Testing the durability of hemp-based mortars under Mediterranean climatic conditions in coastal and inland areas: does the presence of salt alter hemp bio receptivity?

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Abstract

In this study, we assessed the durability of hemp-based mortars by means of accelerated weathering tests, simulating coastal and inland areas with a Mediterranean climate. Mortars were produced with dry hemp hurds (i.e. inner part of the stem) and three binders: dry hydrated lime, lime putty and natural hydraulic lime. Simulation of temperature and relative humidity variations, rainfall and salt attack were carried out in an environmental cabinet and programmed according to the Mediterranean climatic conditions. Only one cycle (lasting 12 days) was simulated, composed of four main steps (each step corresponding to one season). After the test, mortars did not show any apparent damage apart from the loss of some fibres from the edges of samples. Mineralogical (by means of X-ray diffraction), microscopical (by means of environmental scanning electron microscopy) and microbiological studies on samples allowed a more detailed assessment of the response of the three types of mortars to weathering conditions, especially when they were combined with the attack of salt. In particular, it was found that the presence of salt delayed mortar hardening under the simulated weathering conditions and that it induced the colonization of more microbial species. However, salt was mainly washed away by the rainfall simulated in the cabinet; therefore it did not cause any efflorescence on, or damage to, the samples. Among the three mortar types, the one with natural hydraulic lime showed the least deterioration (lowest mass variation and chromatic change) and was attacked by the lowest number of microbe species. This study demonstrates that hemp-lime mortars perform well under Mediterranean climates and that their use in coastal areas is also suitable, provided that natural hydraulic lime is used and that protective measures against salt attack are adopted during the first weeks after mortar application.

Keywords: hemp hurd, lime, weathering, sodium chloride, microbes

1 Introduction

The addition of fibres (e.g. animal hairs, plant fibres) to mortars is an ancient and effective method of improving flexibility, adhesion and strength of these building materials. In addition, hemp hurds (i.e. inner woody part of the stem) provide thermal and insulating properties [1] and are efficient CO_2-sequesters [2], reasons why their use is being strongly encouraged in recent decades for the elaboration of lime-based mortars.

Despite these advantages, hemp hurds increase the water absorption of mortars [3], which may reduce their durability. Moreover, although lime acts as a disinfectant (because of its high pH), hemp mortars are still likely to be subjected to microorganism attack when exposed to humidity variations for long period, due to the organic nature of hemp that enhances mortar bioreceptivity. Since the growth of microorganisms is strongly influenced by conditions of temperature and relative humidity, it is fundamental to study hemp mortar behaviour under real environmental conditions in order to evaluate the advantages and disadvantages of the use of hemp-based building materials in specific areas.

In this work, hemp mortar durability was assessed by simulating one accelerated weathering cycle under the environmental conditions registered during the year 2012 in different cities/countries with a Mediterranean climate. Half of the mortar samples were also exposed to salt spray to simulate the effects of airborne salt on mortar durability in coastal area. Considering that soluble salts may affect microorganism growth, favouring halophytic (i.e. salt tolerant) species [4], we studied how bioreceptivity of hemp-based mortars is influenced by the presence of sodium chloride (e.g. growth of different number or type of microbe species). Hemp mortars were produced with three types of lime (aerial dry hydrated and putty and natural hydraulic), so as to investigate also the influence of the binder on the durability of this peculiar building material.

Although one weathering cycle, corresponding to one year, is not sufficient to fully predict the durability that mortars will exhibit in the building, where they will be exposed to the same conditions repeatedly and for a longer period of time, the present work still represents an important step in the comparative study that needs to be undertaken before the use of any repair materials. Comparing the weathering behavior of hemp mortars made with different lime types, indeed, is helpful to decide which type of lime is most suitable under specific environmental conditions.

2 Materials and Methods

2.1 Materials

Mortars were prepared with commercial hemp hurds (Cannhabitat®, produced by AgroFibre, Euralis, Cazeres, France, and supplied by Cannabric, Guadix, Granada, Spain) and three types of lime: aerial dry hydrated lime in the form of powder (CL90S, [5], produced by ANCASA, Seville, Spain), aerial lime in the form of putty stored under water for 2 years (CL90-S PL, [5], produced by ComCal, Barcelona, Spain); and natural hydraulic lime (NHL3.5, [5], produced by Socli, Italcementi Group, Izaourt, France). The lime:hemp:water dosage by volume was 3:5:2.5 (in the case of the lime putty the same volume of lime and hemp but a lower amount of water were used). Mortars were named as C, N and P, according to the type of lime (CL90S, NHL 3.5 and CL90S PL, respectively). They were cured in the Cannabric factory base for three months under average conditions of T = 17 ºC and RH = 75% before the study.

2.2 Weathering test

In order to simulate Mediterranean climatic conditions a synthetic year of data was obtained from temperature and relative humidity variations and rainfall registered during the year 2012 in different cities/countries with a Mediterranean climate (Fig. 1a). The data were used to provide an accelerated simulation in a Sanyo-FE 300H/MP/R20 environmental cabinet (Fig. 1b), with one year condensed into 12 days.

As shown in Fig. 1, the whole year was simulated (one cycle composed of four main steps), with conditions changing seasonally (a step every season). The values of temperature and relative humidity represented in each step were obtained from the main highest and lowest T and RH values registered during each season of 2012. Rainfall was simulated at the beginning of each season (as indicated by the arrows in Fig. 1) applying mist water by a hose placed inside the cabinet. The time of water application was calculated by converting the amount of rain received seasonally (from millimetres to minutes of application) by means of an experimental rain gauge. To reproduce the same environment in coastal areas, the presence of airborne salt was also simulated, by spraying half of the mortar samples with a half-saturated NaCl solution. The attack of salt was programmed at the beginning of every season as well as the rainfall, to be able to compare sample behaviour in presence and absence of salt. In the cabinet, six samples per mortar type (2×2×4 cm) were placed with the largest surface (4×4 cm) facing the spray.

Samples were weighed (to an accuracy of ±0.01 g) and photos taken at the end of each cycle (season) in order to record any visible change in mortars.

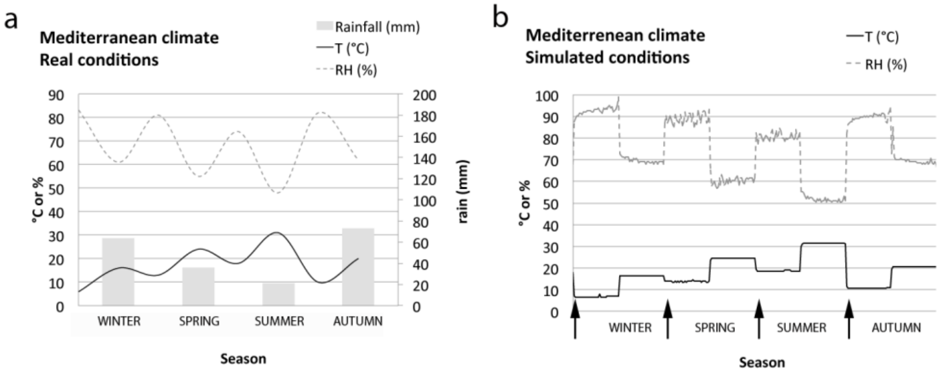

Figure 1: Mediterranean climate: real in 2012 (a) and simulated (b) conditions of temperature (T, in °C), relative humidity (RH, in %) and rainfall over one year cycle. In b, conditions were registered by means of an ibutton® Hygrolog. Arrows indicate the rainfall events.

2.2.1 Study before and after the weathering test

Mortar mineral phases were studied by means of X-ray diffraction (XRD) analysis, using a Panalytical X'Pert PRO MPD diffractometer, with automatic loader. Analysis conditions were: radiation CuKα (λ=1.5405 Å), 3 to 60 º2θ explored area, 45 kV voltage, 40 mA current intensity and goniometer speed using a Si-detector X'Celerator of 0.01 º2θ/s. The interpretation and identification of the mineral phases was performed using the X-Powder© software [6].

Microscopic observations of mortars were performed by means of a Philips Quanta 400 environmental scanning electron microscope (ESEM), which worked at a fixed temperature of 2 ºC. Small pieces of mortars (~5 mm) were directly put in the chamber, which was initially purged 5 times at a range of pressures between 2.5 and 5.5 torr (HR ~ 50% and 100% at T = 2 ºC). Once equilibrium was achieved, pressure was fixed at 3.5 torr (RH ~ 70% at T = 2 ºC).

Mortar chromatic parameters (L*, lightness; a* and b*, chromatism) were measured by means of a portable Konica-Minolta CM-700d spectrophotometer. The CIELab system was chosen [7] and the measurement conditions were as follows: measurement area of 8 mm, D65 standard illuminant and observer 10° with modes SCI/SCE and a wavelength range from 400 nm to 700 nm with a wavelength interval of 10 nm. Ten measurements per mortar type were performed. The overall colour difference (ΔE) of the three mortar types before and after the weathering test was determined as follows.

$$\Delta E = \sqrt{\left((L_1^* - L_2^*)^2 + (a_1^* - a_2^*)^2 + (b_1^* - b_2^*)^2\right)} \qquad (1)$$

where L_1^*, a_1^* and b_1^* are respectively the lightness and the chromatic coordinates of the untreated samples and L_2^*, a_2^* and b_2^* are those of samples after the water absorption-drying or the capillary tests.

Finally, a microbiological study was performed on the mortars after the weathering test. Swab samples (sterilized by ethylene oxide and individually wrapped in peel-pack) deemed suitable for isolations in culture media (Class IIa) (Eurotubo, Deltalab, Rubí, Spain) and adhesive tape samples were collected to characterize the microbial community present in the mortars, both from areas showing colour changes. Samples were inoculated onto plates containing Trypticase soy agar (TSA, Scharlau Chemie S.A., Barcelona, Spain) and Sabouraud chloramphenicol agar (Scharlau) media (100 µL of the suspension obtained per plate) and incubated at 28 ºC for one week. During this period, colonies exhibiting different morphology and appearance were transferred to new culture plates of TSA medium for bacteria and potato dextrose agar (PDA) for fungi, to obtain pure strains. Phenotypic characterization of isolated microorganisms was performed by observation of macroscopic features such as colour, shape and texture of colonies appeared in the culture media. Hyphae, sporangia and spores of fungi have been visualized by staining with lactophenol blue. Bacteria were identified by Gram staining. Observation of the samples was performed with a Leitz Dialux 22 optical microscope with objectives of 60X/100X. Images were obtained with an Olympus Camedia C-5060 camera coupled to the microscope.

3 Results and discussion

3.1 Mortar mass variation and appearance before and after the weathering test

The variations of mortar weight due to the weathering conditions in the cabinet are shown in Fig. 2. Sample weight increased accordingly with the amount of rainwater sprayed in the cabinet and the temperature conditions of each season. As shown in Fig. 1a, indeed, the rainiest periods (winter and autumn) follow the warmest ones (spring and summer) and vice versa. In samples without salt (Fig. 2a), a slight weight increase was registered after the winter season (ΔM ~ 5%), but a much greater water absorption happened during autumn (ΔM ~ 20%), due to both the greater amount of rain water during this season and the higher temperature values simulated in summer (Fig. 1a), which dried samples and increased their water absorption capacity. Samples with salt followed a similar trend during winter and autumn although a lower weight increase was registered (ΔM ~ 3% and 10% after winter and autumn, respectively) compared to samples without salt. This finding could suggest that the presence of salt reduced the water absorption by the hemp hurds or, more likely, that the water was mainly absorbed by the sodium chloride, which was totally or in part washed away, due to the action of falling water. The second reason would explain why the weight increase in samples with salt is not higher than in samples without salt, as expected.

Regarding the behaviour of the three types of lime, the N samples (made with natural hydraulic lime) absorbed less water/salt solution during the weathering test, compared to samples made with aerial lime (C and P). This is in agreement with a previous study on the hygric behaviour of these mortars [8], where it was found that hemp mortars made with natural hydraulic lime have a lower water absorption capacity compared to those made with aerial lime.

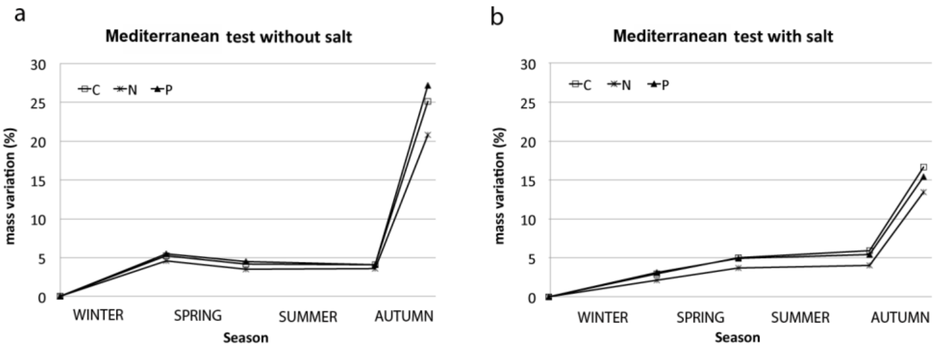

Figure 2: Mass variation (in %) of C, N and P samples (hemp mortars with CL90S, NHL 3.5 and CL90S PL limes, respectively) over one year cycle during the Mediterranean test without (a) and with (b) salt.

Images taken before and after the test show that mortar samples maintained the same appearance during the whole duration of the weathering test, both without and with the application of salt. In particular, Fig. 3 shows the appearance of samples with salt, before and after the weathering test. No efflorescences can be seen on the sample surface but only the loss of few hemp hurds, possibly caused by sample handling more than by the weathering conditions simulated in the cabinet.

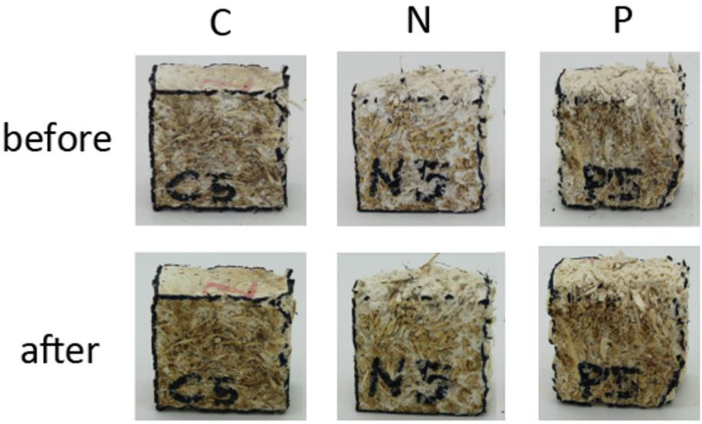

Figure 3: Appearance of C (left), N (middle) and P (right) samples (hemp mortars with CL90S, NHL 3.5 and CL90S PL limes, respectively) before and after the Mediterranean test with salt.

3.2 Mortar mineral phases and microscopic observations

Weathering clearly improved mortar hardening in comparison with control samples not exposed to the 12 days of weathering simulation, as it increased carbonation in samples produced with aerial lime (C, with dry hydrated lime and P, with lime putty) and hydration in samples with natural hydraulic lime (N). This can be observed from the decrease in the content of portlandite (Table 1) that turned into calcium carbonate (mainly calcite but also vaterite) in C and P samples, as well as in the decrease in the content of calcium silicate phases (CS) that turned into calcium silicate hydrates (CSH) in N samples.

Table 1: Semi-quantitative analysis of the mineral phases present in C (a), N (b) and P (c) samples (hemp mortars with CL90S, NHL 3.5 and CL90S PL limes, respectively), carried out by X-ray diffraction after the Mediterranean test without and with salt. Legend: Port: portlandite; Cal: calcite; Vat: vaterite; CS: calcium silicates; CSH: calcium silicate hydrates; Hal: halite; - absent; tr ≤ 2%; * < 10%; ** = 10-20%; *** = 30-50%; **** = 60-80%

C sample						
	Port	Cal	Vat	CS	CSH	Hal
Before weathering	***	***	**	-	-	-
After weathering without salt	*	****	**	-	-	-
After weathering with salt	**	****	**	-	-	tr
N sample						
	Port	Cal	Vat	CS	CSH	Hal
Before weathering	*	***	**	***	**	-
After weathering without salt	*	***	**	**	**	-
After weathering with salt	*	***	**	**	**	tr
P sample						
	Port	Cal	Vat	CS	CSH	Hal
Before weathering	****	***	**	-	-	-
After weathering without salt	**	***	**	-	-	-
After weathering with salt	***	***	**	-	-	tr

The presence of salt, however, delayed this further carbonation, since a slightly higher amount of portlandite was found in all samples compared to those not subjected to salt application. This might have been caused by the closure of some pores in the matrix where sodium chloride precipitated. Halite (NaCl), however, was only detected in traces in the three mortar types and this suggests that it has been mostly washed away during rainfall simulated in the cabinet. This was verified by means of ESEM, as no halite crystals were recognized in the mortar matrix of the three samples with salt. Instead, some spores were observed in the C sample with salt (Fig. 4).

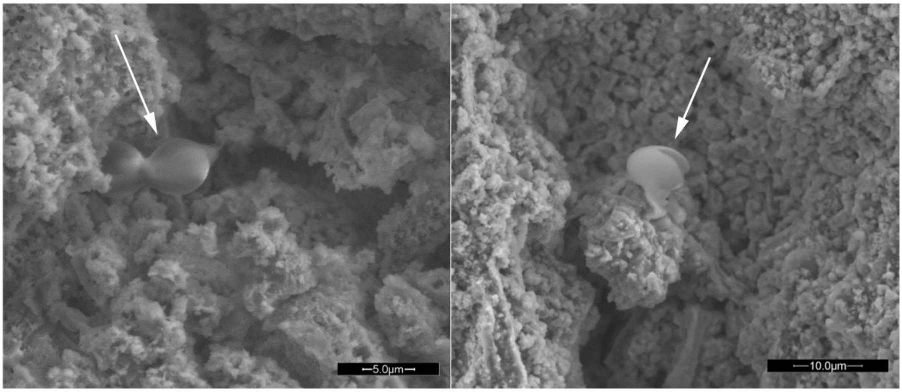

Figure 4: ESEM images of C sample (hemp mortars with CL90S) after the Mediterranean test with salt. Arrows indicate spores.

3.3 Chromatic changes and microbiological study

Sample color was measured before and after the weathering test to determine any chromatic change due to the environmental conditions or the presence of salt. Figure 5 shows that the biggest chromatic change occurred in samples made with aerial lime (especially C, with dry hydrated lime and P, with lime putty) and in those not subjected to salt application.

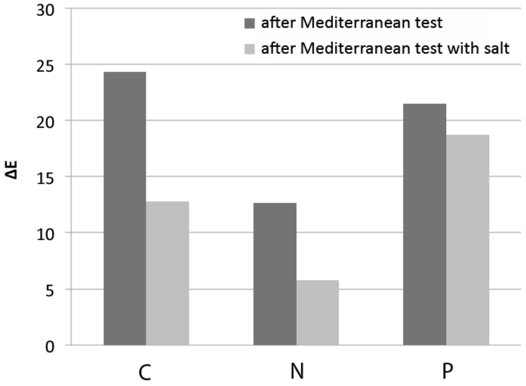

Figure 5: ΔE values of C, N and P samples (hemp mortars with CL90S, NHL 3.5 and CL90S PL limes, respectively) showing the colour difference before and after the Mediterranean test without and with salt, representing the chromatic changes of mortars.

Among the three mortar types, C and P samples were also the ones that presented a bigger mass variation (Fig. 2); therefore they absorbed more water during the test, which may have induced a more intense chromatic change with respect to N samples. The change in color (represented by the ΔE value) resulted in a general yellowing (increase of the b* value) and darkening (decrease in L* value, i.e. loss of lightness) of the sample surface. In samples with salt, this chromatic change might have been balanced by the whiteness conferred by the salt, giving a lower value of ΔE in the three mortar type. These chromatic variations, however, were not recognizable at first sight (Fig. 3), since neither visible efflorescences nor microbial attack (in the form of dark stains or mould) were observed in mortar samples. Notwithstanding, in all samples Gram-positive bacilli (of the Bacillus group, forming endospores) and in C and P samples Gram-positive cocci (of the Micrococcus group) were detected. These are very common bacteria, as they are resistant to most environmental conditions. Also different types of fungi and one type of yeast (*Rhodotorula*) were isolated, as indicated in Table 2.

Table 2: Species isolated in C , N and P samples (hemp mortars with CL90S, NHL 3.5 and CL90S PL limes, respectively) after the Mediterranean test without and with salt.

	C samples		N samples		P samples	
Species	Without salt	With salt	Without salt	With salt	Without salt	With salt
Alternaria					X	X
Aspergillus	X				X	
Chaetomium		X				
Cladosporium		X				
Penicillium	X		X	X	X	X
Phoma		X		X		
Trichoderma		X				
Ulocladium			X			X
Rhodotorula		X				

The bacteria isolated in mortars after weathering were already present in the hemp hurds. The presence of hemp, indeed, is the main cause of bio deterioration in these mortars, due to its organic nature and its ability of retaining large amounts of water. The other species, instead, formed during weathering in the cabinet. The number of species increases in mortars treated with salt, indeed it is higher in samples produced with dry hydrated lime (C samples) and it is the same in the other two mortar types (N and P). In summary: *Aspergillus* was present only in samples without salt; *Chaetomiun*, *Cladosporium*, *Phoma*, *Trichoderma* and *Rhodotorula* were only present in samples with salt; *Alternaria*, *Ulocladium* and mainly

Penicillium were isolated in both samples (without and with salt). Isolated filamentous fungi belong to the phylum Ascomycota, whilst *Rhodotorula* is yeast belonging to the phylum Basidiomycota.

The species isolated in samples with salt are halotolerant/xerotolerant, which means that they can survive and grow in high concentration of salt, although the best growth occurs in the absence of salt [9]. Halotolerant microorganisms are indeed able to grow at low water activity (a_w is the partial vapor pressure in the growing medium (P) divided by the partial vapor pressure of pure water (P_0)). This parameter (a_w) refers to the amount of water metabolically available to microorganisms and it decreases with increasing amounts of solutes in the medium. The lowest values of water activity for filamentous fungi and yeast are $a_w > 0.80$ and $a_w > 0.85$ respectively. The tolerance to low a_w was demonstrated for fungi from Ascomycota and Basidiomycota fila. Yeast and yeast-like *Rhodotorula, Debaryomyces, Aureobasidium, Trichosporum*, as well as filamentous fungi *Cladosporium, Scopulariopsis, Alternaria* [9], *Phoma, Trichoderma* [10], *Chaetomium* [11] and different species of the genera *Aspergillus* and *Penicillium* [12] are described as halotolerant.

When a microorganism is on a substrate with a water activity lower than the needed one, growth stops but the microorganism remains capable of resistance for certain period of time. For spores, the phase of resistance in a salty environment (low a_w) can be considered virtually unlimited. Indeed, spores are very likely to remain in a quiescent state in samples, until environmental conditions are again favourable for their growth.

Water activity is influenced by other environmental parameters such as pH, temperature and relative humidity. In the specific case of the weathering test performed here, the washing of the sample surface undertaken by water during the rainfall period in the cabinet, at least reduced the possibility of salt efflorescences, thus ensuring a limited damage by microorganisms, since the deposit of salt is an ideal habitat for the development of more halofile microbes [4].

4 Conclusions

This study claimed to assess the durability of hemp-based mortars under Mediterranean climatic conditions. In particular, the influence of the presence of sodium chloride on the bio receptivity of these materials was studied here, to compare the behavior of these mortars in coastal vs inland areas.

In general, the behavior of the hemp mortars under variable temperature and relative humidity conditions and rainfall shows improved mortar hardening, with accelerated carbonation and hydration processes in aerial and hydraulic limes, respectively. The same happened in samples subjected to salt attack, although in this case carbonation and hydration were slower than in samples without salt. This indicates that the presence of salt in coastal areas delays hardening of hemp-based mortars, one reason why protective measures to reduce salt attack need to be taken during the first weeks after mortar application.

As expected, the presence of salt influenced mortar bio receptivity, indeed five new species (of fungi and yeast) were isolated only in samples subjected to salt attack, in addition to two types of bacteria already present in the non-weathered hemp mortars. The presence of nutrients (organic matter proceeding from hemp) and pre-existent microorganisms (bacteria) in the material substrate, as well as the variable environmental conditions and the attack of salt were determinant in the bio-colonization of the hemp mortars. Biological attack is an unavoidable deterioration factor in building materials and obviously the bio receptivity is increased when natural components, such as fibers and hurds, are used. However, the addition of hemp hurds to mortars is still a convenient solution in specific cases where the thermal and acoustic insulating properties of a building need to be improved. Investigating the weathering behavior of this peculiar building material allows better information about its response to different climatic conditions with the consequence that specific preventive measures can be taken to ensure longer durability. In this regard, disinfectant pre-treatments of the hemp hurds would be beneficial to reduce mortar bio receptivity.

Regarding the binder type, natural hydraulic lime was found to be the most suitable for use in combination with hemp hurds, since it slightly reduced the water absorption compared to the aerial lime and it also ensured reduced chromatic change and bio deterioration. Samples prepared with natural hydraulic lime, indeed, presented the least amount of fungi species (only three types in all samples), compared to samples with aerial lime (up to seven different species isolated).

It is worth highlighting that mortar samples did not show any apparent damage during or at the end of the durability test, apart from a slight darkening and yellowing of the sample surface, not even evident at first sight. In particular, salt deposition in form of efflorescences was hindered by the continuous washing undertaken by water during rainfall. This means that, using the correct binder (natural hydraulic lime) and taking the necessary preventive measures as suggested above, hemp-based mortars would be totally suitable for use in inland and coastal areas with Mediterranean climate.

Acknowledgements

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References

[1] Newman G. European decortication and fibre make. Biomaterials-back to the future. Plant fibre technology. (2008) From: http://www.compositesinnovation.ca/biofibre_workshop/, consulted on-line in January 2014.

[2] Pervaiz M., Sain M.M. Carbon storage potential in natural fiber composites. Resources, Conservation and Recycling (39) (2003) 325-340.

[3] Nozahic V, Amziane S, Torrent G, Saïdi K, De Baynast H. Design of green concrete made of plant-derived aggregates and a pumice-lime binder. Cem Concr Comp (34) (2012) 231-241.

[4] Altieri A., Pinna D. Monumenti e manufatti in ambienti costieri. La biologia vegetale per i Beni Culturali. Vol. I Biodeterioramento e conservazione. Caneva G. Nugari M.P., Salvadori O. Collana Arte e Restauro, Nardini Ed., Firenze (2005) 196-199.

[5] BS-EN 459-1. Building Lime. Part 1: Definitions, specifications and conformity criteria. BSI, Standards Publication, London (2010).

[6] Martín Ramos, J.D. XPowder. A software package for powder X-ray diffraction analysis. Lgl. Dep. GR 1001/04 (2004).

[7] UNE-EN 15886. Conservation of cultural property - Test methods - Colour measurement of surfaces. Spanish Association for Standardization and Certification, Madrid (2011).

[8] Arizzi A, Brümmer M, Martín Sánchez I, Cultrone G, Viles H. The influence of the type of lime on the hygric behaviour of hemp-based mortars. Submitted to Cem Concr Compos (2014).

[9] Kivistö, A. T., and Karp, M.. 1,3-propanediol production and tolerance of a halophilic fermentative bacterium, Halanaerobium saccharolyticum subsp. saccharolyticum. J. Biotechnol. (152) (2011) 114–124.

[10] López-Archilla, A.I. , A.E. González, M.C. Terrón, and R. Amils. Diversity and ecological relationships of the fungal populations of an acidic river of Southwestern Spain: The Tinto river. Can. J. Microbiol. (50) (2004) 923–934.

[11] Qasemian L , Billette C , Guiral D , Alazard E , Moinard M , Farnet AM . Halotolerant laccases from Chaetomium sp., Xylogone sphaerospora, and Coprinopsis sp. isolated from a Mediterranean coastal area. Fungal Biol. (116) (2012) 1090-1098.

[12] Butinar L, Zalar P, Frisvad JC, Gunde-Cimerman N. FEMS Microbiology Ecology (51) (2005) 155–166.