Impact of stylolite cementation on weathering rates of carbonate rocks

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Short Report

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Abstract

The weathering of carbonate rocks plays a significant role in the evolution of Earth's surface. Such weathering is often accelerated by the presence of stylolites, which are rough, serated surfaces that form by dissolution under burial or tectonic stresses. Stylolites are thought to represent zones of mechanical weakness in rocks, as well as regions in which chemical weathering is enhanced. However, a quantitative framework capable of predicting how stylolites accelerate weathering in carbonates has yet to be achieved. In this study, we first used scanning electron microscopy and wavelength dispersive spectroscopy to characterize the way in which the two sides of individual stylolites are connected at the microscopic scale. In the samples we examined, we found that micrometer-scale calcite bridges span the opposing sides of the stylolites, effectively cementing the rock together. This cement filled 1%-30% of the stylolite volume. We then used a numerical cellular automaton model to simulate the effect that the degree of carbonate cementation has on stylolitic carbonate rock weathering. Our results show that weathering rates decrease non-linearly as the degree of stylolite cementation increases. The effect on overall rock weathering rates is significant: stylolite-bearing rocks with 1% cementation weathered as much as 37 times faster than limestone without stylolites, primarily because of accelerated mechanical erosion. Our results indicate that stylolites could be as important as joints and fractures in accelerating carbonate rock weathering and in the development of karst landscapes, potentially making a major contribution to global carbonate weathering.

1. Introduction

The weathering of carbonate rocks plays a major role in the evolution of Earth's surface. Carbonate rocks comprise 10–20% of Earth's crust (Morse et al. 2007) and cover about 25% of the continental surface (Wang et al. 1999; Goldscheider et al. 2020). On a global scale, the weathering of carbonate rocks is a significant sink for atmospheric CO2, contributing to the regulation of both short-term and long-term climate change (Liu et al. 2011, 2018). On regional and local scales, carbonate weathering can control the evolution of soils and karst landscapes, the composition of groundwater, and the erosion of buildings, sculptures, and historical monuments (Buhmann and Dreybrodt 1985; Doherty et al. 2007).

Like all rocks, carbonate weathering rates are enhanced by the presence of fractures and joints (Goldscheider et al. 2020; Israeli et al. 2021; Zhao et al. 2022; Xu et al. 2022). However, many carbonate rocks also contain stylolites (Larbi 2003; Simpson 2009; Wangler et al. 2016; Aly et al. 2018; Davis 2018), which are rough, serrated surfaces that form by mineral dissolution as a result of diagenetic or epigenetic processes (Rolland et al., 2012; Kaduri, 2013; Toussaint et al., 2018). Because stylolites, like fractures and joints, represent planes of discontinuity they are also be expected to accelerate weathering, and this is supported by both field observations and experiments. For example, at the Western Wall in Jerusalem, Israel, the average erosion rate in stylolitic limestone was found to be over an order of magnitude higher than in rocks that were stylolite-free (Fig. 1; Emmanuel and Levenson, 2014; Emmanuel, 2015). Similarly, an experimental study that tested the effects of heating and drying cycles on limestone cladding showed that stylolite-bearing rock was 3.5 times more susceptible to erosion than samples without stylolites (Aly et al. 2018). Such enhanced weathering is likely to be the result of both physical and chemical processes: as well as representing planes of mechanical weakness (Simpson 2009; Baud et al. 2016; Koehn et al. 2016; Israeli et al. 2021), stylolites can also serve as conduits for fluid flow that facilitate chemical dissolution and the swelling of clay minerals and (Larbi 2003; Wangler et al. 2016).

One of the factors likely to influence the affect that stylolites have on weathering is cementation. In stylolitic carbonates, the gaps between the opposing sides of the stylolite surface are often partially filled with clay minerals, organic matter, and oxide phases (Rolland et al. 2012; Baud et al. 2016; Aly et al. 2018). However, only calcareous cement is likely to bind the opposing sides of the stylolite together. While this can be understood intuitively, the way the interfaces are cemented at the microscopic level has not been examined, and the effect of cementation on weathering rates in stylolite-bearing carbonates has yet to be quantified.

In this study, we use high-resolution imaging to characterize stylolite interfaces in calcareous rocks and propose a conceptual framework and numerical model to describe the impact of stylolite cementation on carbonate weathering rates. Our model includes the effects of both chemical and mechanical weathering, and simulates the rates of denudation in stylolite-bearing carbonate rock possessing different degrees of cementation. We compare our results with reported field rates and discuss the implications of our analysis for karst formation and global carbonate weathering.

2. Methods

2.1 Sampling and high-resolution imaging

To characterize stylolite interfaces, we sampled Cretaceous stylolite-bearing limestone samples from 2 outcrops of the Bina Formation in Jerusalem, Israel: Givat HaTanach (31°46'04.1"N 35°13'32.8"E) and Ramat Shlomo (31°48'28.7"N 35°13'40.8"E). Samples were examined at high-magnification using an Electron Probe MicroAnalyzer (EPMA; JEOL JXA 8230) that provided backscattered electron (BSE) images and wavelength dispersive spectrometry (WDS) maps for 8 elements (Ca, Al, Si, Mg, O, C, K, and Fe).

2.2 Conceptual model

In our model, we assume that the reaction rate along a stylolite depends on the microscopic properties of the stylolite interface. We consider stylolites in carbonate rocks to be gaps that are partially filled by microscopic calcareous cement bridges that connect the opposing sides of the stylolite. These bridges are physically separated from each other by the remaining components filling the stylolite gap: clay minerals, metal oxides, organic matter, and voids. However, these other components do not contribute to the mechanical strength of the stylolite, and we also consider them to be chemically inert. The rock surrounding the stylolite comprises 100% calcareous minerals that can dissolve in contact with fluid.

Using this approach, the volumetric proportion of calcite in the stylolites (V_{cement}) can vary from zero to unity. In fully cemented stylolites (i.e., V_{cement} = 1), there is no effective difference between the bulk rock and the stylolites, and weathering proceeds as if the rock was stylolite free; by contrast, when V_{cement} = 0 the rock has no effective cohesion and the rock will disintegrate immediately. However, at intermediate values (i.e., $0 < V_{cement} < 1$), the weathering rate along the stylolites (i.e., perpendicular to a reactive fluid front) will be quicker than in the surrounding bulk rock because there is less calcareous material to dissolve for a given volume (Fig. 2). Moreover, the rate of dissolution along the stylolite should be inversely proportional to V_{cement}, so that for a limestone in which 10% of the stylolite volume comprises calcite cement, the dissolution rate along the stylolite should be 10 times faster than in the surrounding rock.

This conceptual model serves as the basis for the numerical model described in the next section.

2.3 Numerical model

To determine the impact of carbonate cementation on weathering in stylolite-bearing rocks, we used a numerical code based on a previously published model (Israeli and Emmanuel 2018). The framework simulates the way a rock surface weathers, both chemically and mechanically, when it is in contact with a reactive solution that is dissolving minerals at the fluid-solid interface. In the new simulations presented here, the model rock was assigned a 2D stylolite network pattern (Fig. 3) that was based on the digitization of a stylolite-rich limestone quarry wall from the Avnon Formation in Mitzpe Ramon, Israel (Laronne Ben-Itzhak et al. 2014).

Coded in Matlab®, the model is based on a cellular automaton algorithm. At every step only the pixels that are in direct contact with the fluid can dissolve. In our model domain, a 120 cm × 550 cm stylolite pattern is represented by a 1464 × 6710 pixel matrix with a resolution of 145

pixels cm^{-2}. The pattern is segmented into 2 phases: stylolites and surrounding carbonate rock. Each pixel is assigned a characteristic rate coefficient (distinct from a rate constant in chemical kinetics) that represents the probability of undergoing chemical dissolution, with a high probability corresponding to a high dissolution rate. The algorithm then determines stochastically which pixels will dissolve at every step. The more a pixel is surrounded by a fluid, the higher the probability that it will dissolve during the current step.

Each simulation begins by exposing the uppermost rock surface to fluid. The stylolites do not initially contain a fluid phase. The rock is then weathered chemically in every time step according to the characteristic value of each pixel exposed to the fluid. Every dissolved pixel is assigned a characteristic value of a fluid. After each stage of chemical dissolution, the rock matrix is searched for pixels that are completely surrounded by fluid. Clusters surrounded entirely by fluid are removed, simulating mechanical detachment, and the new pattern becomes the input for the next step of chemical dissolution.

To minimize possible boundary effects, an internal bounding box was defined that represented 53% of the domain. This bounding box began at the top of the domain and continued down to 200 cm above the bottom of the domain; the sides of the box began at a distance of 10 cm from each of the side boundaries. Simulations were terminated when half of the initial pixels inside the bounding box were dissolved. The simulations were run on the whole domain but calculations of weathering rates were only carried out for the region within the bounding box.

To simulate the effect of microscopic scale cementation on dissolution rates at the pixel scale, we assigned a rate coefficient to the stylolite pixels that was different from the surrounding rock. In each simulation, the rate coefficient in the stylolites was assumed to be uniform throughout the domain. We tested the effect of different stylolite rate coefficients on weathering rates. These values ranged from 1 to 100 times greater than the rate coefficient in the carbonate rock, corresponding to V_{cement} = 1 and V_{cement} = 0.01 respectively. In this study, we ran a total of 308 simulations, with 7 repeat realizations completed for each degree of carbonate cementation. The degree of cementation, simulated by the dissolution rate of the stylolite pixels was the only parameter varied in the simulations. The mean weathering rate calculated for the simulations with a rate coefficient of unity (i.e., stylolite-free rock) was used to normalize the weathering rates in all the other simulations.

3. Results And Discussion

3.1 Cement in stylolites at the microscopic scale

In all of the stylolitic limestones we examined, we found evidence for calcite cement at the microscopic scale. Elemental maps of calcium and carbon reveal that $CaCO_3$ bridges connect the bulk rock on opposing sides of the stylolites. These bridges are typically 2−50 µm wide. The degree of bridging, however, was not uniform, even within samples, and ranged from < 2% (Fig. 3a) to as high as 30% (Fig. 3b). Regions not filled by calcite contained a mixture of silicon, aluminum, magnesium, potassium and iron, which is consistent with the presence of clay minerals and metal oxides.

Our observations are consistent with those from previous studies that showed that cementation in stylolites can vary significantly both within and between formations (e.g., Koepnick, 1988; Araújo et al., 2021). Such variability is likely to be the result of a combination of diagenetic and epigenetic factors. For example, clay minerals that fill stylolite gaps are thought to determine stylolite patterns and topology (Ehrenberg et al., 2006; Ebner et al., 2010), and variations in clay content could affect the local degree of cementation. Similarly, hydrocarbons are thought to inhibit calcite precipitation, and the presence of oil or gas during epigenesis could also cause non-uniform cement patterns (Padmanabhan et al., 2015; Humphrey et al., 2019; Koepnick, 1988). Importantly, such heterogeneity means that determining a statistically representative value of stylolite cement for a given rock formation would require a high number of WDS or EDS analyses on different rock samples.

The images of stylolites at the microscopic scale also provide support for our conceptual model of weathering in stylolitic rocks. As long as water that is undersaturated with respect to calcite is able to permeate through the stylolite, the microscopic calcite bridges will dissolve, creating a front of mechanical discontinuity that will enhance weathering. Moreover, the fewer the bridges, the more rapidly this front will progress, and such a mechanism could explain the enhanced dissolution along stylolites observed in some formations (Araújo et al., 2021). In the next section, we show quantitatively how this mechanism accelerates weathering in stylolite-bearing rocks.

3.2 Impact of carbonate cementation on weathering rates

Our numerical model shows that the level of cement has a significant effect on the overall weathering rate (Fig. 4a). Rocks with 1% cement in the stylolites (V_{cement} = 0.01) were found to weather 37 times faster than rocks with no stylolites. However, this effect becomes less pronounced as cementation increases: at 20% carbonate cement (V_{cement} = 0.2), the weathering rate is only about 2.5 times faster than stylolite-free limestone.

The simulations also indicate that accelerated weathering at low levels of carbonate cementation is mainly due to high levels of mechanical disintegration (Fig. 4b). When V_{cement} = 0.01 the proportion of mechanical weathering is 65%; by contrast when V_{cement} = 0.2, the proportion decreases to 13%. This is because when dissolution along the stylolites is rapid, islands of rock surrounded by interconnected stylolites become detached before significant levels of dissolution in the rock matrix can occur. Although the model does not account explicitly for fluid flow through stylolites, we expect that high flow rates will enhance this effect, so that the elevated weathering rates we report should represent conservative estimates.

3.3 Comparison of field weathering rates with model results

By comparing the field weathering rates with our model results, the degree of carbonate cementation along stylolites in real rocks can be estimated. To demonstrate this, we used published data on weathering rates from the Western Wall in Jerusalem, Israel (Emmanuel and Levenson, 2014). At that location, limestone building blocks containing stylolites weather at rates of 23 - 29 mm ky^{-1}, while the rate for stylolite-free blocks is estimated to be 1 - 2 mm ky^{-1}. These values imply that stylolites enhance weathering by a factor of 11.5-29, which would correspond to a degree of cementation of approximately 1-4% (Fig. 5). Significantly, the degree of carbonate cementation predicted by the model is in agreement with that observed in electron microscopy images of the samples we examined here which are from the same formation that is thought to have provided stone for the Western Wall (Fig. 3).

In addition to influencing the weathering rates in monuments and building infrastructure, our findings suggest that carbonate rocks that contain stylolites could be more susceptible to karst formation than stylolite-free rocks. Indeed, in Brazilian limestones from the Potiguar basin, caves and other karst phenomena were found to be strongly associated with the presence of stylolites (Rabelo et al. 2020). In another study in the same region, Araújo et al. (2021) showed that stylolites control karst formation in carbonate rocks exposed to meteoric water, with stylolites increasing in size as a result of weathering until they merge, forming large cavities. Moreover, in a field study in South Italy, Magni (2020) showed that *terra rossa* soil, often associated with carbonate dissolution and karst development, was more common in areas with stylolites than in areas with fractures and joints. This could indicate that stylolites have a greater ability to accelerate weathering than other rock discontinuities. Clearly, such findings suggest that stylolites will enhance the dissolution of carbonate rock in other scenarios, such as the injection of reactive fluids into subsurface reservoirs for enhanced oil recovery and geological carbon storage.

4. Conclusions

In this study, we used high resolution imaging to show that stylolites contain calcite bridges at the microscopic scale that cement the rock together. This cement plays a critical role during weathering rate, and numerical modelling suggests that stylolitic limestones with a low degree of cementation will weather more than 30 times faster than stylolite-free limestones.

While our results demonstrate the importance of cementation in stylolites during carbonate weathering, our model has several limitations. First, we only tested one example of a 2D anastomosing stylolitic network; other kinds, such as isolated stylolites and bedding parallel stylolites, could behave in a very different manner, particularly in 3 dimensions. Second, our model assumes that the degree of cementation is uniform throughout the stylolite network; in reality, cementation could exhibit significant spatial variability that could affect overall weathering patterns and rates. However, incorporating such variability into future simulations in a meaningful way would require quantitative data from real stylolite networks. To achieve this, a reliable method to quantify cementation patterns in stylolites at spatial scales beyond the microscopic level would need to be developed.

Factors other than cementation are also likely to play an important role in determining the impact of stylolites on weathering rates. Stylolites often contain smectite clay minerals that expand in contact with water, increasing stress within the stylolite and enhancing the mechanical weathering rate. Fluid flow too is expected to accelerate the effects described here. Such effects could be included in more sophisticated models in the future.

Because stylolites accelerate weathering at the outcrop scale, they will also contribute to enhanced carbonate weathering on the global scale. However, estimating this contribution is challenging. While stylolites are ubiquitous in carbonate rocks, there is no generally accepted value for their abundance, making any assessment of their global impact on weathering poorly constrained. To resolve this issue, further studies are required that provide reliable estimates for the frequency of stylolites in carbonate rocks.

Declarations

The authors have no relevant financial or non-financial interests to disclose.

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Figures

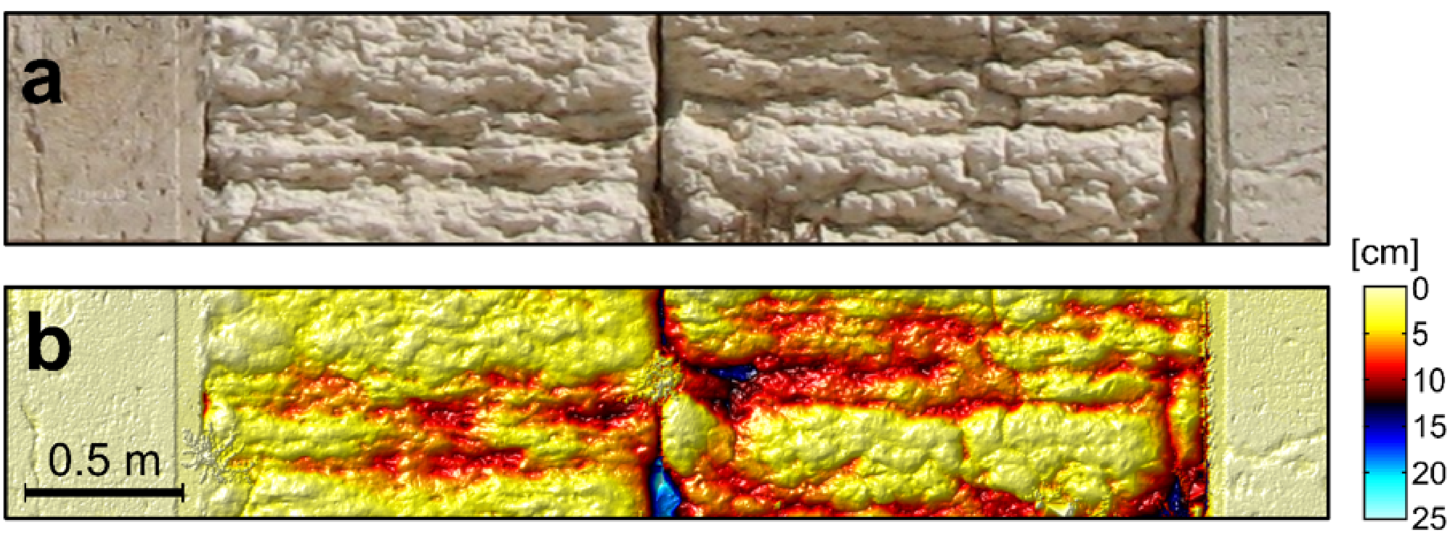

Figure 1

Example of preferential weathering of stylolite-bearing rocks at the Western Wall, Israel. (a) Digital photograph; and (b) a surface retreat map. The two central blocks have average weathering rates of 23 – 29 mm ky^{-1}, in contrast to the flanking blocks at the sides that have weathered at a rate of 1-2 mm ky^{-1}. Modified from Emmanuel and Levenson (2014).

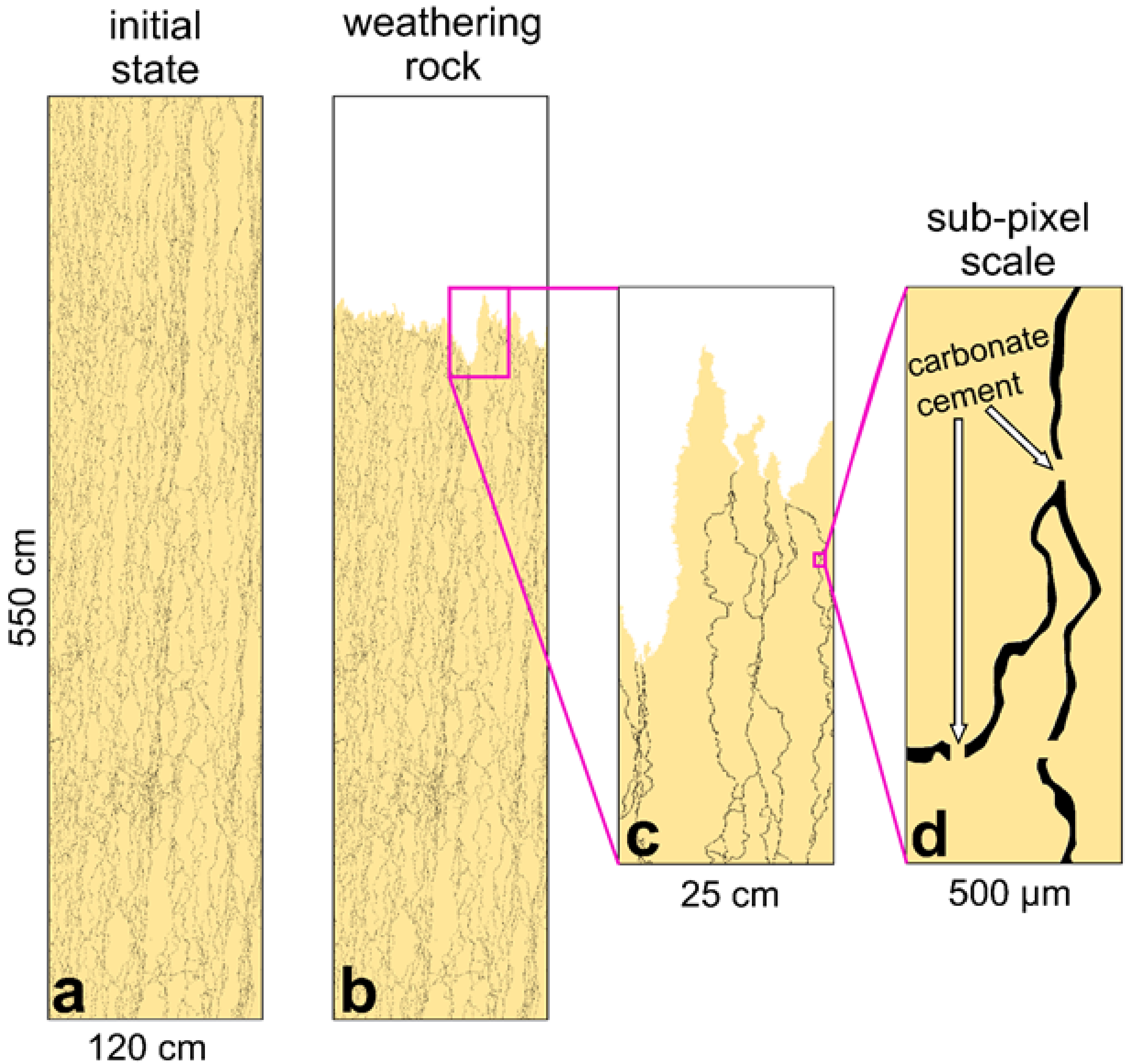

Figure 2

Representation of the digitized stylolitic limestone used in the numerical model. (a) Model domain at the beginning of the simulation and (b) during the simulation. Black lines represent the styolites, yellow represents the surrounding carbonate rock, and white represents the fluid phase. When a grain is surrounded by fluid it becomes detached. (c) Zoom in shows the roughness of the weathering interface resulting from mechanical detachment. (d) Sub-pixel scale representation of the carbonate cement bridges (highlighted by the white arrows) between two opposing stylolite sides. When there are fewer bridges, the rate at which the dissolution front moves along the stylolite will be greater. This results in the stylolite losing its cohesion more rapidly, leading to accelerated mechanical weathering.

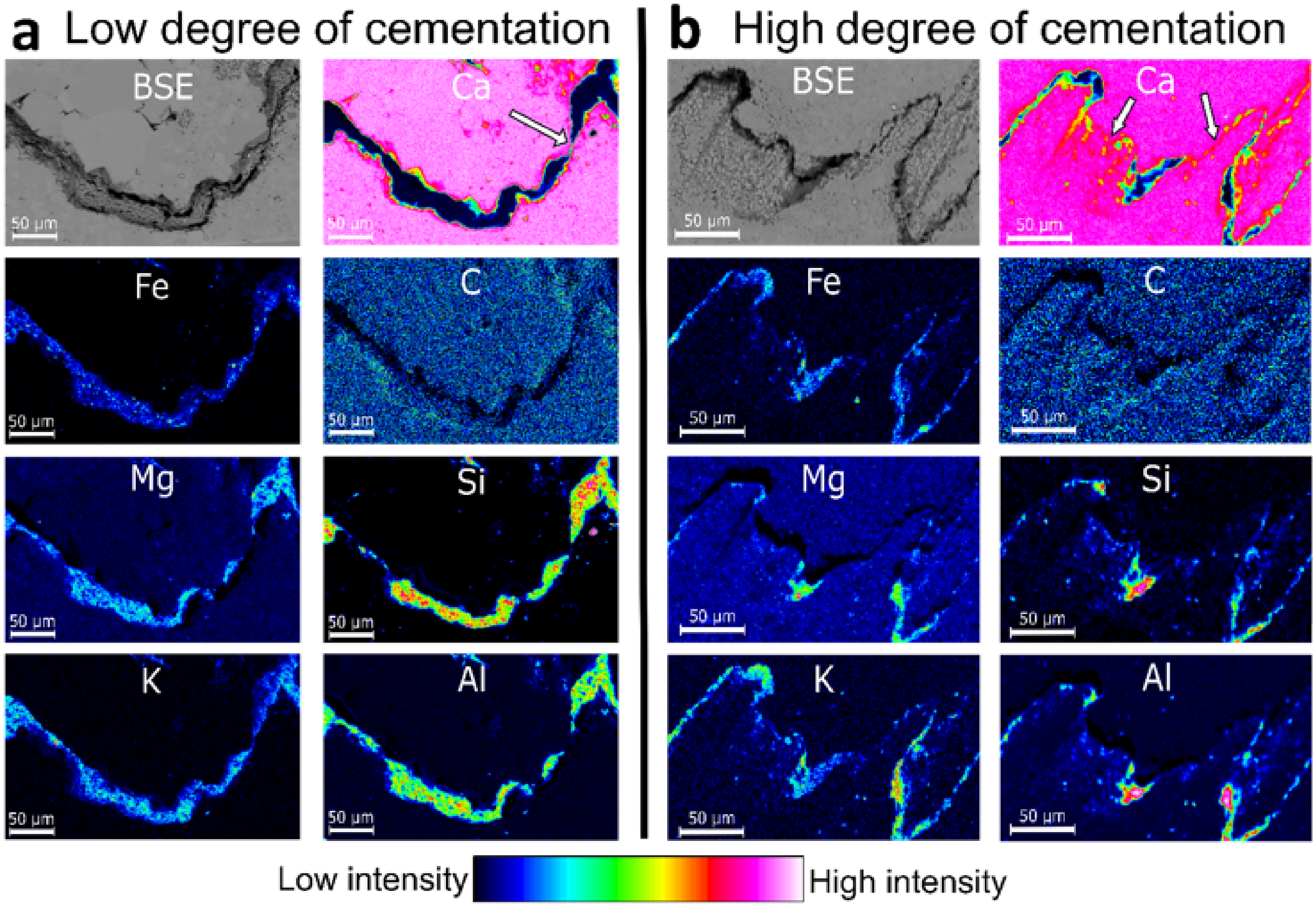

Figure 3

Backscattered electron (BSE) image and elemental (WDS) maps of stylolitic limestone from 2 different locations from the Bina Formation in the Judean Hills, Israel. (a) Sample with a low degree of cementation sampled from Ramat Shlomo exhibiting ~2% of calcite cementation (highlighted by the white arrow). (b) Region with a high degree of calcite cementation (~30%; white arrows) in a sample from Givat HaTanach. Elemental maps are shown for Ca, C, Fe, Si, Mg, Al, and K. In both samples, much of the stylolite is filled with clay minerals and Fe-oxides.

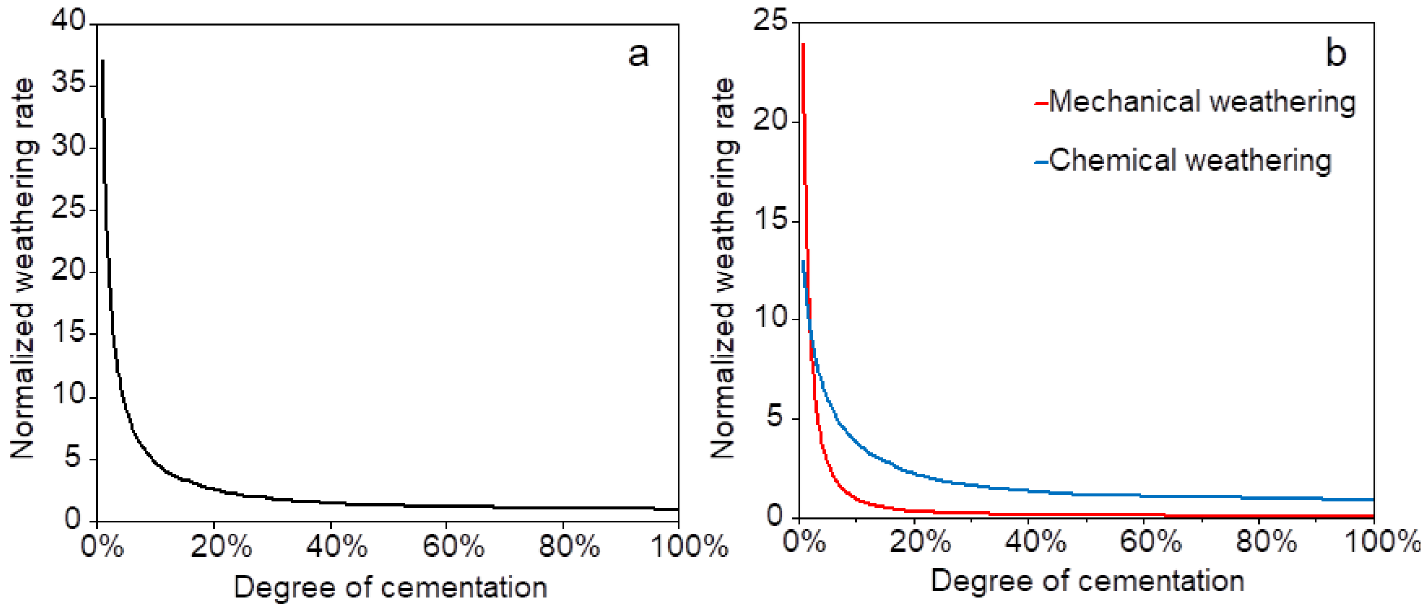

Figure 4

Simulated weathering rates as a function of the degree of carbonate cementation. (a) Total rate; and (b) mechanical and chemical rates. Rates are normalized to the total rate of weathering at 100% cementation. At high levels of cementation, weathering is primarily chemical; at low levels of cementation, mechanical weathering dominates.

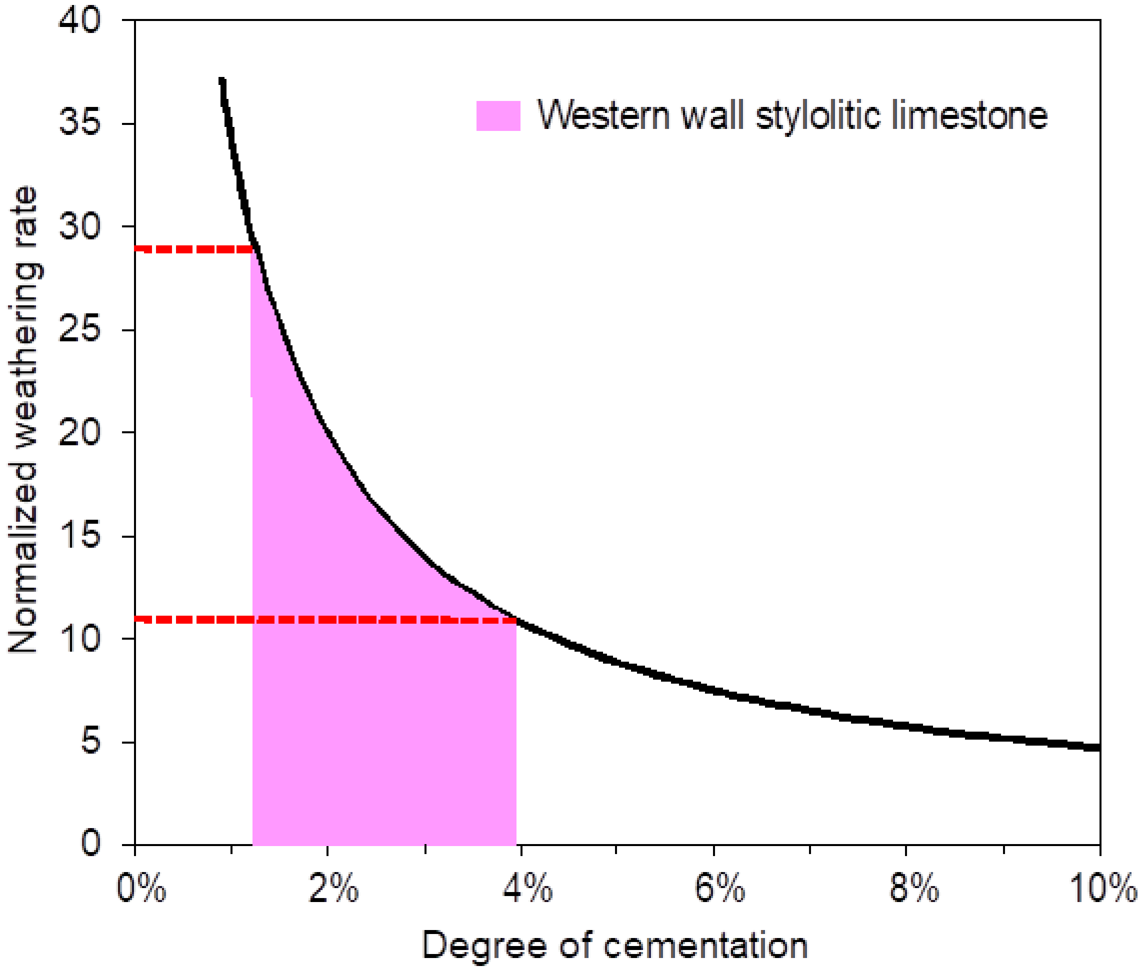

Figure 5

Prediction of degree of cementation in Western Wall stylolitic limestone based on a comparison of measured weathering rates with the numerical simulations. The horizontal red lines represent the range of weathering rates in stylolite bearing rocks normalized to weathering rates in stylolite-free rocks estimated for the Western Wall based on data reported by Emmanuel and Levenson (2014). The pink region shows the corresponding range of carbonate cementation predicted by the simulations (1.3% to 3.9%).