Architectural Heritage and Climate Change: Performance Prediction of Exposed Natural Building Stone to Rainfall through a Combination of Laboratory Leaching Tests and X-ray Computed Tomography

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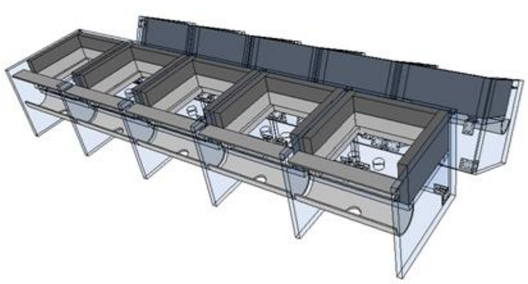
Introduction

Natural stone has been over the centuries among the most common and prominent materials used in construction. The natural and irreversible process of ageing results in a progressive deterioration of the original properties, whose level and rate are determined by the interaction of environmental factors and the stone's intrinsic features. Water represents one of the most important causes of decay for historic buildings; atmospheric precipitation is responsible for the surface recession of carbonate stones, especially in areas where the presence of pollution induces a decrease in the rainwater pH (Grossi and Brimblecombe, 2007). In connection to the climate change issue, recession rate in high-rainfall regions of Europe is expected to increase, due to the rise in carbon dioxide atmospheric concentration (Bonazza *et al.*, 2009). However, not only the mineral composition, but also the structural properties are important factors influencing the performance. Experimental testing carried under laboratory conditions found the existence of correlation between the stone's microstructure and its degradation caused by wet deposition (Franzoni and Sassoni, 2011). This highlights the need for a specific and targeted evaluation of the individual case to inform decision-making about conservation and selection of stone for necessary material replacement.

Aim and Methodology

The aim of this work is the performance evaluation of different calcareous sedimentary stones when exposed to surrogate rainfall, through a unique combination of chemical and morphological analyses with multivariate data treatment. The applied methodology combines accelerated ageing with X-ray computed tomography (XCT) and inductively coupled plasma mass spectrometry (ICP-MS), to evaluate the structural and compositional properties of the specimens and measure the level of mineral dissolution resulting from the interaction with water. A custom-built apparatus has been specifically designed for the artificial laboratory weathering (Fig. 1a), consisting of multiple independent exposure units, each accommodating one sample of dimension 5x5x1 cm (Fig. 1b). For the preliminary test here described triplicate specimens of three stone types containing different amounts of carbonate minerals and porosity were selected, i.e., Bernese sandstone, Pitairlie sandstone and Cadeby limestone (Fig. 2a). In addition to the compositional and structural requirements, these specimens were preferred for their wide employment in historic buildings and conservation works. The surrogate rainwater (10^{-4} mol L^{-1} HNO_3 at pH 4) was delivered to the samples at regular intervals of time during the course of 14 days. Performance was estimated in terms of mineral dissolution, through the determination of the concentration of cations (Ca^{2+}, K^+, Mg^{2+} and Na^+) in the run-off water collected every two days during the experiment. Principal component analysis (PCA) was used to simplify the complex dataset by identifying latent variables that captured the maximum variance. This feature was employed to verify the discrimination between responses of different stone typologies and identify the most influential variables in the performance variation.



a

b

Figure 1. Costum-built apparatus for the artificial exposure of the samples a) 3D model of the original design b) the apparatus in use with the replicates in their exposure units

Results

The statistical treatment of the data showed that the three stone typologies were well differentiated based on their response to the leaching action of surrogate rainwater. The PCA score plot displaying the cation concentrations measured in all of the leachates collected indicated three clusters, each relating to a type of stone (Fig. 2b). As expected, calcium and magnesium, originating from the dissolution of dolomite and calcite, were the variables with the highest discriminatory power. Overlap between some of the samples belonging to different typologies may have arisen from a fault in the water delivery system, which distributed surrogate rainwater unevenly in a few cases. The relationship between the amount of calcium released and the volume of water supplied exhibited a positive trend only for the less porous lithotypes, while the opposite behaviour was found for the most porous specimen, i.e., Bernese sandstone (Fig. 2c). At the end of the test, the different types of stone showed different mass-loss percentage, but the precision between replicates of the same typology that received comparable amounts of water was good, i.e., 0.010 ± 0.005 for Pitairlie sandstone, 0.017 ± 0.003 for Bernese sandstone, and 0.042 ± 0.001 for Cadeby limestone, indicating equivalent performance.

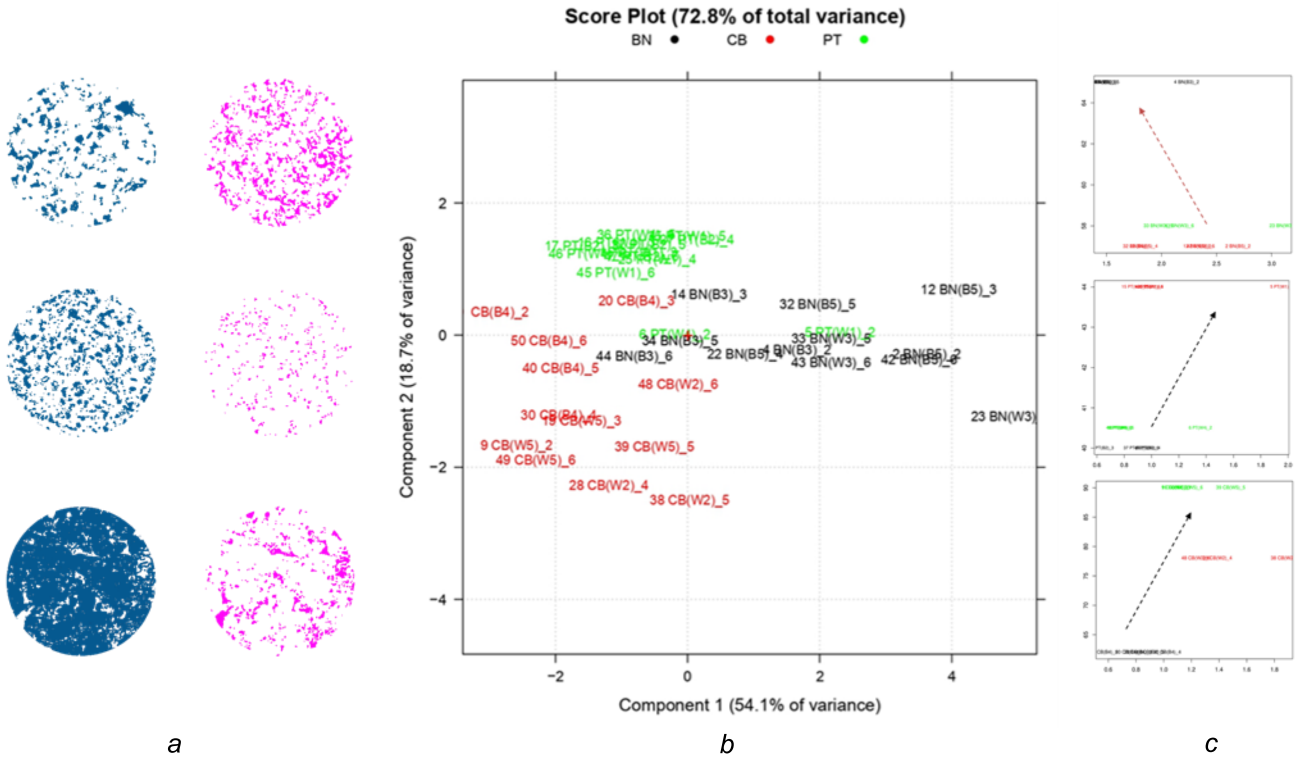


Fig. 2. Results from the X-ray computed tomography and statistical treatment of the dataset a) Segmentation of three samples sections acquired by XCT showing carbonate minerals content (blue) and porosity (pink); from top to bottom: Bernese sandstone, Pitairlie sandstone, and Cadeby limestone b) Score plot from the principal component analysis c) Correlation for the three specimens between volume of water delivered and total calcium concentration in the run-off water

Conclusions and future work

This preliminary test revealed good discrimination between individual responses to the action of surrogate rainwater. The weathering process was mainly controlled by the dissolution of the carbonate minerals. It was also found that not all specimens responded equally to amount of water delivered. The most porous type exhibited a negative relationship, indicating a direct effect of the rainfall event duration over the dissolution process. This circumstance might suggest a connection with the more rapid evaporation of water experienced by highly porous lithotypes. These findings highlight the necessity to further investigate the relationship between the weathering process and the chemical and physical properties of the stone. Future testing will involve the analysis of a wider range of stone typologies. XCT carried before and after the leaching test will support the assessment of the parameters that are influential in controlling the rate of degradation and its progression, offering a visual representation of the alteration. In predicting the stone performance, the main challenge is represented by the complexity of the system under analysis, where a high number of variables are actively involved in controlling the response. Such considerations emphasize, as it has been has been previously identified (Viles, 2013), the need for further developments in this field, combining laboratory and field exposure testing with the non-destructive monitoring of existing buildings. The employment of digital technologies might support this resolution, and provide a mean for the non-destructive assessment of the stone performance. This will help to create the much-needed fundamental research on stone decay and conservation, with a particular focus on climate change.

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Conflict of Interests Disclosure

The authors have no conflicts of interest to declare.

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