

Hygric properties of Euville limestone

Beatrice Cerrai^{*} , Katrin Schollbach , H.J.H. Brouwers

Department of the Built Environment, Eindhoven University of Technology, P.O. Box 513, Eindhoven 5600 MB, the Netherlands

ARTICLE INFO

Keywords:

Limestone
Porosity
Water transport
Degradation

ABSTRACT

Understanding how microstructural characteristics influence water transport and durability in natural building stones is crucial for predicting their long-term performance and guiding conservation practices. This study investigates the hygric behaviour of the Euville limestone, a building stone widely used in European architecture, through an integrated experimental approach which combines porosity, vapour diffusion, water absorption, and drying analyses. While prior studies have addressed the bulk properties of this stone, this work explicitly characterises the anisotropy of these properties due to sedimentary layers. The results show that Euville limestone has a high water absorption capacity (up to 6 wt%) and a considerable difference between total (14%) and open porosity (12%). Pronounced anisotropy in capillary absorption was observed, with higher uptake parallel to the bedding planes, while compressive strength remained nearly isotropic. The prolonged drying (14 days) highlights the tendency of the material to retain moisture.

1. Introduction

Sedimentary rocks, particularly limestones, have historically been popular construction materials that are incorporated in buildings as interior and exterior masonry or cladding, having either a decorative or structural use [1]. Euville limestone is a variety widely employed in France [2], Belgium [3], the Netherlands, and in Germany [4] for historical and contemporary architecture since the late 19th century. This limestone is made of crinoid fossils which are bonded together by a carbonate cement [5]. The stone has a homogeneous appearance, presenting a white-beige colour and a homogenous distribution of the grains. The sedimentary layering is not visible. The grain cohesion, providing the stone strength, is determined by the bonding between the grains and the cement. Due to its microstructure, this limestone is characterised by a high open porosity, which influences its interaction with moisture and durability [6].

The deterioration of limestones is intrinsically linked to the role of water as a transport and reaction medium [7,8]. Specifically for Euville, the weathering effect is represented macroscopically as either grain loss or a progressive internal weakening, where the stone appears intact due to a hardened external crust, but the interior has lost most of its strength. The severity of such deterioration is dictated by the petrographic characteristics of the stone. Properties such as the dimension and distribution of the grains constituting the stone and their packing, which creates the pore network, regulate capillary suction, moisture retention time,

and susceptibility to freeze-thaw cycles [9–12]. Further cracking and efflorescence formation are caused by the crystallisation of soluble salts, which is governed by the hygric behaviour [13]. Additionally, moisture retention promotes biological colonisation, which can affect the interaction of the material surface with the environment and induce aesthetic changes [14]. Therefore, a thorough characterisation of the hygro-thermal behaviour is crucial for understanding the durability of Euville limestone.

Euville limestone has been the subject of several petrophysical studies. For instance, Bellanger *et al.* [10] examined the frost resistance of carbonate grainstones, including Euville, focusing on water storage and transport. In the context of Heritage Conservation, the stone's compatibility with repair mortars was evaluated by Lopez-Arce *et al.* [2], providing data on porosity, strength, and vapour permeability. Both studies report similar results for open porosity, but other results are not comparable because they have measured different quantities. However, neither studies include the potential influence of sedimentary layers, as information about the stones orientation after cutting is often not available and difficult to determine later on.

The water absorption of Euville has been evaluated in relation to bedding orientation by Pestre *et al.* [15] and resulted in a substantial difference. However, there is no comprehensive research that addresses the directional anisotropy in water transport for Euville limestone and a significant knowledge gap remains regarding the integrated moisture behaviour of the stone.

^{*} Corresponding author.

E-mail address: b.cerrai@tue.nl (B. Cerrai).

<https://doi.org/10.1016/j.conbuildmat.2026.147365>

Received 30 December 2025; Received in revised form 9 June 2026; Accepted 4 July 2026

Available online 7 July 2026

0950-0618/© 2026 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

This study addresses this gap by providing a comprehensive characterisation that correlates sedimentary layer orientation with mechanical strength as well as water transport, offering new insights into how the microstructure and sedimentary layers influence its properties. Drying kinetics and water vapour permeability of the stone are assessed and related to its porosity to better characterise the moisture behaviour. Water adsorption rates are estimated using the Karsten tube, a standard tool used in conservation practice [16,17].

2. Materials and methods

2.1. Material

The samples investigated in this work have been collected from the construction company *Vandekerckhove NV* (Belgium). The sedimentation layering orientation was marked on the stones when they were extracted from the quarry. The depositional layers are indicated on each block by two or three parallel lines drawn on the block itself. The intact blocks of approximately $20 \times 30 \times 15 \text{ cm}^3$ have been cut into $5 \times 5 \times 5 \text{ cm}^3$ cubes for mechanical and hygric testing. A picture of the intact blocks used in this research is presented in Fig. 1.a.

Alongside the blocks picture, the optical microscope images (Fig. 1.b and Fig. 1.c) show the microstructure of the Euville limestone. This stone is classified as a Middle Oxfordian biosparite (grainstone) extracted from north-eastern France [18]. It is characterised by crinoid fossil fragments of calcite (CaCO_3) with average dimensions of 2 mm [3] which are bonded by a syntaxial calcite cement [5]. The grain cohesion, providing the stone strength, is determined by the bonding between the grains and the cement. The internal architecture is articulated in sedimentary layers, which, resulting from its depositional history, create the microstructural anisotropy that is the focus of the transport analyses presented in this study. An example of the packing between the grain and the cement is presented in Fig. 1.b and in Fig. 1.c. Those are microscope pictures of a thin section made from the intact blocks collected. The fossil grains are surrounded by the calcitic cement, which grows directly from the grain's surface, creating both optical and structural continuity between the two phases. While macroscopically uniform, the calcitic cement is susceptible to dissolution and recrystallisation. This degrades the grain-cement interface, leading to granular disintegration and reduced structural integrity [9,19]. Such transformation determines grain detachment and, as a result, the macroscopic crumbling of the stone, which is the unique degradation pattern observed for this limestone.

2.2. Methodology

The total porosity was derived after the measurement of ρ_s (skeletal density) using a helium pycnometer (Micromeritics AccuPyc II 1340).

For the skeletal density measurement, 8 intact Euville samples have been examined. They were dried and crushed in an agate mortar to approximately 5 g of powder and measured with the pycnometer.

The total porosity p_{tot} has been determined following the equation:

$$p_{tot} = 1 - \frac{\rho_b}{\rho_s} \quad (1)$$

Where ρ_b is the bulk density, calculated as the ratio between m_{dry} and V of the 12 cubic blocks examined in this study. To evaluate the hygric transport properties, it is necessary to distinguish between water-accessible porosity and total porosity. The latter refers to all voids in the solid matrix, including closed pores. The porosity accessible to water, or p_{open} , corresponds to the percentage of material that can be occupied by water. It has been calculated on the 12 blocks of $5 \times 5 \times 5 \text{ cm}^3$ after the water absorption test, following the formula:

$$p_{open} = \frac{(m_{sat} - m_{dry})/\rho_w}{V} \quad (2)$$

The open porosity was also assessed using Mercury Intrusion Porosimetry (MIP) on small samples measuring approximately 5 mm and weighing 2 g each. A representative sample was dried at 70°C before performing the MIP measurement with an Autopore IV. The analysis was conducted using a pressure range from 0.0141 MPa to 206.81 MPa, allowing for the detection of pore throat diameters from approximately 88.35 μm down to 0.0066 μm . The results obtained by MIP are compared with the open porosity value calculated using Eq. (2).

The masonry blocks of Fig. 1.a were cut into $5 \times 5 \times 5 \text{ cm}^3$ cubes, using a water-cooled diamond saw, and distributed for the different experiments. Before testing, all of the prepared blocks were dried in an oven at 70°C until mass consistency. After drying, the specimens were cooled to ambient temperature in a desiccator over silica gel to prevent moisture absorption.

A set of ten cubes was used to estimate the Uniaxial Compressive Strength (UCS) of the limestone. The measurement has been performed following the procedure of the standard EN 1926 [20] and performed with a 250 kN Tension-Compression machine Instron. The compressive strength has been considered as a characterisation test and to address the presence of anisotropy for this property.

The moisture transport has been studied by the water vapour diffusion resistance factor (μ , adimensional). This value is determined employing the cups method as defined in the standard ISO 12572 [21]. For this experiment, ten samples of Euville of 1 cm thickness and an area of $5 \times 5 \text{ cm}^2$ have been cut with a water-cooled diamond saw. The testing cups contain either a desiccant (calcium chloride, dry cup) or water (wet cup). The assembly of samples, which is shown in Fig. 2, resulted in ten cups, five of which were for the dry and the others for the wet cup

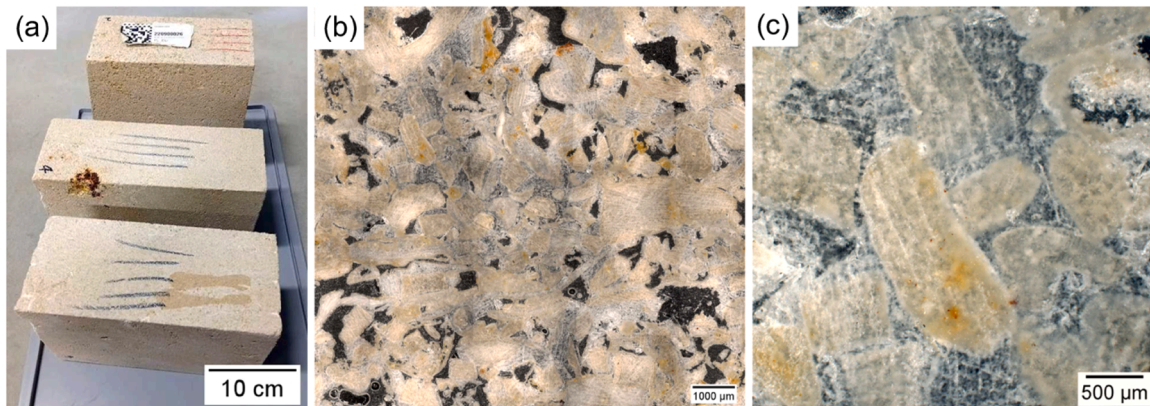


Fig. 1. (a) Blocks of intact Euville, used to prepare the sample cubic blocks; (b) and (c) optical microscope pictures of Euville limestone thin section.

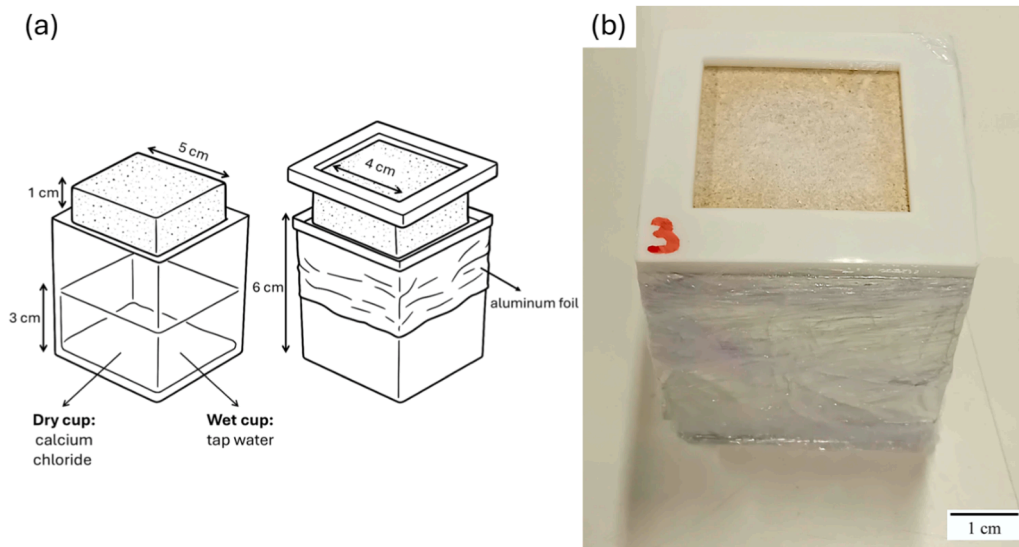


Fig. 2. (a) Schematic diagram of the water vapour permeability experiment with the wet and dry cups method; (b) picture of the sample assembly.

experiments. Fig. 2.a shows how the sample cups were constructed: the cups were filled with either calcium chloride or water up to a thickness of approximately 3 cm from the bottom of the cup. Then the stone sample is placed and covered with a lid-frame, which exposes an area of $4 \times 4 \text{ cm}^2$ for vapour exchange between the material and the environment. Such a prepared assembly was wrapped in one layer of aluminium foil (approximately 0.05 mm thickness), and lastly, a plastic foil wrap was created on top of the aluminium foil in order to seal the whole system. All prepared cups were placed in a climate chamber at $20 \pm 1^\circ \text{C}$ and $60 \pm 2\% \text{ RH}$. The weights of the test assemblies are monitored every 24 h until five successive calculations of the mass change are constant within $\pm 5\%$ of the mean value for the single specimen weight.

Anisotropy in the water transport has been investigated by performing water absorption tests on limestone cubes of $5 \times 5 \times 5 \text{ cm}^3$. All the water absorption experiments were performed in a controlled laboratory environment at $20 \pm 2^\circ \text{C}$ and $50 \pm 5\%$ relative humidity. The water absorption at atmospheric pressure and the water absorption by capillarity have been determined by observing the procedures described in the standards EN 13755 [22] and EN 1925 [23]. In the following

sections, each block is indicated by a number and a letter, which indicate the orientation of the sediment layers, either vertical (“V”) or horizontal (“H”) and the sample number. The presented study aims to evaluate the eventual influence of the orientation of sedimentary layers on the water transport properties. Specifically, of 12 samples, 6 blocks have been positioned with sedimentary planes parallel to the water level (H), and the other 6, perpendicular to it (V). The same set has been kept for both total and capillary water absorption experiments. The diagram shown in Fig. 3 illustrates the positioning of the samples during the water absorption experiments.

After performing the capillary absorption experiment, the water-saturated samples were wrapped in one layer of aluminium foil, leaving only one surface where evaporation was possible. Furthermore, the samples were wrapped again in plastic foil to ensure good adhesion of the aluminium to the stone. Such prepared samples were then placed in a climate-controlled chamber, which was kept at $20 \pm 1^\circ \text{C}$ and $60 \pm 2\% \text{ RH}$. The specimens were placed at approximately 5 cm distance from each other to ensure uniform vapour diffusion, and their weight was monitored until the mass was stable (defined as a mass change of less

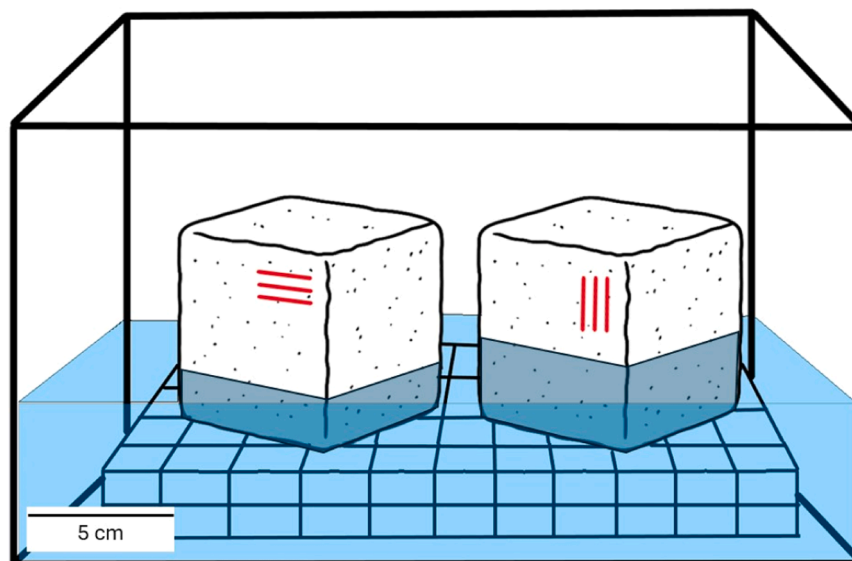


Fig. 3. Schematic diagram of the water absorption experiment by capillary action. The stone block on the left has the stratification planes (indicated by three red lines) oriented perpendicular to the water absorption direction (H), while the block on the right has them oriented parallel (V).

than 0.01% over a 24-hour interval). The drying properties of the material have been determined following the procedures in the standard EN 16322 [24]. A representative diagram of the blocks prepared for the drying test is proposed in Fig. 4.a.

The Karsten tube water penetration test was performed to provide a comparison with the laboratory-standardised experiments for water absorption, as it is a common, non-destructive and portable technique. The instrument consists of a graduated tube welded to the lower end of a cylinder cell of 30 mm diameter that is sealed to the block surface with plasticine putty produced by Mohren. Each tube was filled with 4 mL of tap water, and the water level was monitored until complete absorption. This comparative approach allows for the validation of simplified on-site tools against laboratory data obtained with standardised experiments. A schematic diagram representing the two different conditions in which the experiment has been conducted is presented in Fig. 4.b and Fig. 4.c.

3. Results and discussion

3.1. Porosity and compressive strength

A summary of the calculated and measured densities together with the results of the porosity study is presented in Table 1.

The average total porosity p_{tot} is $14.16 \pm 0.68\%$, with 12.98% as minimum and 15.21% as maximum based on measuring 12 samples. The calculated coefficient of variation is 4% and together with a variance of 0.47, it indicates low relative variability of the data.

The open porosity p_{open} has been determined from the total water absorption test as explained in 2.2 and resulted in an average of $12.57 \pm 0.55\%$, with 11.79% and 14.09% as minimum and maximum. For the

sample set, the coefficient of variation is 4%, and the variance is 0.30, demonstrating the good consistency of the results. In the porosity study of Euville by Leger and Luquot [25], the difference between open and total porosity indicates the existence of closed pores. Unexpectedly, p_{open} from MIP is only 7.14%, a difference with p_{open} by water absorption indicates the presence of open voids or cracks that are outside the MIP measurement range (upper limit is $88.35 \mu\text{m}$ pores). Such porosity is visible in Fig. 2.b and can explain the difference. The "ink-bottle" effect could also contribute to the observed difference. While water is a wetting fluid that spontaneously penetrates the network via capillary action, mercury is non-wetting and requires high pressure to enter. Salvini *et al.* [26] reported that MIP may underestimate open porosity if the ratio between the pore dimension and the pore throat is high, as mercury might not be completely drained during depressurisation [27]. This leads to an underestimation of the open porosity. Furthermore, the cubes used for water absorption are more representative than the small sample required for MIP.

Table 1

Density and porosity of Euville limestone. The MIP open porosity was calculated on a single representative sample, therefore, the standard deviation is not specified.

Skeletal density ρ_s (g/cm ³)	Bulk density ρ_b (g/cm ³)	Total porosity p_{tot} (%)	p_{open} (%) by water absorption	p_{open} (%) by MIP
2.54 ± 0.04	2.17 ± 0.02	14.16 ± 0.68	12.57 ± 0.55	7.14

In the literature, the open porosity of Euville appears with a wide range of values, from 10% to 20% [3,10,28,29]. This variability is not surprising due to the heterogeneity of the stone, which is extracted from different sections of the quarry.

The pore size distribution resulting from MIP is presented in Fig. 5. The pore sizes show a bimodal distribution, with a maximum around $1 \mu\text{m}$ and $40 \mu\text{m}$. The distribution matches the results of Leger and Luquot [29] and Bellanger *et al.* [10], who classified the larger pores as resulting from intergranular porosity. The bimodal distribution, together with the high ratio between maximum and minimum radii, indicates the presence of a complex pore network. This configuration facilitates dual transport mechanisms: small pores control capillary rise and vapour sorption, whereas large pores enable faster liquid transport and storage capacity. The gap between open and total porosity values confirms the presence of closed or semi-closed pores, which may act as a water reservoir. During freeze–thaw cycles, these closed voids contribute to internal pressure build-up and microcracking, explaining why Euville might be subjected to frost damage comparable to that of more porous limestones reported in the literature [15,30].

When estimating the durability of a stone as construction material, both porosity and strength should be considered [6]. Limestones have compressive strength ranging from 6 to 130 MPa and porosity going from 0% to even 50% [15].

The compressive strength of Euville varies between 12.4 MPa and 38.3 MPa [31], which is consistent with other UCS presented for biocalcarenes [30]. Among the 10 tested samples, the average compressive strength is 27.53 ± 3.26 MPa. Samples having the sedimentary layers oriented parallel to the applied force direction (Fig. 6.a) have an average of 27.99 ± 3.03 MPa, while the perpendicular ones (Fig. 6.b) of 27.07 ± 3.78 MPa. While a numerical increase is noted for the perpendicular orientation, the standard deviation overlaps, suggesting that the Euville limestone does not exhibit mechanical anisotropy. A two-sample *t*-test ($\alpha = 0.05$) produced $t = 0.48$ and $p = 0.65$, showing that the difference between the two orientations is not statistically significant and is likely related to natural variability rather than anisotropy. In addition, the pattern created by the cracks does not seem to recall the stratification planes disposition, but instead, the cracks created by the applied

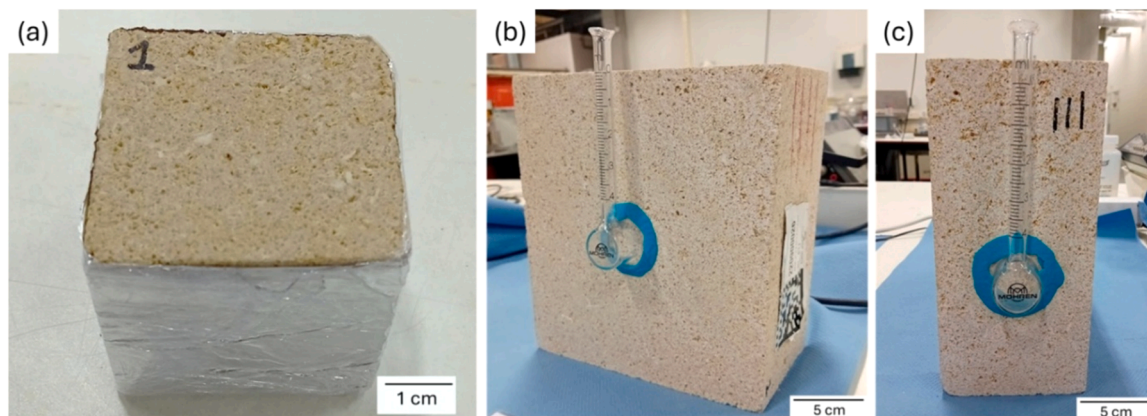


Fig. 4. (a) Picture of the samples assembly for the drying experiment. On the right, the Karsten tube experiment set-up: in (b), the water absorption direction is perpendicular to the stratification planes, which are indicated by three parallel lines. In (c), the water absorption plane is parallel to the stratification planes.

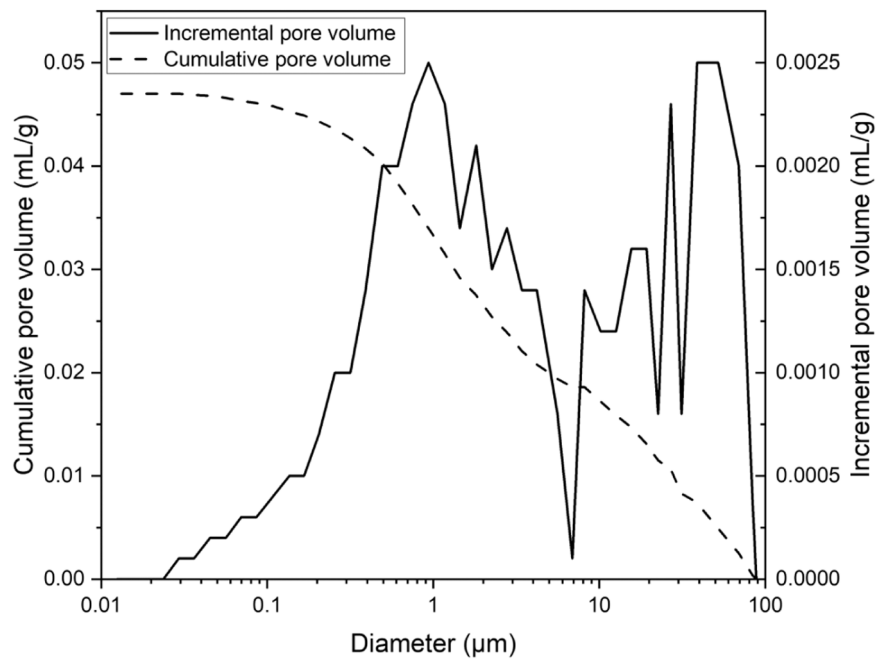


Fig. 5. MIP pore size distribution. In the graph are displayed both the cumulative and the incremental pore volume as a function of the pore diameter.

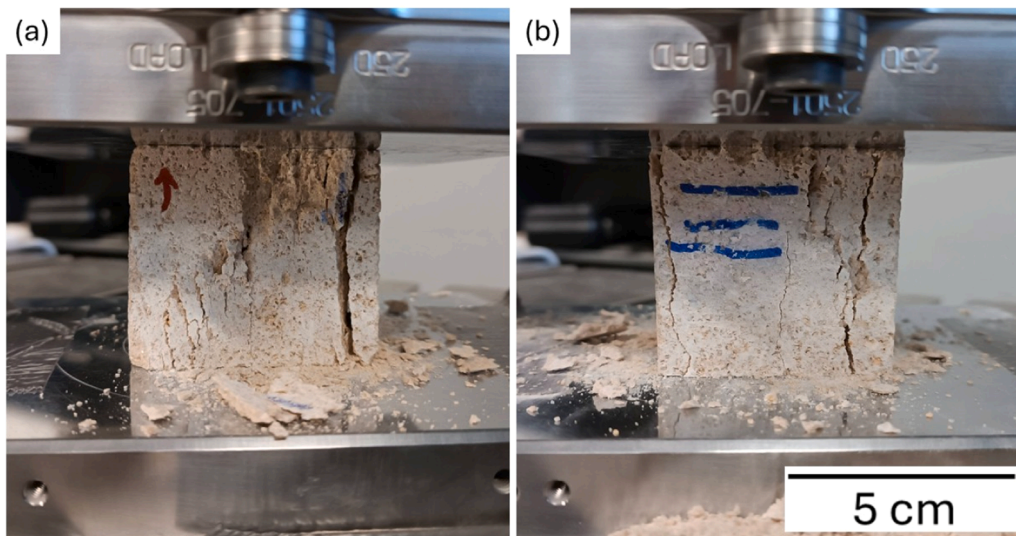


Fig. 6. UCS test pictures. In (a), the stratification planes are oriented parallel to the compressive force (V). In (b), the sedimentary planes are perpendicular to the direction of the compressive force (H).

pressure have the same orientation (Fig. 5). This behaviour indicates that the sedimentary layering does not function as a primary plane of mechanical weakness. But it can eventually be ascribed to the deterioration of the syntaxial calcite cement. The cement grows in structural continuity with the crinoid fragments, creating a robust, interlocking mineral matrix that effectively merges the depositional layers. Consequently, the stone maintains a high degree of structural homogeneity under compressive loading.

3.2. Vapour transport and drying properties

The water vapour permeability assessment with the dry and wet cup method allowed for the determination of the moisture flow rate in the samples, resulting in an average vapour diffusion resistance factor of $\mu_{\text{wet}} = 21.58 \pm 4.63$ and $\mu_{\text{dry}} = 35.78 \pm 5.58$ for the wet and the dry cup,

respectively. For the wet cup experiment, the minimum calculated value is 13.77, while the maximum is 24.95, and the coefficient of variance for the five analysed cups is 21%. At the same time, the results obtained for the dry cup range from 30.06 to 42.20, and the coefficient of variance for the five calculated values is 16%. Such high values of variability might be attributed both to the natural origin of the material and the relatively small size of the sample, which can produce different results.

The calculated water vapour permeabilities are $5.46 \times 10^{-12} \pm 5.74 \times 10^{-13}$ kg/m·s·Pa for the dry cup and $8.53 \times 10^{-12} \pm 6.88 \times 10^{-13}$ kg/m·s·Pa for the wet cup experiment. The wet cup experiment coefficient of variation is very low with 6%, as the calculated values range from 8.01×10^{-12} kg/m·s·Pa to 9.44×10^{-12} kg/m·s·Pa. The dry cup experiment shows higher variation with a coefficient of variation of 13% and 4.74×10^{-12} kg/m·s·Pa and 6.41×10^{-12} kg/m·s·Pa as the minimum and maximum calculated values, respectively.

Even though the resulting values are different from the ones measured by Pestre *et al.* [15], presenting resistance factors of $\mu_{\text{wet}} = 56$ and $\mu_{\text{dry}} = 149$, the relation between the values remains the same, with the dry cup method resulting in higher values than the wet cup one. Specifically, the water vapour resistance factors are higher for the dry cup experiments and the water vapour permeability is higher for the wet cup ones. This effect might be due to the increased water content in the stone during the wet cup experiment. Notably, in the dry cup experiment, there is only vapour diffusion through the material, while for the wet cup experiment, the high moisture content in the cup might favour surface diffusion and capillary condensation, for which the transmission rate of water vapour is increased [32]. As a result, the resistance of the material towards water vapour transmission is lower for the wet cup experiment. On the contrary, for the same reasons, the water vapour permeability results in higher values for the wet cup. In this work, both experiments have been performed in order to get a comprehensive assessment of the water vapour transport behaviour in the Euville limestone. This is because, while with the dry cup the water vapour flow is calculated, with the wet cup, the susceptibility of the material towards moisture retention is assessed.

In addition, limestones similar to Euville (15–20% porosity) present vapour resistance values similar to the calculated ones, while more porous varieties present smaller values of water vapour resistance, but similar water vapour permeabilities [15], thus suggesting the importance of the vapour diffusion resistance factor for building materials characterisation. The water vapour diffusion is an important property of a porous material since it influences its drying behaviour in the final steps of the process.

The drying behaviour of the Euville limestone is discussed here as the process is strongly influenced by the RH of the air. In particular, the removal of water from a wet material is governed by evaporation, which depends on the difference between the material's humidity and that of the environment.

The drying curve correlating the mass of water removed from the samples over time is shown in Fig. 7. The drying process stops when equilibrium between the material and the environment is reached. The

experimental configuration guarantees water evaporating from a single surface. As a result, water rises under the influence of capillary action. The movement of liquid water to the sample surface, where it evaporates, typically governs the initial drying phase. In this phase, the drying rate is influenced by the properties of the liquid, as well as by the microstructure. The drying rate remains constant throughout the first phase, or linear drying period. When the evaporation rate slows down because of liquid water removal and the vapour diffusion process starts, the second drying phase begins. The drying process of the Euville took around 14 days. The mass stabilisation of the samples is represented in Fig. 7 by the convergence of the drying curves towards a horizontal asymptote. The endpoint of the experiment corresponds to the hygroscopic equilibrium moisture content of the stone at the test environmental conditions ($20 \pm 1^\circ\text{C}$, $60 \pm 2\%$ RH). Analysis of the oven-dry and drying experiment final stable mass across all 12 blocks yields a residual moisture content ranging from 0.84 to 1.32 wt%, with a mean of 0.96 ± 0.13 wt% at the end of the test. The apparent convergence toward zero in Fig. 7 is a visual artefact of the graph scale.

The first drying phase is defined by the detection of a linear decrease in water content that appears to end after 4 days. The drying rate for this phase was calculated using the first derivative of the water content with respect to time. The drying rate of the water vapour transmission phase (second stage of drying) is determined as the slope of the linear part of the curve displayed as a function of the square root of time. There is a water loss of $-0.17 \pm 0.1 \text{ kg/m}^2\cdot\text{h}$ during the first stage of drying, with a minimum value of $0.15 \text{ kg/m}^2\cdot\text{h}$, and $0.20 \text{ kg/m}^2\cdot\text{h}$ as maximum. The drying rate of the second phase is $-0.23 \pm 0.1 \text{ kg/m}^2\cdot\text{h}^{1/2}$, with $-0.22 \text{ kg/m}^2\cdot\text{h}^{1/2}$ as the minimum value, and $-0.24 \text{ kg/m}^2\cdot\text{h}^{1/2}$ as the maximum. The measured drying process duration is in line with previous research [14,33]. According to the study of Karaca [34], who examined the water absorption and drying behaviour of several types of stones, the drying phase is always longer than the time required to obtain water saturation, except for the most porous variety of limestone considered. Actually, the pore network structure is a fundamental characteristic of the material which influences its drying behaviour [35].

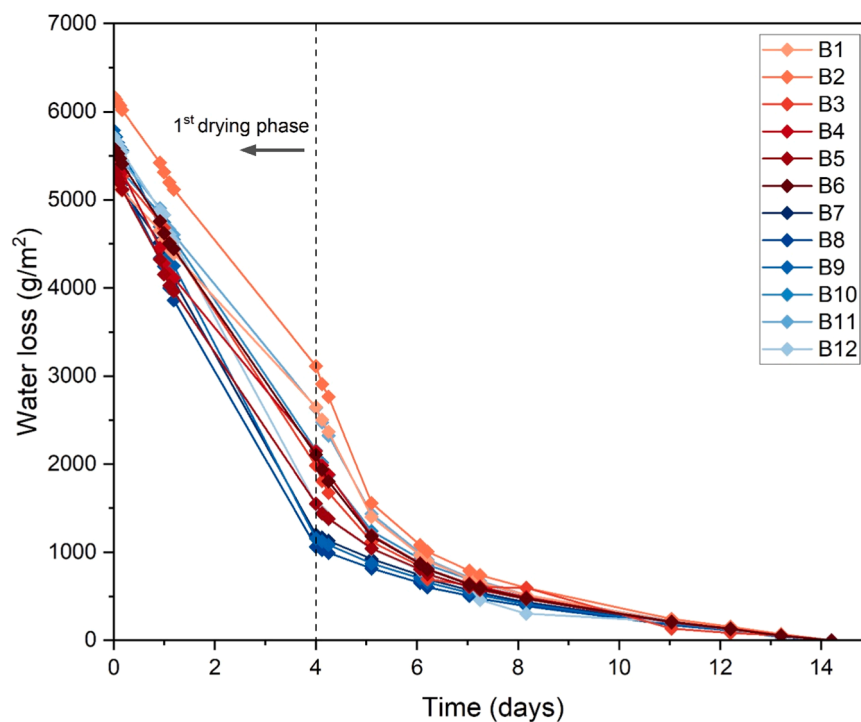


Fig. 7. Drying curves of the Euville limestone are represented by the decrease in water content per unit of surface against time for each examined block. Each one is nominated with the letter “B” for block and a number.

The drying and vapour diffusion experiments illustrate how the pore network influences the ability of the stone to release moisture after wetting. The long drying period of approximately 14 days indicates that Euville limestone retains moisture for extended durations in an environment of approximately 60% RH, which is common for the climate where Euville is used (in France, Belgium, and the Netherlands). This behaviour suggests the presence of narrow throats connecting larger pores, creating capillary condensation zones that delay evaporation. The observed drying behaviour is in line with the MIP results (Fig. 5). The bimodal distribution shows a significant volume of macropores ($40\ \mu\text{m}$) and a secondary population of narrow throats ($1\ \mu\text{m}$). During the first stage of drying, the larger pores at the surface empty rapidly. However, as the drying front recedes, the transport of liquid water to the evaporation surface is governed by the $1\ \mu\text{m}$ throats. The high capillary suction associated with these smaller diameters, combined with the 'ink-bottle' effect where macropores are shielded by these narrow connections, creates a bottleneck that slows the drying rate. This microstructural restriction is the primary driver for the prolonged second drying stage, as the narrow throats facilitate local capillary condensation zones that delay the final stabilisation of the mass.

The slow drying observed in Euville limestone has significant implications for its durability as a building stone material. Within temperate or humid climates, moisture is the primary driver for several degradation mechanisms. Firstly, the retention of moisture within the pore network facilitates the dissolution of calcium carbonate, the migration of carbonate and soluble salt ions, and subsequent crystallisation in the outer part of the material [9,36,37]. If the stone remains wet for long periods, with evaporation occurring primarily at the surface, the newly formed crystals originate in that outer part, leading to the removal of calcite from the stone core. This influences the cohesion of the grains with the cement, possibly leading to the disintegration observed in the weathered Euville. Secondly, the high surface humidity maintained during the slow first stage of drying creates an ideal environment for the colonisation of micro-organisms, which can lead to undesired staining and biochemical alterations of the calcitic matrix [14].

3.3. Water transport anisotropy

The water sorption of porous material under natural conditions is caused by either capillary imbibition or sorption from total immersion. The water behaviour in a porous medium strongly depends upon the network geometry. The transport properties and storage capacities are linked to pore radii, connected pore networks, and their ability to trap air bubbles.

Whether from rain and water runoff or from capillary water rising, the measurement of water absorption from total immersion provides a good quantification of the highest degree of saturation reached under natural conditions [10].

In Fig. 8, the total water absorption is shown. Here, two different phases can be distinguished. The first is characterised by rapid water absorption, during which the stone absorbs most of the water. In the case of Euville, this period lasts about 72 h and results in $4.86 \pm 0.22\ \text{wt}\%$ of absorbed water on average. The average total water absorption value for the 12 cubes examined at the end of the experiment (saturated samples) is $5.78 \pm 0.24\ \text{wt}\%$ with 4% as coefficient of variation. This limestone can thus be regarded as a highly absorbing material because the results are greater than the literature values for construction limestones, which range from 0.1 wt% to 3.0 wt% [38]. Fig. 8 also shows that the difference in water adsorption between specimens oriented parallel and perpendicular to the water level is small. Specifically, blocks from V7 to V12, which were oriented parallel to the water absorption direction, present an average value of $5.80 \pm 0.31\ \text{wt}\%$ with 5% as coefficient of variation and the perpendicular set, from H1 to H6, of $5.74 \pm 0.11\ \text{wt}\%$ with 2% as coefficient of variation. These values do not differ significantly from each other, thus suggesting the absence of structural anisotropy. A mathematical verification made through a two-sample *t*-test ($\alpha = 0.05$) results in $t = -0.38$ with $p = 0.72$, demonstrating the absence of a statistically significant difference between the two sample sets (horizontal and vertical orientations). The duration of the total water absorption at atmospheric pressure test, necessary to reach saturation, is 15 days. This period is considerably longer than the ones measured for other varieties of limestone, which, in general, correspond

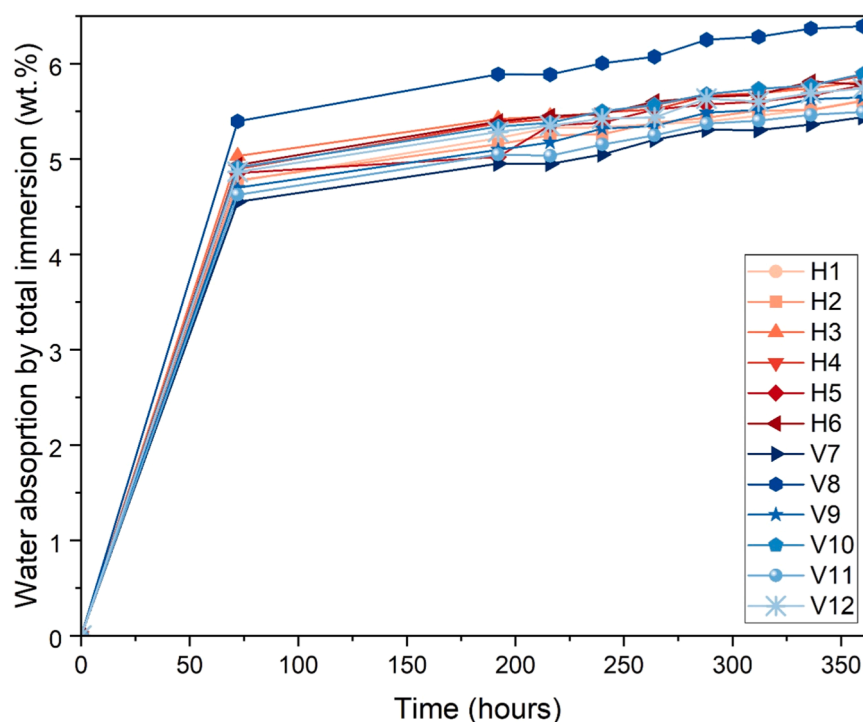


Fig. 8. Total water absorption at atmospheric pressure. Each curve represents the water absorption weight percentage against time for each block. The number indicates the single block, while the letters indicate the samples orientation: "V" is for vertical and "H" is for horizontal.

to a duration of less than a week [34,35].

The same mechanisms of absorption in two stages is evident with the capillary absorption test. The plot in Fig. 9 presents the weight increase per unit of surface area over the square root of time. First, the material absorbs water quickly, resulting in a linear increase, which is also used to calculate the capillarity coefficient. The second phase is linked to reaching the saturation condition which is visually identified by a plateau. The capillary coefficient for the whole set of samples corresponds to $77.47 \pm 9.19 \text{ g/m}^2\text{s}^{1/2}$. This value is in accordance with the range that has been presented by Benavente *et al.* [30] for eight varieties of calcarenites, similar types of limestone. At the same time, it differs from the results for the Euville that are present in the literature. The averaged capillary coefficient value calculated by Lopez-Arce *et al.* [2] is $45.60 \text{ g/m}^2\text{s}^{1/2}$, Lubelli *et al.* [31] and Schröer *et al.* [14] report $20.90 \text{ g/m}^2\text{s}^{1/2}$ and $60.56 \text{ g/m}^2\text{s}^{1/2}$, respectively. The same value, but calculated for the two sets of samples, is significantly different. The capillarity coefficient for the set oriented perpendicular to the water absorption direction (horizontal samples) is $71.72 \pm 8.46 \text{ g/m}^2\text{s}^{1/2}$, while the parallel (vertical samples) resulted in $83.22 \pm 5.90 \text{ g/m}^2\text{s}^{1/2}$. The two values differ by more than 10% from each other, thus indicating the existence of a difference in the water transport due to the sedimentary layers of the stone. Similar results are reported by Pestre *et al.*, having $71 \text{ g/m}^2\text{s}^{1/2}$ for the parallel and $42 \text{ g/m}^2\text{s}^{1/2}$ for the perpendicular directions. A two-sample *t*-test ($\alpha = 0.05$) was used to determine the statistical significance of capillary absorption anisotropy in specimens orientated parallel and perpendicular to the water absorption direction. The test resulted in $t = -4.61$ with $p = 0.0058$, suggesting a significant difference between the two orientations. The results obtained support the possible existence of anisotropic capillary water transport in Euville limestone, with higher absorption rates found in samples orientated vertically (parallel) to the water absorption direction.

A redistribution of water inside the material is determined by a stationary condition, which occurs after the imbibition phase ends (plateau in the graph of Fig. 9). In this second phase, the liquid creates a continuous network in which air can still be trapped between

macropores. The water flow in the stone is primarily controlled by capillary forces that take place in pores of $0.1 \mu\text{m}$ to $2500 \mu\text{m}$ [30]. The transition between the two phases is reflected by a slope shift in the capillary absorption curve. For this experiment, the slope change occurs around 60 h after the start of the test. It took around 8 days for the samples to reach the saturation condition. This is considerably faster than the time it took to reach the same condition during the total absorption experiment. The difference suggests that capillary action is the most efficient mechanism of water transport. As a result, high capillary coefficient measurements may indicate a high absorption capacity and, consequently, indicate a possible vulnerability of the material to water-induced degradation. In this process, the orientation of the pore network, which is determined by the sedimentary layering, is fundamental.

The Karsten tube is a tool used for measuring the water absorption of construction materials outdoors. Besides *in situ* measurements, another advantage is the vertical tube, useful for measuring the water absorption of façades. In the presented experiment, the vertical Karsten tube has been used on a block of Euville of $23 \times 22 \times 10 \text{ cm}^3$ (Fig. 4.b and Fig. 4. c). The aim is to test the efficiency of the instrument in measuring the water absorption per unit of surface of the stone and further evidence the presence of anisotropy in the water transport.

The graph presented in Fig. 10 shows water absorption curves per unit of surface area in relation to the square root of time, measured using the vertical Karsten tube. It is clear that when placed parallel to the sedimentary layers water absorption is much faster. The absorption of 4 mL of water takes 6 min for the parallel direction and 14 min for the perpendicular direction.

The water absorption per unit of surface was calculated considering only the linear part of the graph, thus excluding readings during the first 3 min of the experiment. The resulting values are significantly higher than those measured with the capillary absorption. Specifically, $422.60 \text{ g/m}^2\text{s}^{1/2}$ and $563.05 \text{ g/m}^2\text{s}^{1/2}$, for the perpendicular and parallel direction, respectively. These values are significantly higher than those from the standard capillary test, which are $71.72 \text{ g/m}^2\text{s}^{1/2}$ for the

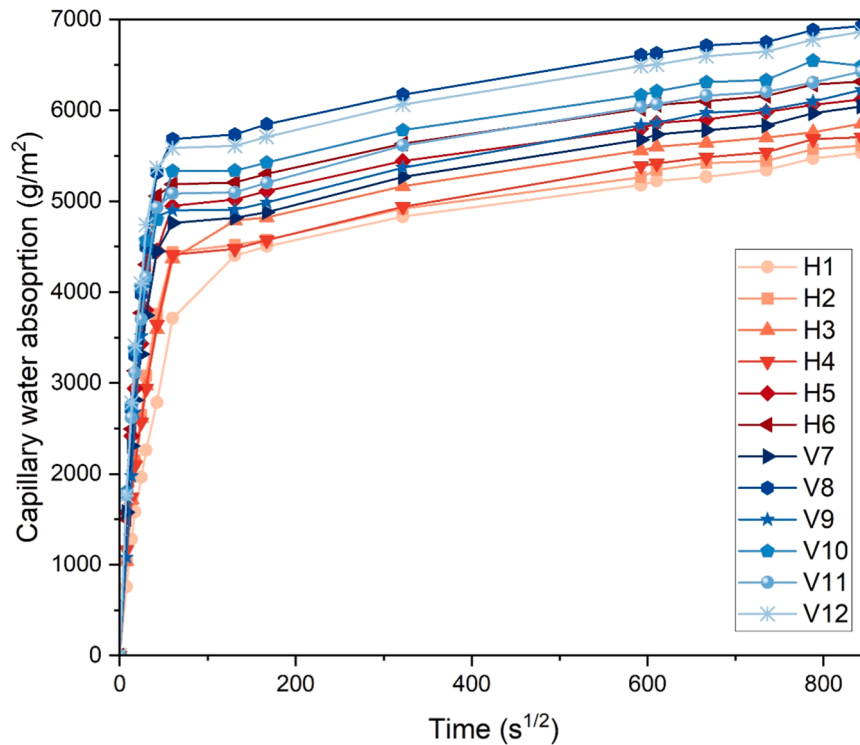


Fig. 9. Capillary water absorption curves. The amount of water absorbed per unit of surface is plotted as a function of time. The number indicates the single block, while the letters indicate the samples orientation: "V" is for vertical and "H" is for horizontal.

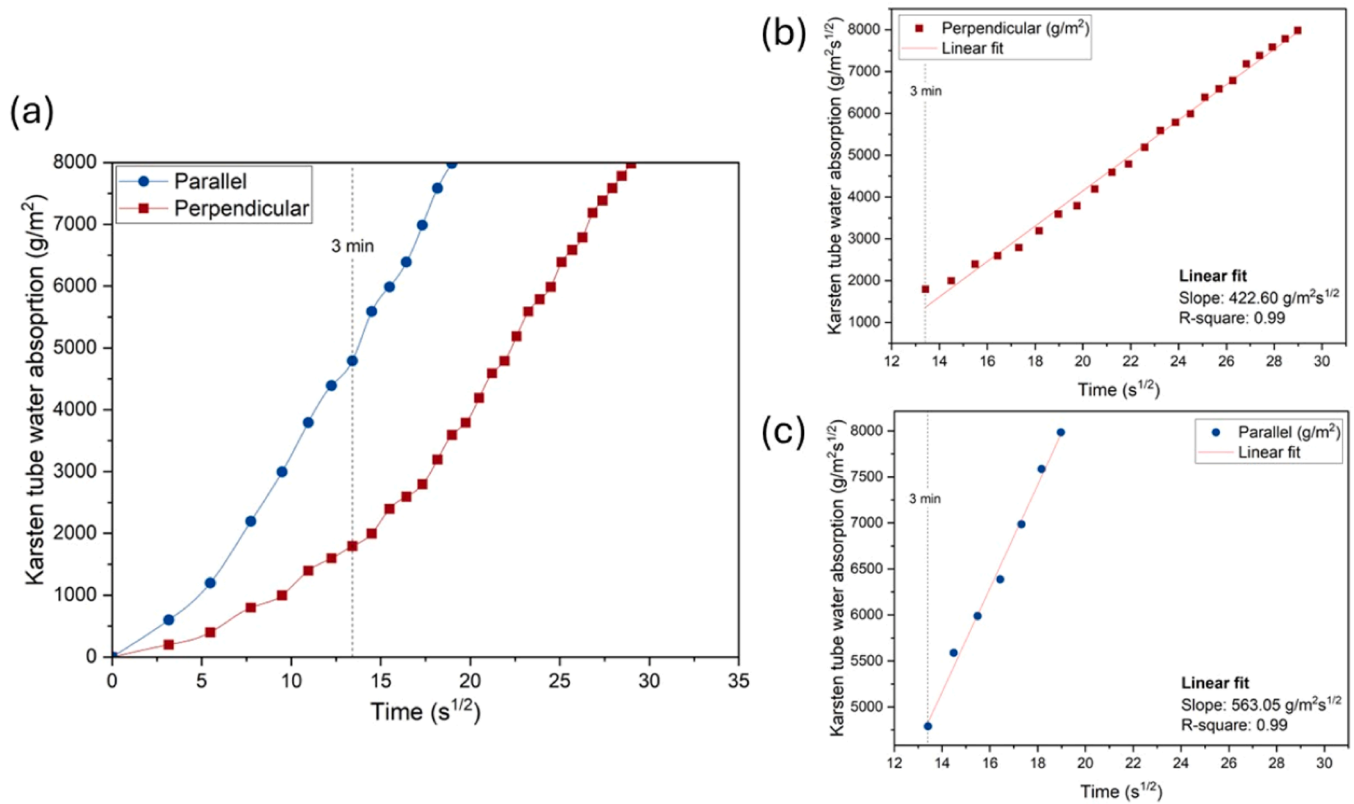


Fig. 10. Water absorption with the vertical Karsten tube. (a) Amount of water absorbed per unit of surface by the Euville block. (b) Linear fit of water absorption perpendicular to the sedimentary layers. (c) Linear fit of water absorption parallel to the sedimentary layers. (c) Linear fit of water absorption parallel to the sedimentary layers.

perpendicular and $83.22 \text{ g/m}^2 \text{s}^{1/2}$ for the parallel. The observed difference might be since the Karsten tube measurement is based on a relatively small absorption area, $5.72 \times 10^{-5} \text{ m}^2$, and the water absorption speed can be accelerated by the presence of voids or cracks that are hidden under the surface [16]. Additionally, in the Karsten tube setup, water spreads laterally into the dry stone surrounding the tube, whereas the standard laboratory capillary absorption test is restricted to one-dimensional vertical transport. Consequently, the Karsten tube measures an absorption influenced by the three-dimensional connectivity of the pore network and local surface defects. Despite the difference in the results, Karsten tube experiments can be useful to quickly estimate the influence of the stone anisotropy on hygric properties, for mapping surface heterogeneity and identifying cracks, or to evaluate the influence of treatments on water absorption, like water repellent applications [17]. For example, the experiment can be effective in a survey of an extended area where the variability of the measurement can indicate the presence of cracks and voids.

4. Conclusions

This study provides a comprehensive assessment of the moisture transport behaviour of the Euville limestone, integrating microstructural, hygric, and mechanical analysis, while also taking sedimentary layers into account.

- Euville limestone displays a bimodal pore size distribution (maxima at $1 \mu\text{m}$ and $40 \mu\text{m}$) and a significant difference between total (14.16%) and open (12.57%) porosity, revealing the presence of closed pores. This structure explains both rapid liquid ingress through interconnected macropores and slow moisture release through a microporous network.

- MIP underestimates open porosity compared to water absorption (7.14% by MIP and 12.57% by water absorption) due to the presence of large pores, which are above the detection limit of MIP and possibly to the ink-bottle effect. However, the technique still gives valuable insights into the size distribution of smaller pores.
- Compressive strength is 25.73 on average and shows no anisotropy.
- The overall drying duration of the Euville limestone was 14 days. During the first phase, the drying rate was $-0.17 \text{ kg/m}^2 \cdot \text{h}$ and $-0.23 \text{ kg/m}^2 \cdot \text{h}^{1/2}$ during the second. This suggests the presence of narrow throats connecting larger pores, creating capillary condensation zones that delay evaporation. This is also consistent with MIP results.
- The vapour diffusion resistance factors are higher for the dry-cup ($\mu=35.78$) than in the wet-cup ($\mu=21.58$) condition, indicating increased permeability at higher relative humidity due to surface diffusion and capillary condensation.
- Capillary absorption and Karsten tube tests consistently demonstrated anisotropic water transport, with higher absorption rates observed when water flow was oriented parallel to the depositional layers. This anisotropy arises from grain packing along bedding planes. Even though the rates measured with the Karsten tube are higher, it still demonstrates the anisotropy in water absorption. Understanding the directional dependence of water uptake offers actionable knowledge for both construction and conservation practice. Building blocks cut parallel to bedding may be more susceptible to degradation, given the higher water absorption capacity. Therefore, careful consideration of block orientation is fundamental. Moreover, the identification of prolonged moisture retention underscores the need for adequate drying periods and ventilation strategies.

The characterisation of moisture transport and microstructural

anisotropy in this study provides the necessary physical basis for assessing the stone's susceptibility to environmental stressors such as freeze-thaw cycles. While experimental freeze-thaw cycling was outside the scope of this initial characterisation, it represents the next critical step in broader research related to the Euville conservation research campaign.

CRedit authorship contribution statement

Katrin Schollbach: Supervision. **H.J.H Brouwers:** Writing – review & editing, Supervision. **Beatrice Cerrai:** Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

Funding

This research received no specific grant from any funding agency in the public, commercial, or not-for-profit sectors.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors would like to express their sincere gratitude to Mr. Hendrik Tolboom, natural stone specialist at the Cultural Heritage Agency of the Netherlands (RCE), for his invaluable guidance and for introducing the authors to the complexities of stone conservation and the specific challenges associated with Euville limestone. We also thank him for his practical assistance in sourcing intact Euville limestone blocks from the Vandekerckhove NV masonry stockpile.

Data availability

Data will be made available on request.

References

- [1] S. Siegesmund, A. Torok, *Building Stones. Stone in Architecture: Properties, Durability*, Springer Berlin Heidelberg, 2011, pp. 11–95, ch. 2.
- [2] P. Lopez-Arce, et al., Physico-chemical stone-mortar compatibility of commercial stone-repair mortars of historic buildings from Paris, *Constr. Build. Mater.* 124 (Oct. 2016) 424–441, <https://doi.org/10.1016/j.conbuildmat.2016.07.076>.
- [3] J. Dewanckele, T. De Kock, G. Fronteau, H. Derluyn, P. Vontobel, M. Dierick, L. Van Hoorebeke, P. Jacobs, V. Cnudde, Neutron radiography and X-ray computed tomography for quantifying weathering and water uptake processes inside porous limestone used as building material, *Mater. Charact.* 88 (Feb. 2014) 86–99, <https://doi.org/10.1016/j.matchar.2013.12.007>.
- [4] J.J. Hughes, T. Howind, Science and Art: A Future for Stone. Proceedings of the 13th International Congress on the Deterioration and Conservation of Stone," in Proceedings of the 13th International Congress on the Deterioration and Conservation of Stone, University of the West of Scotland, Paisley, Sep. 2016.
- [5] P.A. Scholle, D.S. Ulmer-Scholle, *A Color Guide to the Petrography of Carbonate Rocks: Grains, textures, porosity, diagenesis*, American Association of Petroleum Geologists, 2003.
- [6] R. Prikryl, Durability assessment of natural stone, *Q. J. Eng. Geol. Hydrogeol.* 46 (4) (2013) 377–390, <https://doi.org/10.1144/qjegh2012-052>.
- [7] M. Laho, C. Franzen, R. Holzer, P.W. Mirwald, Pore and hygric properties of porous limestones: a case study from Bratislava, Slovakia, *Geol. Soc. Spec. Publ.* 333 (2010) 165–174, <https://doi.org/10.1144/SP333.16>.
- [8] M. Steiger, A.E. Chardia, K. Sterflinger, *Weathering and Deterioration. Stone in Architecture: Properties, Durability*, Springer, Berlin Heidelberg, 2011, pp. 227–316, ch. 4.
- [9] E.M. Winkler, Weathering and weathering rates of natural stone, *Environ. Geol. Water Sci.* 9 (2) (1987) 85–92.
- [10] M. Bellanger, F. Homand, J.M. Remy, Water behaviour in limestones as a function of pores structure: application to frost resistance of some Lorraine limestones, *Eng. Geol.* 36 (1993) 99–108, [https://doi.org/10.1016/0013-7952\(93\)90022-5](https://doi.org/10.1016/0013-7952(93)90022-5).
- [11] S. Siegesmund, R. Snethlage, *Stone in Architecture: Properties, Durability*, Springer Berlin Heidelberg, 2011, <https://doi.org/10.1007/978-3-642-14475-2>.
- [12] T. Briole, et al., Experimental study of microstructural controls on the weathering of carbonate rocks, *Geochem. Soc. (Dec. 2022)*, <https://doi.org/10.46427/gold2022.10188>.
- [13] R.M. Espinosa Marzal, G.W. Scherer, Crystallization of sodium sulfate salts in limestone, *Environ. Geol.* 56 (3–4) (Dec. 2008) 605–621, <https://doi.org/10.1007/s00254-008-1441-7>.
- [14] L. Schröer, T. De Kock, S. Godts, N. Boon, V. Cnudde, The effects of cyanobacterial biofilms on water transport and retention of natural building stones, *Earth Surf. Process. Landf.* 47 (8) (Jun. 2022) 1921–1936, <https://doi.org/10.1002/esp.5355>.
- [15] T. Pestre, E. Antczak, Franck Brachelet, D. Pallix, Multiphysical Charact. Limest. Energy-Effic. Sustain. Build. Compon. (2022), [https://doi.org/10.1061/\(ASCE\)MT.1943](https://doi.org/10.1061/(ASCE)MT.1943).
- [16] R. Hendrickx, Using the Karsten tube to estimate water transport parameters of porous building materials: the possibilities of analytical and numerical solutions, *Mater. Struct. /Mater. Et. Constr.* 46 (8) (Aug. 2013) 1309–1320, <https://doi.org/10.1617/s11527-012-9975-2>.
- [17] R. Duarte, I. Flores-Colen, J. de Brito, A. Hawreen, Variability of in-situ testing in wall coating systems - Karsten tube and moisture meter techniques, *J. Build. Eng.* 27 (Jan. 2020), <https://doi.org/10.1016/j.jobe.2019.100998>.
- [18] J. David, M. Roux, Current model of bioclastic production by pedunculated crinoids. Application to the entrobic limestone of Euville (Oxfordian of the Meuse), *Comptes Rendus De l'Acad. émie Des. Sci. -Ser. IIA-Earth Planet. Sci.* 330 (2) (2000) 111–116, [https://doi.org/10.1016/S1251-8050\(00\)00107-5](https://doi.org/10.1016/S1251-8050(00)00107-5).
- [19] G. Fronteau, C. Schneider-Thomachot, E. Chopin, V. Barbin, D. Mouze, A. Pascal, Black-crust growth and interaction with underlying limestone microfacies, *Geol. Soc. Spec. Publ.* 333 (2010) 25–34, <https://doi.org/10.1144/SP333.3>.
- [20] EN 1926, Test. Methods Nat. Stone - Determ. uniaxial Compress. Strength. (2006), 2006.
- [21] ISO 12572, Hygrothermal Perform. Build. Mater. Prod. - Determ. Water Vap. Transm. Prop. - Cup Method. (2016).
- [22] EN 13755, Natural stone test methods - Determination of water absorption at atmospheric pressure, CEN, 2008.
- [23] EN 1925, Nat. Stone Test. Methods - Determ. Water absorption Coeff. capillarity (1999).
- [24] BS EN 16322, Conserv. Cult. Herit. - Test. Methods - Determ. Dry. Prop. (2013).
- [25] M. Leger, L. Luquot, Importance of Microstructure in Carbonate Rocks: Laboratory and 3D-Imaging Petrophysical Characterization, *Appl. Sci.* 11 (9) (2021), <https://doi.org/10.3390/app11093784>.
- [26] S. Salvini, C. Coletti, L. Maritan, M. Massironi, F. Balsamo, C. Mazzoli, Exploring the pore system of carbonate rocks through a multi-analytical approach, *Environ. Earth Sci.* 82 (23) (Dec. 2023), <https://doi.org/10.1007/s12665-023-11234-1>.
- [27] S. Roels, J. Elsen, J. Carmeliet, H. Hens, Characterisation of pore structure by combining mercury porosimetry and micrography, *Mater. Struct.* 34 (Mar. 2001) 76–82.
- [28] G. Li, Y. Miao, R. Li, Ultrasonic S-wave to obtain shear modulus and matrix permeability of D'Euville Limestone, *Geofluids* 2022 (2022), <https://doi.org/10.1155/2022/5126760>.
- [29] M. Leger, L. Luquot, D. Roubinet, Role of mineralogical, structural and hydrodynamic rock properties in conduits formation in three distinct carbonate rock types, *Chem. Geol.* 607 (Sep. 2022), <https://doi.org/10.1016/j.chemgeo.2022.121008>.
- [30] D. Benavente, M.A.G. del Cura, R. Fort, S. Ordóñez, Durability estimation of porous building stones from pore structure and strength, *Eng. Geol.* 74 (1–2) (Jul. 2004) 113–127, <https://doi.org/10.1016/j.enggeo.2004.03.005>.
- [31] B. Lubelli, R.P.J. van Hees, T.G. Nijland, J. Bolhuis, A new method for making artificially weathered stone specimens for testing of conservation treatments, *J. Cult. Herit.* 16 (5) (Sep. 2015) 698–704, <https://doi.org/10.1016/j.culher.2015.01.002>.
- [32] C. Hall, G.J. Lo, A. Hamilton, Water vapour permeability of inorganic construction materials, *Mater. Struct. /Mater. Et. Constr.* 57 (2) (Mar. 2024), <https://doi.org/10.1617/s11527-023-02281-y>.
- [33] H. De Clercq, M. Jovanović, K. Linnow, M. Steiger, Performance of limestones laden with mixed salt solutions of Na₂SO₄-NaNO₃ and Na₂SO₄-K₂SO₄, *Environ. Earth Sci.* 69 (5) (Jul. 2013) 1751–1761, <https://doi.org/10.1007/s12665-012-2017-0>.
- [34] Z. Karaca, Water absorption and dehydration of natural stones versus time, *Constr. Build. Mater.* 24 (5) (May 2010) 786–790, <https://doi.org/10.1016/j.conbuildmat.2009.10.029>.
- [35] Y. Özcelik, A. Ozguven, Water absorption and drying features of different natural building stones, *Constr. Build. Mater.* 63 (Jul. 2014) 257–270, <https://doi.org/10.1016/j.conbuildmat.2014.04.030>.
- [36] D. Camuffo, Physical weathering of stones, *Sci. Total. Environ.* 167 (1995) 1–14.
- [37] C. Franzen, P.W. Mirwald, Moisture content of natural stone: static and dynamic equilibrium with atmospheric humidity, *Environ. Geol.* (Aug. 2004) 391–401, <https://doi.org/10.1007/s00254-004-1040-1>.
- [38] R. de Sousa Camposinhos, *Stone Cladding Engineering*, 1, Springer, Dordrecht, 2014, <https://doi.org/10.1007/978-94-007-6848-2>.