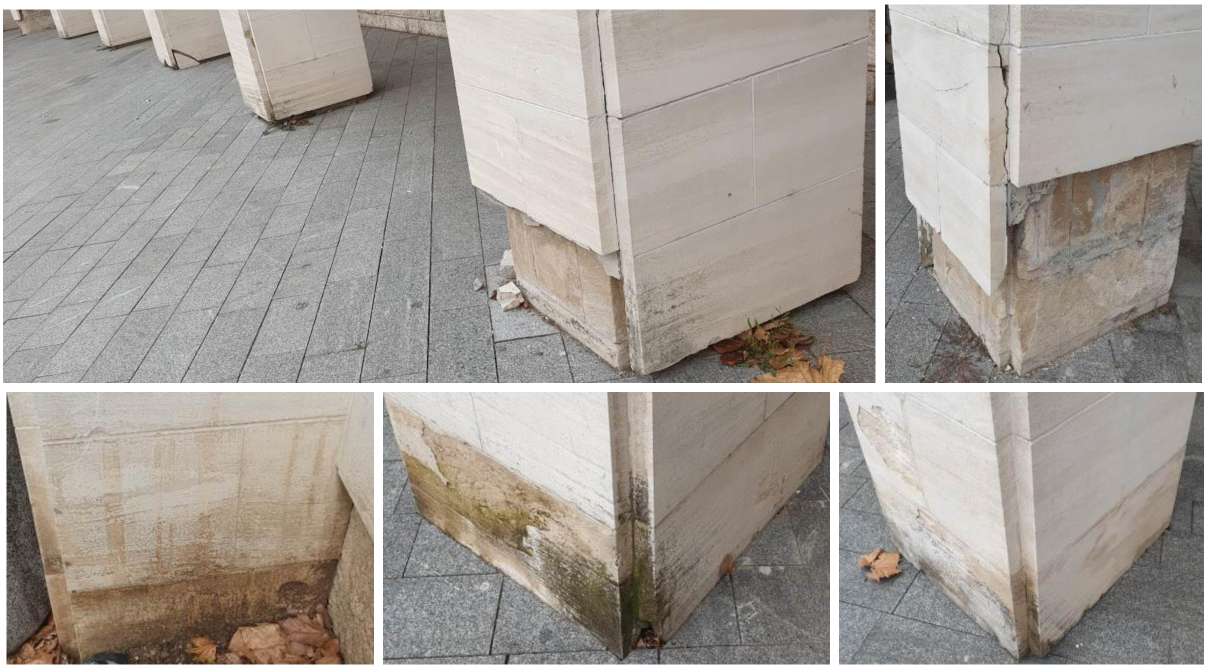
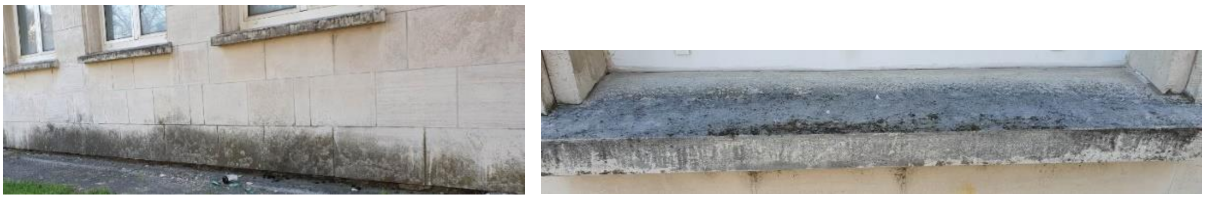


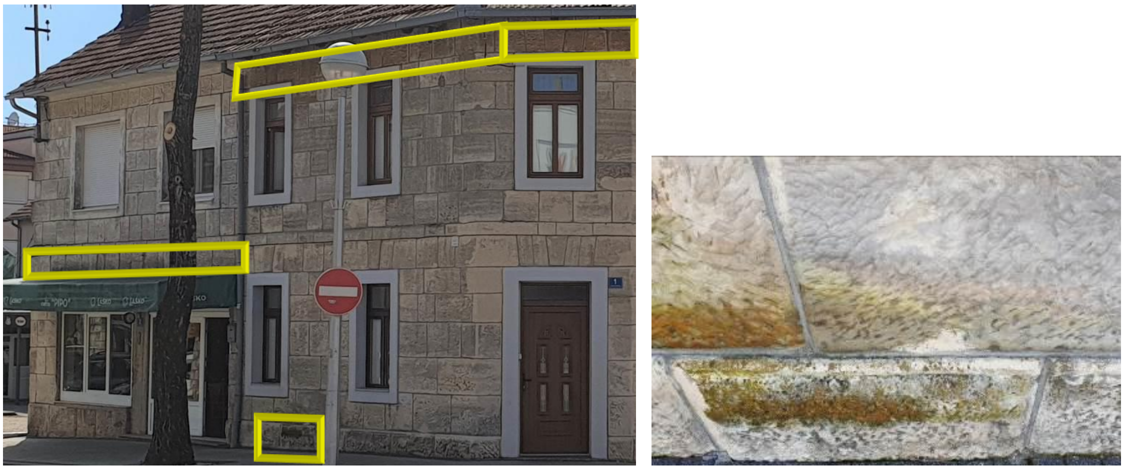
Figure 18-22. Deterioration of the lower part of the facade made of (Mostar) miljevina, which is used for decorative and protective covering in the form of a thin stone base of the Croatian Center Herceg Stjepan Kosača in Mostar due to the physical impact of atmospheric water and moisture, (photo: K. Šaravanja), [21]



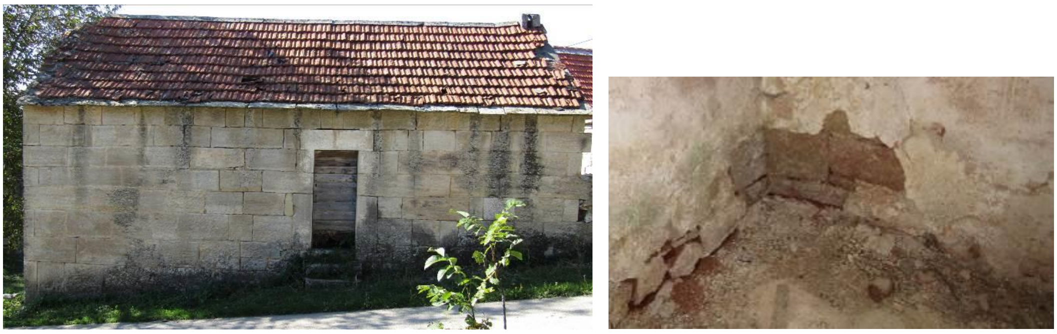
Figures 23-24. Deterioration of the lower part of the facade made of (Mostar) miljevina, which is used for decorative and protective cladding in the form of a thin stone base and stone sills under the windows of the Croatian Center Herceg Stjepan Kosača in Mostar due to the physical impact of atmospheric water and moisture, with a detail of the sill under the window, (photo: K. Šaravanja), [21]

Knowing the limitations of the application of *tenelija* and *miljevina* related to high water absorption, the builders in Mostar often used another type of stone for lower parts of buildings.

Although *miljevina* has a dense, fine-grained structure in which no cementation and recrystallization of calcite has occurred, so it keeps moisture for a long time, and so the effect of temperature changes and ice can cause surface loosening and deterioration of stone, a number of constructed buildings, especially those in higher parts of the municipality of Posušje, were inspected with visible good preservation of the built-in *(Posušje) miljevina*, and changes on the stone are present only in the lower parts of the buildings as a result of the capillary rise of rainwater (and snow drifts) over decades.



Figures 25-26. The effect of atmospheric water under the roof gutters and above the awning on a house in Posušje made of (Posušje) miljevina, more pronounced in the lower part of the building due to the capillary rise of rainwater (and snow), with detail (fungi and lichens), (photo: K. Šaravanja), [21]



Figures 27-28. Degradation of the lower part of the northern facade (and foundation) of the house in Tomića Brig near Posušje, as a result of the capillary rise of rainwater and snow drifts, or the cycle of freezing near the ground; Due to the surface of the surrounding ground in a slope, surface water flow and flooding of the wall are present; The roof did not have a gutter, so rainwater fell from the roof onto the ground and bounced off onto the facade together with soil particles (left); detail of the interior of the house, as an illustration of the degradation (right), (photo: K. Šaravanja), [21]

According to Frano Oreč, there is a problem of increased moisture in old buildings made of *miljevina*, especially those that were not built according to the rules of the profession, so it is necessary to make sure that expansions joints are sealed with binder, and that they are renewed and maintained. Some of the buildings were founded with a different, harder type of stone, with the idea that there would be no moisture problems in this way, which did not prove to be justified, [23].

2.5 Action of frozen water in stone (action of frost)

The action of frost can play a significant role in the destruction of built-in stone, especially stone sensitive to frost in colder climates with low temperatures and humid conditions (large number of freeze/thaw cycles), particularly stone already damaged by other processes, such as salt crystallization. The consequences are changes in the mineralogical composition and mechanical properties of stone (blocks of masonry stone and stone ornaments). Disintegration of grains occurs, as well as the removal of the stone layer by scaling and peeling.

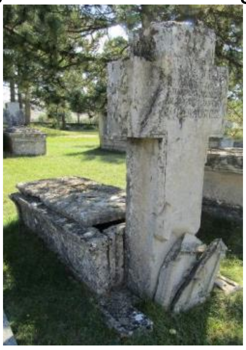
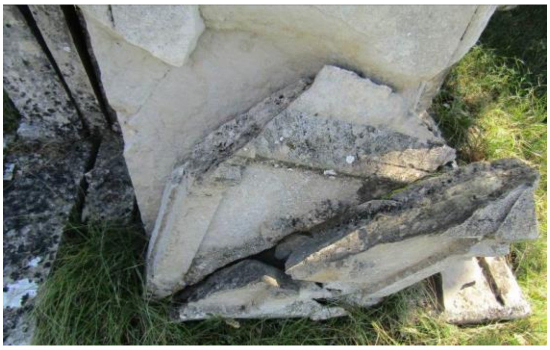
The susceptibility of stone to frost depends significantly on its porosity and pore size distribution, with the stone with the smallest pore size being the most susceptible to frost damage. Frost resistance generally decreases with increased pore volume that is accessible to water.

After conducting appropriate laboratory tests of the analyzed freshwater limestones, the resistance to frost is questionable. From the tests in the 1960s until today, contradictory results have been obtained for *tenelija* - that it is "resistant to frost", that it is "poorly resistant to frost", and that it is "not resistant to frost". The holes around the balls of *tenelija* ooids represent a space for the expansion of ice during freezing, so the possibility of stone cracking is lower.

The somewhat better resistance to frost damage in *tenelija* compared to *Mostar miljevina* indicates that due to its coarse-grained structure, water is drained out of the stone faster, or it indicates a shorter retention of water in the stone structure, which is also supported by the large difference in the results obtained from the test of water absorption by the process of saturation by boiling in relation to absorption under atmospheric pressure. A greater filling of pores with water by 33% was obtained for *tenelija*, compared to 13% in *Mostar míljevina*, where there is no space left for the expansion of ice, which leads to destructive tensile stresses inside the stone.

Related to the frost resistance test, in addition to the difference in the "age" of the stone "freshly extracted" from the quarry and the "old" stone that is installed in the buildings, I note that the frost tests were performed according to different standards, since the crystallization test with sodium sulfate is ten times more aggressive to stone than the conventional frost test with cycles of freezing and thawing (the effect of negative temperatures on water-saturated stone), and that the crystal lattices of sodium sulfate and magnesium sulfate are different, the consequence of which is different results of the durability of natural stone, especially freshwater limestone, so a corrected test method should be proposed. It is obvious that we have a problem with laboratory tests of the frost resistance of freshwater limestones that are not in accordance with the behavior of this stone in real conditions over tens or even hundreds of years, since there is no accepted test method that fully reproduces the natural conditions in which freshwater limestones are found in free atmosphere. The best evidence of its qualities and durability is the stone incorporated in numerous buildings from the near and distant past. Thus, for example, although made from the youngest sediments of the freshwater limestone *(Posušje) miljevina*, the Roman sarcophagus from Vinjani near Posušje, two millennia old, made in shallow rustic of regular shapes, as well as the groups of stećak tombstones at the site of Ričina near Posušje, half a millennium old, resist the test of time and harsh climatic influences of harsh winters with cold north wind for so long, while heavy rainfall (raindrops and hail) can also cause a number of damages to the stone, aggravated by low temperatures and atmospheric pollution. There is also the influence of hot and dry summers with large daily temperature expansions, and the chemical action of rainwater (corrosion), especially at air temperatures slightly above zero, because then the water contains the most dissolved CO_2, or it has the lowest pH-value.

Bearing in mind the combination of the above climatic parameters, the general condition of buildings and monuments made of freshwater limestone in Mostar and Herzegovina (and beyond) is relatively good, even that of stećak tombstones made of *(Posušje) miljevina* in Ričina near Posušje, in harsh climatic conditions for hundreds of years, which indicates not only resistance to frost, but yet more broadly indicates the overall durability/stability of this stone. The deterioration of the analyzed local freshwater limestones is such that some terms related to the deterioration of the built-in stone are not correct, such as: "damage", "disintegration", "decomposition", "degradation", "deterioration" and "decay", but the term "change" of stone is more appropriate, since every stone after extraction and installation is subject to aging or to certain changes.

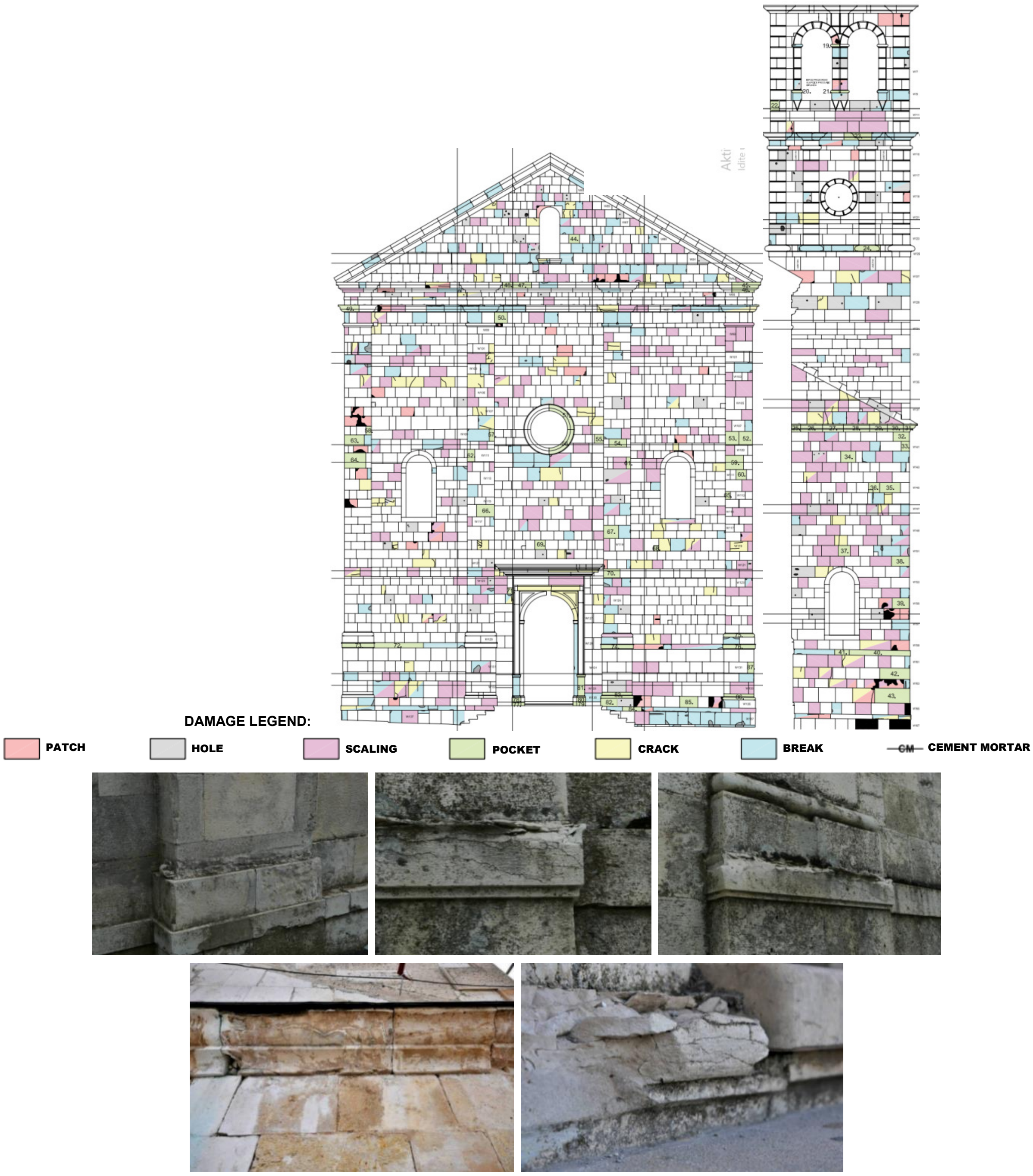



Figures 29-30. Different types of damage to tombstones made of freshwater limestone muljika at the cemetery in Livno, the lower part of which is destroyed by the action of frost, (photo: K. Šaravanja), [21]

The best evidence of the properties and durability of the analyzed freshwater limestones is the stone incorporated in numerous buildings from the near and distant past.

Although made from the youngest sediments of the freshwater limestone *(Posušje) miljevina*, the Roman sarcophagus from Vinjani near Posušje, two millennia old, made in shallow rustic of regular shapes, as well as 3 groups of stećak tombstones at the site of Ričina near Posušje, half a millennium old, resist the test of time and harsh climatic influences of harsh winters with cold north wind and heavy rainfall.

Of course, there is also damage caused by frost, where the damaged parts were replaced. Such an example is the facade of the church of St. Peter and Paul in Gorica in Livno, where mineralogical and petrographic analysis of the stone established that several varieties of limestone from the same deposit were incorporated (pedogenetically altered limestone of the packstone-grainstone type). Of the harmful salts in the stone, the existence of harmful sulfates, namely calcium sulfate (gypsum), with a concentration at the limit of harmfulness, has been sporadically proven, so it could contribute a little to damage to the stone. The most severe damage to the stone is caused by cycles of freezing and thawing of water that enters the stone through cracks, and between the blocks through damaged joints. As the stone does not have a high water absorption, the damage is not evenly distributed, but occurred in places where there was most likely a crack or where through a damaged joint water remained deeper and longer in the stone, and the freezing caused new cracking and expansion of the crack and the damage. Due to the very pronounced daily temperature changes on the facade, in the summer months there was damage to the lime joints and stresses on the microcracks in the stone, which in the winter allowed the penetration of water and the daily cycles of freezing and thawing. Due to the insufficient height of the sheet metal edging on parts of the cornices of the western facade and bell tower, due to heavy rains or melting of snow, water penetrated above the sheet metal into the stone, passed through the wall cover and through the joints of stone blocks emerged even several meters below the cornice. At the copper sheet metalwork on the edges of the wall coverings, due to oxidation or formation of patina, the corrosion products of the copper sheets were washed and poured down, forming green stains on the surface of the stone, [7].

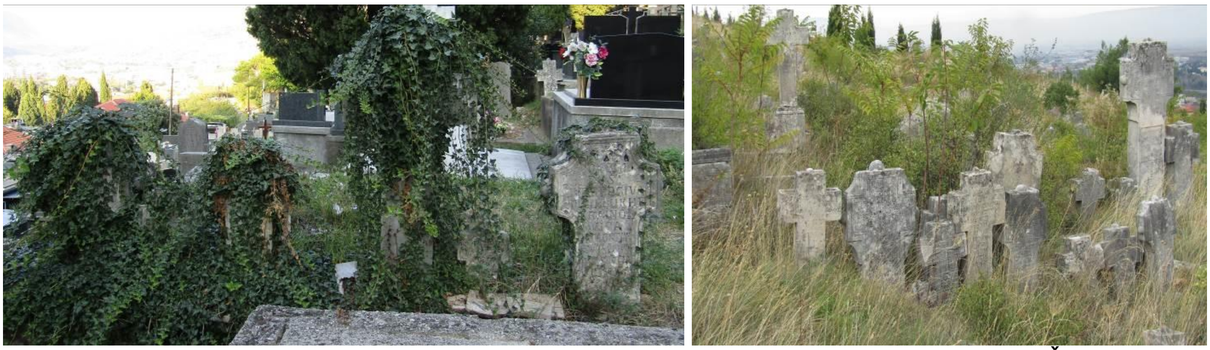


Figures 31-37. Scheme of damage to the western facade (left) and the western facade of the southern bell tower of the Church of St. Peter and Paul on Gorica in Livno (right) (made by: Croatian Restoration Institute, Zagreb, 2011); The disappearance of certain parts of the pilaster base due to severe damage caused by aggressive climatic impacts; Completely destroyed parts of the dividing cornice at the top of the plinth on the western facade of the church; Deposits of low weeds in the form of algae, lichens and mosses are also observed (figures above); Deterioration of the stone plastic of the dividing cornice on the western side of the southern bell tower due to the penetration of atmospheric agents and the destruction of the stone by freezing processes (below, left); Detail of profiled stone plastic on which cracks, flaking and breaking off of stone material have occurred (below, right), [7]

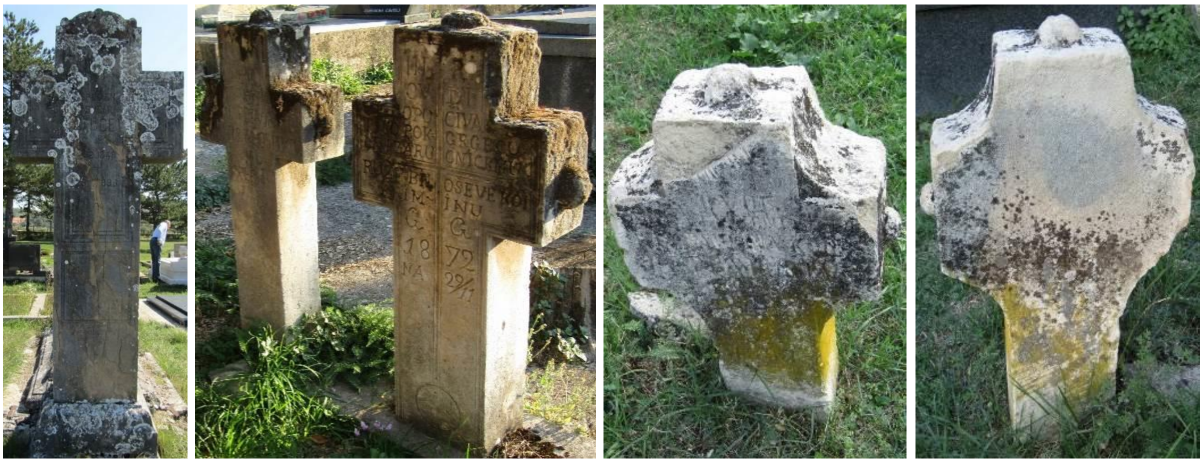
2.6 Organic pollution (biological covering)/biological colonization

Biological covering or biological colonization is often present on stone surfaces exposed to rain. It needs to be removed, especially higher plants in the form of tree-like, bushy and/or grassy outgrowths, grown from joints and cracks in the stone, into which wind and rain bring leaves and earth over time. They not only obscure the view of the monuments, but by developing roots they penetrate the structure of the stone and create high pressure and physically destroy the stone. The transfer of nutrients from the roots to the plant affects the carbonate structures in stone. Other degradation processes on stone are also possible. Biological covering retains water, which can cause other forms of stone corrosion.

On surfaces exposed to rain, stone is often contaminated with biological pollution in the form of lower plants - microscopic fungi, lichens, bacteria, molds, algae and mosses, which we recognize as coatings of different colors (black, greenish, yellow or brown), most often as grayish-black accumulations on the stone, which turn green in contact with moisture; lichens are yellowish-brown in color, [14], [26].



Figures 38-39. Ivy vegetation as an example of biological covering of tombstones in Šoinovac cemetery in Mostar (left); Tombstones in the old part of the Bjelušine cemetery in Mostar (right), which is neglected and overgrown with low wild vegetation, and a large number of monuments are overgrown with lichen and moss. During the war, all tall vegetation was removed from the cemetery area, which to a certain extent had previously protected the monuments from the sun and western winter winds, (photo: K. Šaravanja), [21]



Figures 40-43. Covering of tombstones made of miljevina with biological cover in the cemetery of St. Ivo in Livno (left) and in the Šoinovac cemetery in Mostar (other 3 figures); Biological covering (lichens, moss, fungi,...) use stone as habitat and/or food, (photo: K. Šaravanja), [21]

The influence of all low plants, especially microscopic fungi and lichens, is manifested in a wide range of harmful effects on the stone epidermis, [27]. Through metabolic processes, they produce various organic and inorganic acids, causing chemical destruction (dissolution) of the limestone surface and loss of the surface layer, which is why this kind of stone is lighter in color after removal of the colonies. We are talking about a number of harmful effects on the stone surface, from visual deterioration and loss of finishing details in some places, to difficult legibility (especially when the stone is soaked with rain).

On certain parts of the facades algae and lichens were visible to the naked eye, while in some places in dry climate conditions they imperceptibly cover the entire façade, [28].

The most persistent and resistant of these organisms are lichens, yellowish-brown in color, which creep into the pore space of the stone, deep between the mineral zones, and are able to destroy/lower the surface of the stone, which is visually deteriorated, the sharpness of the relief is wiped off, and legibility is difficult, while the finishing details in some places are irretrievably lost. They develop in conditions of high humidity, although they can develop with very little humidity and survive at low temperatures. Their negative influence is manifested in a wide range of harmful effects on the surface layer of stone, which becomes less resistant to other harmful agents and weathering factors, which leads to faster deterioration of the monument. Their hyphae can penetrate very deeply into the structure of the stone, creating a dense network of tiny holes, so the stone begins to deteriorate faster under the action of weathering factors (scaling of stone layers). They are most often found on the north side, where the moisture in the stone stays the longest, [2].

With their metabolic activity, bacterial populations and algae produce polymers, a slimy mass that protects them from drying, radiation and erosion, taking suspended particles from the air and forming hard deposits and patinas on the stone. The increase in biomass on stone surfaces stimulates the production of nutrients, which enables the colonization of other microorganisms, thus accelerating the processes of biological decomposition of stone, [29]. However, the most harmful for stone are endolithic lichens that produce the most corrosive substances in the form of organic and inorganic acids, [26], [30]. They penetrate into the pore space of the stone by several millimeters. This lowers the surface of the stone, wipes out the sharpness of the relief, and opens the way to other degradation processes.

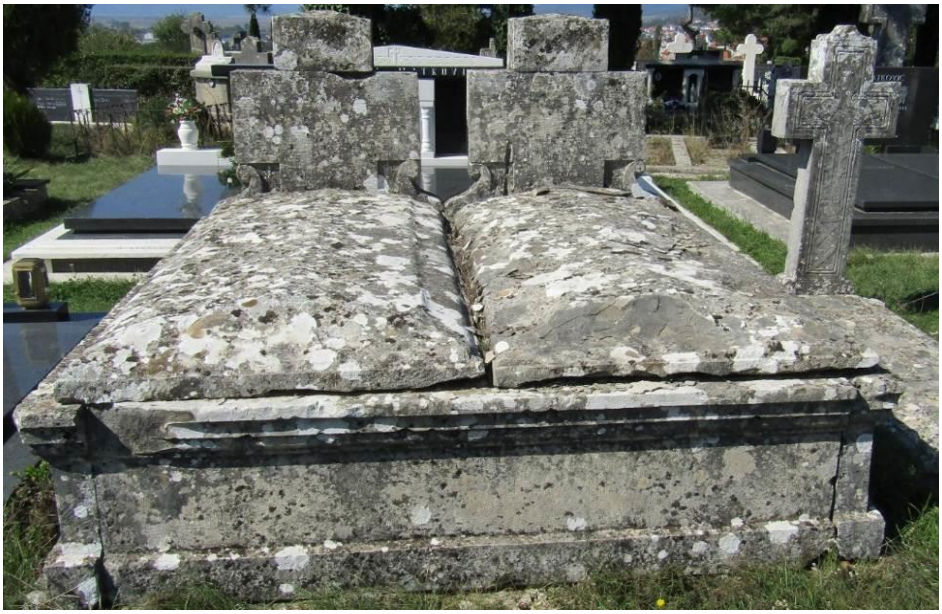
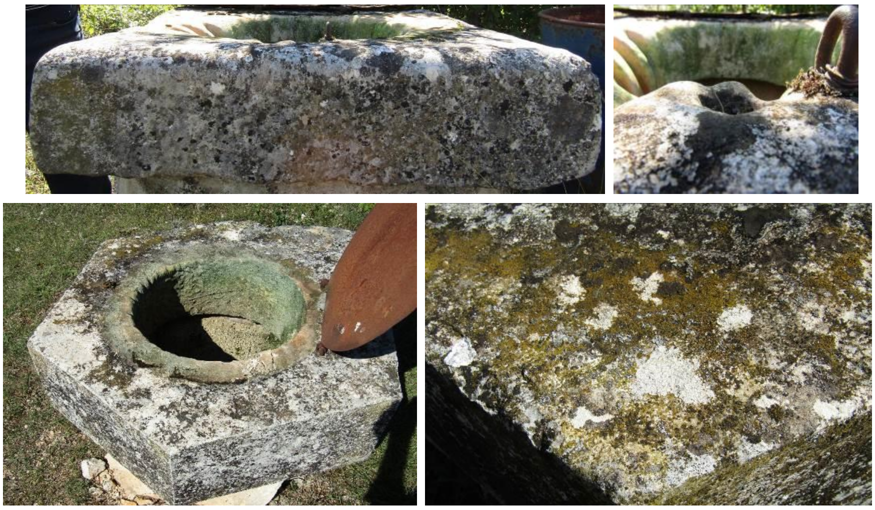


Figure 44. Lichens and mosses covering tombstones in the cemetery of St. Ivo in Livno, (photo: K. Šaravanja), [21]

Figures 45-47. Lichens and mosses covering tombstones made of miljevina in the Kolo cemetery near Tomislavgrad. The cover plate made of freshwater limestone, 200 cm long, 50-80 cm wide, and about 5 cm thick, is interesting as a certain proof of its stability in harsher climatic conditions. Engraved motifs are very rarely damaged. It should also be borne in mind that the cemeteries used to be forested with pines and oaks, which today are present much less or not at all, and this influence of trees on the stone of the monuments is very difficult to retroactively analyze, (photo: K. Šaravanja), [21]

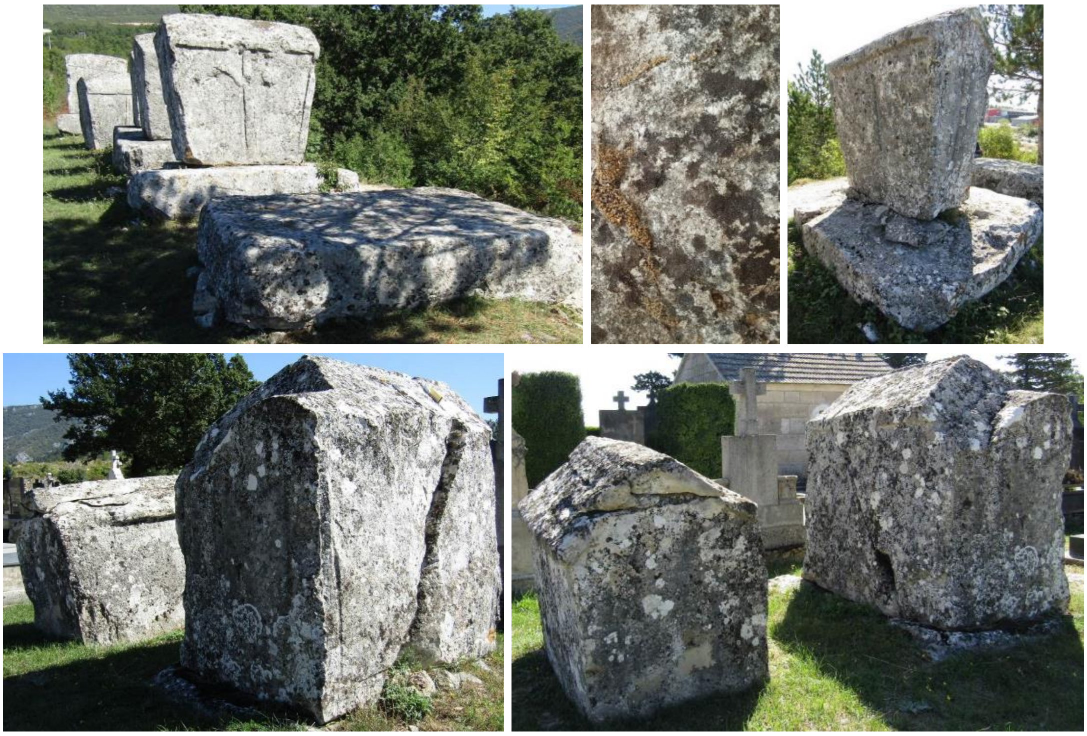


Figures 48-51. The crowns of stone cisterns made of (Posušje) miljevina covered with lichens and moss at the location of the Kovači hamlet, southwest of Posušje, with details, (photo: K. Šaravanja), [21]

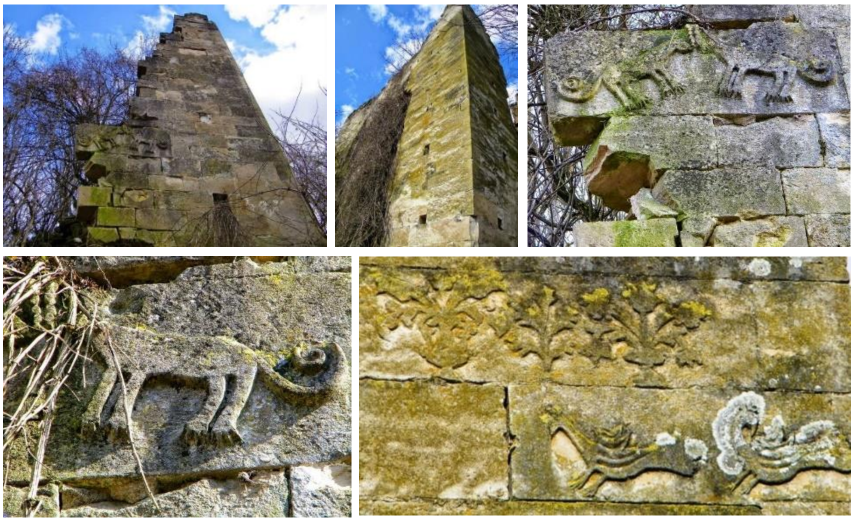
As part of the microflora conservation and restoration studies (2014-2016) on stone stećak tombstones from the locality of Crljivica near Cista Velika, field studies and collection of lichen cover from the surface of the stećak tombstones were carried out, where as many as 18 different species were found, with 5 dominant species. Several species of mosses were observed on the surface of the stećak tombstones, and a wide spectrum of bacteria, fungi, molds, cyanobacteria and green algae was registered by sampling and sowing on nutrient media. Their distribution and total frequency have not been analyzed in detail as for lichens. Both laboratory and in situ tests of the effectiveness of available commercial biocides were carried out, [29].

A visual inspection of the surface of the stećak tombstones at the Ričina site near Posušje, which is not too far from the stećak tombstones from the Crljivica site, shows a similar wide range of microorganisms, which should be investigated. Still, the general condition of stećak tombstones made of *(Posušje) miljevina* in Ričina near Posušje is relatively good, bearing in mind the harsh climate conditions to which they were exposed for hundreds of years, which indicates not only the resistance to frost, but even more broadly indicates the overall durability/stability of this and other freshwater limestones.

Figure 52. Part of an orthophoto with the positions of 3 groups of stećak tombstones on Ričina near Posušje, [31]



Figures 53-57. Seven stećak tombstones of the main group of stećak tombstones, marked "1" in Figure 52 (figures above) and two significant and preserved gabled stećak tombstones as part of the modern cemetery, marked "3" in Figure 52 (figures below) made of (Posušje) miljevina on Ričina near Posušje; A wide range of microorganisms (lichens, bacteria, cyanobacteria and fungi) is visible on the stećak tombstones, (photo: K. Šaravanja), [21]



Figures 58-62. A good example of preservation of freshwater limestones miljevina in the remains of the Čengić bays' tower in the village of Rataji near Miljevina in eastern Bosnia. In addition to the patina, the presence of biological covering in the form of moss, lichens and fungi is noticeable, (photo: Zaim Bešović, 2015)

3. CONCLUSIONS

Freshwater limestones are incorporated in numerous stone buildings and/or monuments, where complex processes of changes, or weathering (degradation) of stone occur as a result of its interaction with various environmental conditions (external factors): the effects of temperature, humidity and quality of air, winds, precipitation, acidic rain, exhaust gases, micro-particles and organisms in the air and water, harmful salts, radiation, soil, biological colonization, along with the destructive action of man.

Geologically young formations, as compared to geologically older ones, compact (dense) limestones, are less crystalline, less dense, and thereby less resistant to the processes of chemical degradation (dissolution and sulfation) and deterioration caused by crystallization (salts and frost).

In addition to diagenesis, their durability is also affected by the method of cutting and installation of stone blocks, so stratification and secondary fracturing must be taken into account during processing.

Old builders used analyzed local types of freshwater limestones in construction, knowing exactly their possibilities and advantages related to the exceptionally easy processing and realization of more delicate buildings, but also limitations related to considerable to high porosity, small bulk density, high water absorption, relatively low strength, poor resistance to wear and questionable resistance to frost.

Due to the high degree of porosity and the slow release of moisture, water fills all the pores and the basic material itself, which significantly affects the strength. With water-saturated *(Mostar) miljevina*, water eliminates the influence of other factors and is the only factor influencing strength. Therefore, it was desirable for it to be installed on surfaces that are not directly exposed to rain and gusts of wind, so the surfaces of residential, religious and public buildings in Mostar made of *miljevina*, which are exposed to wetting and moisture, are protected by facades, with the application of another type of stone in the lower part of the building that is in contact with the ground (for foundations and basement), or more recently they are protected with highly water-repellent impregnating agents.

In contrast to *Mostar miljevina*, numerous residential and commercial buildings in the municipality of Posušje were built with facades of *(Posušje) miljevina*, similar to *tenelija* in Mostar.

After carrying out appropriate laboratory tests of the analyzed freshwater limestones, frost resistance/stability is questionable to say the least, especially since there is no accepted test method that fully reproduces the natural conditions in which freshwater limestones are found in free atmosphere, for tens and hundreds of years. Thus, for example, although made from the youngest sediments of the freshwater limestone *(Posušje) miljevina*, the Roman sarcophagus from Vinjani near Posušje, two millennia old, made in shallow rustic of regular shapes, as well as the groups of stećak tombstones at the site of Ričina near Posušje, half a millennium old, resist the test of time and harsh climatic influences of harsh winters with cold north wind for so long, while heavy rainfall (raindrops and hail) can also cause a number of damages to the stone, aggravated by low temperatures and atmospheric pollution. There is also the influence of hot and dry summers with large daily temperature expansions, and the chemical action of rainwater (corrosion), especially at air temperatures slightly above zero, because then the water contains the most dissolved CO_2, or it has the lowest pH-value. Bearing in mind the combination of the above climatic parameters, the general condition of buildings and monuments made of freshwater limestone in Mostar and Herzegovina (and beyond) is relatively good, even that of stećak tombstones made of *(Posušje) miljevina* in Ričina near Posušje, in harsh climatic conditions for hundreds of years, which indicates not only resistance to frost, but yet more broadly indicates the overall durability/stability of this stone. The deterioration of the analyzed local freshwater limestones is such that some terms related to the deterioration of the built-in stone are not correct, such as: "damage", "disintegration", "decomposition", "degradation", "deterioration" and "decay", but the term "change" of stone is more appropriate, since every stone after extraction and installation is subject to aging or to certain changes.

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