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AN ANALYTICAL INVESTIGATION AND A MYCOLOGICAL STUDY OF SOME ARCHITECTURAL STUCCO MONUMENTS IN THE PERIOD OF MAMLUK AND OTTOMAN RULE OF THE ISLAMIC ERA, EGYPT.

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ABSTRACT

Architectural stucco carvings suffer from many aspects of deterioration due to environmental corrosion factors, especially moisture. Moisture is the key factor for the establishment and spread of fungi within the stucco granules, and it leads to the surface decomposition and fall of the stucco surface parts near the end of the incubation period. Analyses and investigations have been conducted on the stucco material, including XRD and ESEM-EDX analyses. As the main stucco component, Gypsum is occasionally replaced with a Hemi-hydrated phase, such as bassanite in one sample, in addition to traces of halite, calcite and silicates. Relative humidity, temperature and fungal infection were monitored to assess their interrelationship and impact on stucco deterioration. Isolated fungi were identified and tested for their ability to grow on mockups with the same components of examined archaeological stucco. Eventually, moisture and salinity are expected to activate specific physical and chemical reactions and fungal growth,

الملخص

تعاني المنحوتات الجصية المعمارية من العديد من جوانب التدهور بسبب عوامل التآكل البيئي ، وخاصة الرطوبة. الرطوبة هي العامل الرئيسي لتوطين وانتشار الفطريات داخل حبيبات الجص، التي تؤدي إلى تحلل وسقوط أجزاء من سطح الجص قرب نهاية فترة الحضانة. تم إجراء الفحص و التحليل لمادة الجص ، بما في ذلك تحليلات XRD و ESEM-EDX . الجبس هو المكون الرئيسي للجص في كل العينات، ولكن قد تم استبدالة بالباسانيت bassanite في احد العينات ، بالإضافة إلى انه قد تم الحصول على آثار من الملح والكالسيت والسيليكات في غالبة العينات. كما تم قياس الرطوبة النسبية ودرجة الحرارة والعدوى الفطرية لتقييم العلاقات المتبادلة وتأثيراتها على تدهور الجص. تم تحديد واختبار الفطريات المعزولة لمعرفة قدرتها على النمو على نماذج بالأحجار الطبيعية مع نفس مكونات الجص الأثري الذي تم فحصه. وفي نهاية المطاف ، نستطيع القول ان أن الرطوبة والملوحة ادت إلى تنشيط بعض التفاعلات الفيزيائية والكيميائية وتشجيع نمو الفطر ، مما ادى الى البهتان والسمرة والسواد والتلف البيولوجي للأسطح الجصية.

which facilitate dullness, brownness, blackness and the biodeterioration of stucco surfaces

KEYWORDS
stucco, fungi, museum, archaeological mosques, semi-simulation, XRD- EDX analyses

الكلمات الدالة
الجص؛ الفطريات؛ متحف؛ المساجد الاثرية؛ تحليلات XRD و ESEM–EDX

INTRODUCTION:

Architectural stucco carvings in mosques, ancient houses, palaces and those exhibited in museums are in danger; therefore, their degradation causes high economic impacts. Egypt is rich in stucco carvings, whether as decorative architectural elements inside buildings or as museum pieces like alternate heads, stucco masks and cartonnage which belong to the Pharaonic period (Afifi, 2011; Madkour, Afifi and Abd El-Fatah 2018; Abdel-Ghani, Afifi & Osman, 2020). In the Islamic period, stucco monuments vary among stucco windows, muqarnas, murals and mihrabs (Kamel et al., 2014a; Kamel et al., 2015; Eissa et al., 2015; Ali, Afifi & Lofty 2021). Gypsum was used as a plaster to cover rough stone surfaces with a layer of white mortar in Pharaonic temples and Islamic houses, mosques and palaces (Ali, Afifi, & Lofty, 2021; Shaheen 2005). It was used throughout different eras in the manufacturing of various decorative elements in Islamic buildings, and it was distinguished by its tendency to floral, calligraphy, and geometrical decorations. The first Islamic artistic style was of the Umayyad era, which used prominent stucco engraved on a large scale in the decoration of palaces. This style was followed by the Abbasid artistic style, which was influenced by Persian art. In the Fatimid era, the decorations elevated to the leafy forms and Kufic inscriptions, called arabesque. In the Ayyubid era, the decorations became more meticulous and complex as in domes (Ismail, 2006). The Mamluk era, especially the marine Mamluk period, was the golden age for stucco decorations (Kamel et al., 2014a).

Conversely, in the Ottoman period, the stucco industry deteriorated, and the use of marble and copper flourished. Finally, the Islamic eras bequeathed many stucco monuments of delicacy, perfection and beauty to us. It affected the minds of the beholders, especially the ancient monuments not only in Egypt but also in all countries belonging to the Islamic eras in the world (Geweely et al., 2020).

Gypsum is frequently used in Egypt because its raw material is abundant, it is easy to obtain, and the degree of its burning is low. However, rising temperatures lead to gypsum's dehydration and its transformation into a hemihydrate form called bassanite. It is an inorganic substance and is considered a harsh environment that does not directly contribute to microbial growth. Moreover, the existence of stucco carvings in open environmental conditions makes them vulnerable to various damage factors, like fungi. These factors are difficult to control in the spaces that were open to the elements and became, in turn, major causes of the natural damage to the ancient stucco monuments (Madkour Afifi & Abd El-Fatah, 2018). Temperature and humidity are reported to encourage the growth of fungi (Jain et al., 2009; Montanaria et al., 2012). As one of the most diverse organisms following insects in diversity, fungi are major contributors to the deterioration of archaeological works (Karpovich-Tate & Rebrikova 1990; Schabereiter-Gurtner et al., 2001; Gorbushina, et al., 2004; Scheerer, Ortega-Morales &

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Gaylarde 2009; Pepe, et al. 2010; Pangallo et al., 2012; Kavkler, et al. 2015, 51-59; Rosado 2015, 78-83; Geweely & Afifi 2011; Geweely et al., 2022). In historic buildings, churches and archaeological tombs, ascomycetes are frequently recorded on paints, while basidiomycetes and zygomycetes are commonly isolated from murals and the surrounding environment (Karpovich-Tate & Rebrikova 1990). Nevertheless, the most famous fungi found in the indoor environment of buildings are, *Aspergillus, Alternaria, Penicillium, Cladosporium* and *Stachybotrys*. They have a high enzymatic activity that enables them to damage the structure of any archaeological material (Abdel-Alim 2003; Sterflinger & Piñar 2013; Rosado et al., 2015; Geweely et al., 2019).

This paper focuses on the study of some architectural and museum archaeological stucco that belonged to Mamluk and Ottoman Rule of The Islamic Era in Cairo, Egypt. The study was carried out in four selected historical museums, mosques and houses: Gamali Yusuf Mosque, Tatar El-Higaziya Mosque, Bayt El-Kredlea Museum (Gayer-Anderson Museum) and Zeinab Khatoon House. The study explores the state of these antiquities in terms of their general appearance, type, function, origin, date of construction and method of manufacture. Moreover, the study focuses on the obvious problems of fungal damage and the surrounding factors which may facilitate the deterioration process in the museum environment. Those aspects can be identified through close-up visual examination, analysis and investigation. X-ray Diffraction Analysis (XRD) and Energy Dispersive X-Ray Analysis (SEM-EDX) are used to determine the chemical composition and the fine structure of the examined ancient stucco carvings. The isolation of fungi was undertaken to assess their role in biodeterioration. The effect of moisture on fungal colonization on simulated stucco surfaces was also carried out.

METHODS

STUDY STUCCO ANTIQUITIES

Stucco mihrab decorations of Gamali yusuf mosque:

The mosque belongs to the Circassian Mamluks period (784-932 AH/1382-1517AD) (Figure 1.a).

It suffered from numerous causes of deterioration, such as weakness, the fragility of the surface and the change of stucco surface color to gray due to dust, especially in the deep areas of the decoration.

Stucco decoration of arched recesses in Tatar El-Heigaziya mosque (Figure 1.b):

The mosque belongs to the Bahri Mamluk period (648 to 784 AH: 1250 to 1382 AD). The texture of the decoration is a perforated stucco element for three and a half roses in a head position that occupies most of the chest of that veil (Williams 2008). Tatar El-Higaziiya Mosque: The sample site called Dakhla (the arched recesses with stucco elements) is found to the left of the ascender to the western Ewan (palace) on the second level of the walls and above the Quranic scripture tape. The mosque exists in a densely populated area; the main street to the mosque contains small shops, vegetable and fruit carts, and an accumulation of garbage bags and dead animals around the mosque walls. In addition, the lack of both a good sewage network and drainage pipelines in the area led to the rise of moisture levels in the soil, which in turn is transferred to the walls of the mosque. Damages appear in the color change of stucco to brown, yellow or gray. There are also meandering cracks in the surface of the gypsum, which is covered with dust (Afifi et al. 2021).

Stucco window of bayt el-kredlea museum (gayer-anderson museum, Figure 1.c.):

It belongs to the Ottoman period (923 - 1213 AH, 1517 - 1798 AD). The museum consists of two houses: the first is the House of Amna Bint Salem, established in 947 AH / 1540 AD, and the second is the house of Muhammad bin Al Haj Salem Al-Jazzar, which is known as Bayt el-Kredlea established in 1041 AH / 1631 AD. It is also called the Gayer Anderson Museum. The colored glass stucco window (in the Byzantine Hall of the museum) is jammed and does not open. Its purpose was either constructive to reduce the weight of the building's facades or aesthetic, as these windows allow a small amount of colored light to enter the place (Afifi et al., 2020a). Fungal damage occurs with the discoloration of some areas where the characteristic white color changes to dark yellow or brown. The samples were collected from falling parts below the window flap.

Stucco window of the archaeological house of zeinab khatoon (Fig. 1.d):

It belongs to the era of the Mamluk Circassians, 784-932 AH / 1382-1517 CE). It was established during the reign of Sultan Al-Ashraf Qaitbay and has undergone several changes and restorations over the historical ages, most recently in 1996. The stacked window with colored glass is in one of the house's empty rooms, which is not utilized. It overlooks the open courtyard of the house, and it is exposed directly to the sun. Consequently, it is exposed to high temperatures, especially in the summer. The left window sash suffers from multiple forms of damage, such as a change in the white stucco color to brown and gray in more than one place. The wall below the window is exposed to moisture, which rises within the wall to the height of 80 cm, and this increases the level of moisture that reaches the window (Williams, 2008; Afifi et al., 2020b; Afifi et al., 2021). The sample was taken from a part beside a crack in the lower right section of the window.

Relative humidity and ambient temperature

The average temperature and relative humidity were determined by a Thermometer and a Hygrometer device (AcuRite 02067M) at the smearing time.

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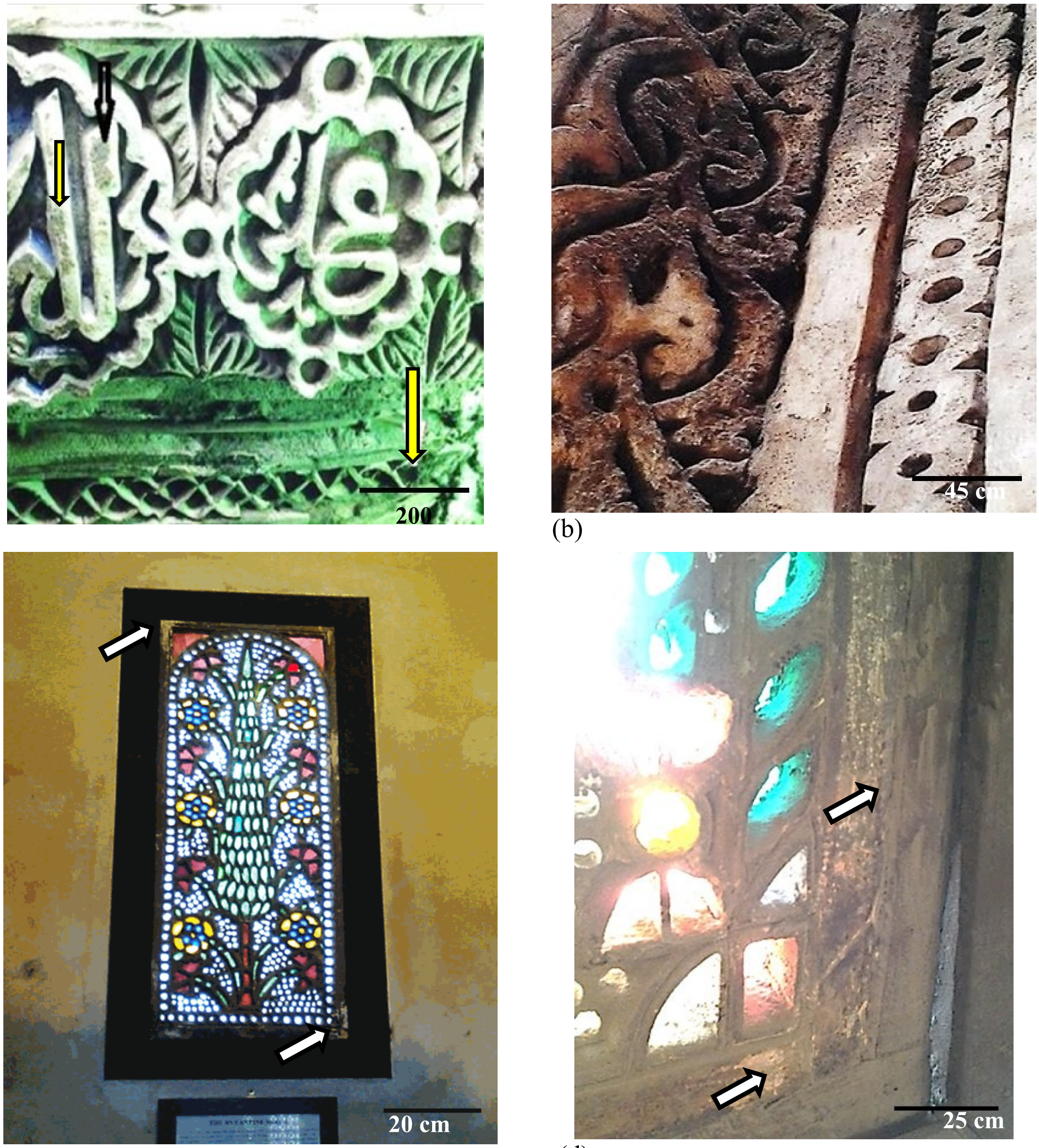

(a) (b) (c) (d)

Figure 1 Studied archeological stucco carvings: (a) sample 1 of Gamali Yusuf mosque (damages appear as changes in the color of the plaster to gray in Mihrab Wall inscriptions, 400x250 cm); (b) sample 2 of Tatar El-Higaziiya mosque window 90 x 145 cm (lower left part of the window, showing damages as a change in the color of the plaster to black, brown, yellow, gray, meandering and cracks in the surface of the gypsum; (c), Sample 3 of Bayt el-Kredlea museum window 100x40cm (damages on the upper and lower part of the window in The Byzantine Hall of the Museum as discoloration in some places where the characteristic white color is changed to dark yellow and brown; d, sample 4 of Zeynab Khatun house where the left flap of the stained glass window (100x100 cm) suffers from multiple forms of damage in the form of change in the color of the stucco from white to brown and gray in more than one place. The rest of the window shows a light change in the color of the plaster.

STUCCO ANALYSIS

An analysis is fundamental to identify the products of the stucco components deterioration, including the experimental and all conservation processes (Afifi et al., 2021; Geweely & Afifi 2011; Geweely et al., 2014). Stucco samples were analyzed using Environmental SEM with an Energy Dispersive X-Ray (EDX) unit. X-ray Diffraction Analysis (XRD) has been done by using a Diffractometer type PW1840 Philips (Corradiation, Fe filter at 40 kV, 30 mA and scanning speed 0.02/s) by X-ray powder diffraction method (Connolly 2005, EPS400-002). The dA and the relative intensities were obtained from the produced charts and were compared with JCPDS files (Smith and Berry 1967). Uncoated samples were carefully analyzed in a Philips Environmental Scanning SEM Microscope (model XL30 2000, a low discharge mode of 20-30 kV, 0.8 Tor Equipped with scattered electron diffusion meters (BSD)). The EDX analysis was performed (counting time 120-180 seconds, minimum concentration Weight: 0.1% by weight to 1% by weight, analysis depth of about 0.5-1.0μ). The accuracy of the quantitative results was 2-5% for element Z> 9 (F) and 5-10% for light elements B, C, N, O and F.

ISOLATION OF FUNGI

Sterile swabs (114 swabs) were taken from the stucco surfaces of the selected archeological objects that show a change in the stucco color under aseptic conditions. Pre-sterilized labels were placed on the surface of the stucco in different places. After removal, they were placed directly in Czapek Dox agar plates and incubated at 28◦C for 4 to7 days (Rose et al. 2004; Santucci et al. 2007; Rassana, et al. 2017). Bacterial growth was prohibited by adding Penicillin G and Streptomycin. Between measurements, samples were swabbed with 70% ethanol. After that, the growing fungal colonies were counted, isolated and stored on agar slides for further investigation. The isolated species are classified as very frequent (> 20%), frequent (10-20%) and infrequent (<10%) based on the frequency of occurrence (Tan and Leong 1989).

Semi-simulation experiment: moisture effect on fungal colonization of stucco

Preparation of experimental samples

According to the results of archaeological stuccos analytical and investigation studies, the experimental samples were prepared with the same components and proportions for each sample site with different sizes (5 x 5 cm), (2.5 x 2.5 cm) and (2.5 x 5 cm) for each fungus with a total number of 72 stucco models. Some drawings were performed and carved in deep, prominent, flat figures on the cutting surface. The surface was sterilized with alcohol. In the first experiment, stucco models were placed in sterile petri-dishes. Fungi and fungal consortiums from each site were tested. Fungal spores in suspension (1x106 /ml) were distributed on dry stucco surfaces using dry sterile cotton swabs in the sterilization chamber. They were incubated for three months at room temperature with daily observation. A second experiment was designed where the surfaces of the gypsum

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models were moisturized using wet cotton scrolls. After that, the models were infected with the same types of fungi and concentration of spores. The samples were left for three months and were followed up weekly to record any appearance of fungal growth on their surfaces.

STATISTICAL ANALYSIS

To determine the relationship between the fungi and the measured environmental variables, on the one hand, and salt impurities of the stucco, on the other hand, CCA (canonical correspondence analysis) was used through the Past program.

RESULTS AND DISCUSSION

The Egyptian cultural heritage, especially in an open museum environment, is constantly exposed to numerous climatic changes and in situ conditions which cause major internal and external damage to stucco carvings. Overall, each museum, ancient house or mosque varies in the level of fungal spores (Table 1, Figure 2) because their growth depends on air currents and air exchange in rooms resulting from ventilation or opening and closing doors. Therefore, fungi cause a persistent problem in museums due to the restorers and officials' lack of information, experience and training. Eventually, archeological mosques and museums are considered to be giant incubators that offer an exclusive microenvironment for the growth of various fungi.

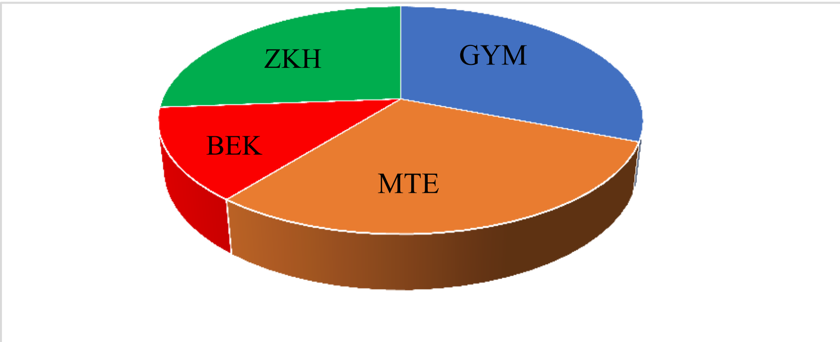

Figure 2 Bulk of fungal total colony count from Gamali Yusef Mosque (GAM), Byzantine Hall (BYZ), Mosque of Tatar El Higaziya (TAT) and Zeinab Khatoon house (ZYK).

INTERACTION BETWEEN FUNGI AND MEASURED PARAMETERS

The biodeterioration of archaeological materials is a well-recognized problem in Egypt, where environmental factors enhance the growth of microorganisms, especially fungi. Twelve fungal species were isolated and identified from the stucco samples of the studied museums (Table 1). A. niger and A. flavus (100%) were the predominant species, followed by A. flavipes, Alternaria alternata and Penicillium canescens (75%). The remaining species were very frequently isolated. The dominance of Aspergillus spp. and other hyphomycetes like Alternaria has been reported on archeological materials (Sterflinger, 2010). These fungi are significantly present in the Egyptian atmosphere because of the deterioration of internal air quality (Gutarowska & Zakowska, 2002; El-Morsy, 2006; Verdier, et al. 2014), causing aesthetic damage (Sterflinger, 2010; Mesquita 2009) and the biodeterioration of archeological materials (Johansson, 2008). Their high activity causes severe erosion (Sterflinger, 2010; Burford, Fomina & Gadd

2003; Hoffland et al. 2004). Temporal changes in relative humidity (R) and temperature (T) have a statistically similar effect on fungal distribution, where T and R changes in the winter are 18oC to 21oC and 66% to 69%, respectively (Table 2). In this regard, appropriate temperature and

Table 1: Occurrence of fungal species in studied contaminated stucco carvings of ancient Egyptian mosques, houses and museums.

Fungal species \ Sample Location		1	2	3	4	% occurrence
A. niger van tieghem.	Ani	+	+	+	+	100
A. flavus link ex Gray.	Afl	+	+	+	+	100
A. flavipes (Bain & Sart.) Thom & Church	Aff	+	+	+		75
Alternaria alternata (Fr.) Keissler	Alt		+	+	+	75
Penicillium canescens	Pca	+	+		+	75
P. chrysogenum Thom	Pch	+			+	50
P. citrinum Thom	Pci	+		+		50
Fusarium sp.	Fus		+		+	50
A. ustus (Bain.) Thom & Church.	Aus	+				25
A. clavatus Desmazieres	Acl			+		25
A. oryzae (Ahlburg) Cohn	Aor		+			25
Cladosporium cladosporoides	Cla		+			25

1; Mihrab Wall decoration of Gamali Yussef Mosque, 2; The Dakhla of Tatar El Higaziya Mosque, 3; stained colored glass plaster window in The Zeinab Khatoon house.4; Glass plaster window in The Byzantine Hall of the Museum of Bayt el-Kredlea,

Table 2. Record of the measured average of relative humidity (R) and Ambient Temperature (T) during sample collection in winter (W) and summer (S) month.

Variables		R %		T ºC	
Location		S	W	S	W
Gamali Yusef Mosque	(GAM)	34%	67 %	31º	20º
Tatar El Higaziya Mosque	(TAT)	33%	69 %	32º	21º
Byzantine Hall	(BYZ)	27%	66 %	30º	18º
Zeinab Khatoon house	(ZYK)	30%	66 %	28º	19º

humidity was reported to be between 18ºC-20ºC and between 50% - 60%, respectively (Montanaria et al., 2012), where it was stated that a relative humidity over 55% encourages the growth of fungi (Jain et al., 2009). Therefore, humidity between 66%-69% may help fungal establishment at an adequate temperature between 18º-21ºC. The relation between the isolated fungi and the stucco at recorded T, R and salinity was statistically evaluated by using canonical correspondence analysis CCA (Figure 3). It is the relationship between the first two axes, where fungi are organized based on their

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count on these axes. The relative distance of a species to a site indicates the relative abundance of that species on that site. Fungi on the edge of the diagram are often less frequent. The dominant fungi (A. niger, A. flavus and A. flavipes) are seen to be less affected by changes in T, R and salinity, so they are gathered at the center of the ordination biplot. The same conclusion was previously reported (Filipello, Airaudi & Barchi 1997; El-Morsy,1999). Because there are few variations in daily records of T and R, their effect on fungal distribution is not apparent. The stucco halite content was alike. However, temperature changes were more effective on the stucco material than the fungus, which flourished in the presence of available moisture and organic pollutants, as indicated later in this investigation.

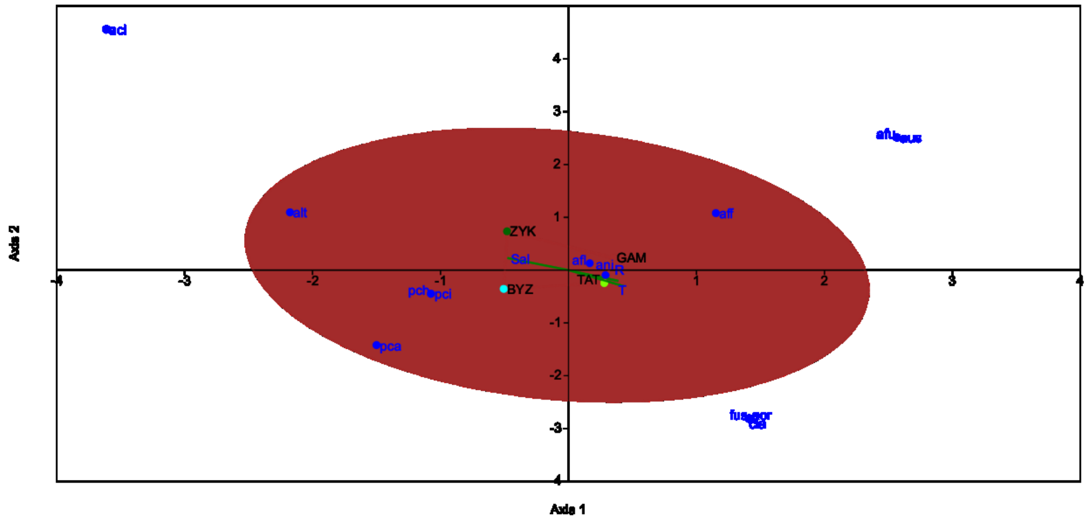

Figure 5 CCA ordination biplot showing relationships between fungal entities ( ) and median values of environmental variables (R=Relative Humidity; T= Temperature) in winter and salt constituents of the plaster (Sal) in tested ancient Egyptian mosques, houses and museums (GAM= Gamali Yussef Mosque; BYZ= Byzantine Hall; TAT= Mosque of Tatar El Higaziya; ZYK= Zeinab Khatoon house). Convex hull in the center (the intersection of all sets containing X). Ellipse 90% enclose species recorded in at least two locations. Fungal Abbreviations in Table 1.

STUCCO ANALYSIS:

X-ray diffraction (XRD) and SEM-EDAX analyses illustrated the main gypsum components in addition to some impurities that fluctuated between the two tests (Table 3). The presence of impurities in the two analyses varies because they are extraneous elements whose source is unknown and are distributed in unequal proportions in the gypsum. Thus, they appear in some but not all samples.

Table 3. X-ray diffraction (XRD) and dispersive (EDAX) analyses of collected Samples from stucco carvings of some ancient Egyptian mosques, houses and museums.

studied site	Sample position	Analysis		
		X-ray dispersive analysis (EDAX)	X-ray fraction analysis (XRD)	
			Components	%
Gamali Yussef Mosque (sample 1)	a. Above the decoration towards the fracture	Calcium (Ca), Sulphur (S).	Gypsum ($CaSO_4.2H_2O$) Silica (SiO_2) Bassanite ($2CaSO_4. H_2O$)	38% 17% 44%

	b. Inside the fracture within the decorations	Ca, S, Lead (Pb), Copper (Cu), Si	Gypsum SiO_2 Bassanite	38% 17% 44%
Mosque of Tatar El Higaziya (sample 2)	a. Above the Dakhla from the left side	Ca, S, Aluminium (Al), Cu, Si	Gypsum SiO_2 Calcite ($CaCO_3$)	57% 19% 23%
	b. The dark-colored layer in the Dakhla	Ca, S, Al, Copper, Si	Gypsum SiO_2	81% 19%
Glass plaster window in The Byzantine Hall of the Museum of Bayt el-Kredlea (sample 3)	a. from some of the falling parts of the bottom of the window	Ca, S, Chloride, Al, Silicon, Iron (Fe), Potassium (k)	Gypsum Sodium Chloride	64% 36%
	b. from the side beside the crack in the lower right part of the window	Ca, S, Chloride, Al, Si, Fe, Pb, K	Gypsum Sodium Chloride	64% 36%
Stained colored glass plaster window in The Zeinab Khatoon house (sample 4)	a. Part beside the crack in the lower right part of the window.	Ca, S, Chloride Al, Silicon, Fe, Pb, K	Gypsum Sodium Chloride Bassanite	47% 24% 29%
	b. Part beside the crack in the lower right part of the window	Ca, S, Chloride Na, K, Strontium (Sr), Barium (Ba)	Gypsum Sodium Chloride Bassanite	47% 24% 29%

X-RAY DIFFRACTION ANALYSIS

Gypsum (CaSO4.2H2O) was recorded as the major component of the stucco, except in sample 1, where bassanite (2CaSO4•H2O) was higher (44%) than gypsum (38%). That may be attributed to the high temperatures over the years, especially in the summer, which led to the transformation of the gypsum into a semi-anhydrous phase, bassanite. Bassanite, sand (in the form of SiO2), calcite (CaCO3), halite (NaCl) and anhydrite CaSO4 (11%) were frequently detected (Table 3). The highest silicate percentage was obvious in samples 1 (17%) and 2 (19%). Lead, copper, iron, potassium, aluminum, strontium, and barium were found as impurities in the gypsum powder. Calcite was detected but only in sample 1 (Table 3), probably as a result of the fungal conversion of the contaminants calcium oxides to CaCO3 producing stucco deformation in certain archeological materials (Verrecchia, Dumont & Verrecchia, 1993; Garvie et al. 2000; Arocena, Zhu & Hall, 2003; Kolo and Claeys, 2005). Despite the dark growth of fungi on Dakhla's sample, the gypsum structure appears unaffected by quartz impurities (Figure 4). Halite (NaCl) accumulation was present in a high percentage in samples 3 (36%) and 4 (24 %). The high percentage of sand and halite (NaCl) in some samples indicates that they might have been used in the stucco's original mixture. Moreover, the presence of salt in substrates supports the growth of microorganisms, especially fungi, in samples 3 and 4 (white salt efflorescence was seen in moist areas), where isolated fungi are primarily xerophilic and utilize even micronutrients; moreover, they may use osmosis to produce enough pressure to displace the metal grains (Burford, Fomina & Gadd, 2003; Otlewska et al., 2017). In addition, it was reported that halite crystallization and accumulation were found to cause the decay of porous materials of ancient houses in a variety of environments (Goudie & Viles, 1997), where salt filling the pores exert pressure on the pore walls of the ancient materials (Scherer, 1999). Thus, halite is directly or indirectly involved in ancient stucco impairment.

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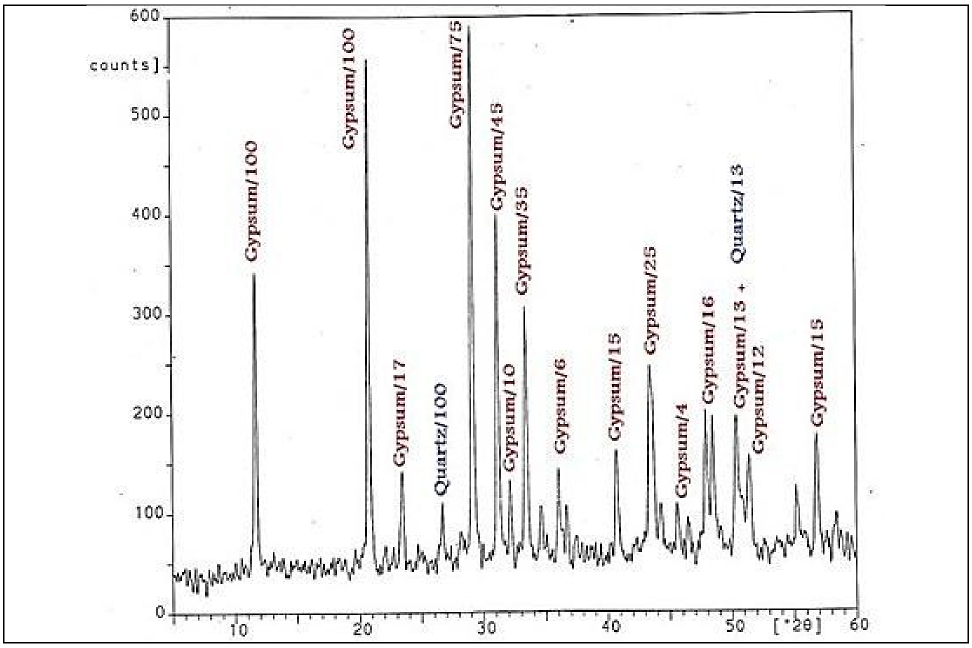
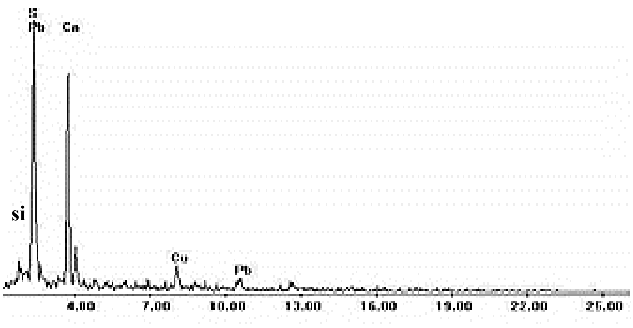


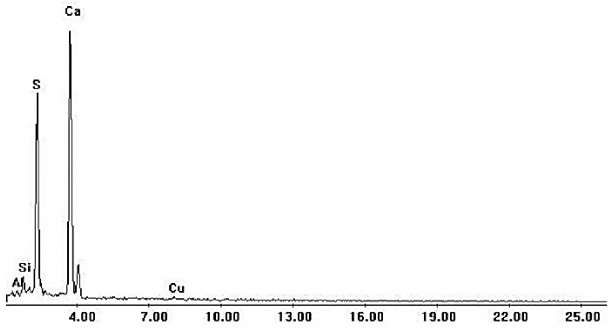
Figure 4 X-ray diffraction analysis of a sample of a dark-colored layer of The Dakhla (arched recesses with plaster elements) in Tatar El-Higaziiya mosque.

ESEM - X-RAY DISPERSION ANALYSIS

SEM results and XRD analysis (Table 3, Figure 5 a- e) illustrated that calcium Ca and sulphur S were the basic elements in the gypsum and bassanite, as well as other elements such as Na, Cl, Fe, Pb, K, Al, K, Al, Si, Sr and Ba. In Figure 5a (Gamali Yusef Mosque), calcium and sulphur were abundant because they are fundamental elements in the gypsum (38%) and the bassanite (44%). Copper and lead were found in a small percentage; their presence can be impurities in the gypsum dough. The same finding was detected in Dakhla at Tatar El-Higaziiya Mosque (Figure 5b) where calcium and sulphur were abundant and represented 57% of the gypsum and 23% of calcite with impurities of copper and aluminum. Regarding the glass window in the Byzantine room of Bayt El-Kredlea Museum, SEM results for samples from the falling parts below the windows (Figure 5c) illustrated that Ca and S are the major elements representing the gypsum (64%) followed by Na and Cl representing NaCl (36%) in addition to impurities of Fe, Pb, K, Al, and Si. In Figure 5d (the stained-glass window in Zeinab Khatoon's House), Ca and S are major constituents and represented 47% of the gypsum and 29% of bassanite, while Na and Cl represented 24% of Sodium Chloride. Other elements such as Fe, Pb, K, Al, Si, Sr and Ba were detected. The presence of Si in the form of silicon dioxide, SiO_2, indicates its contamination with sand. The characteristic lamellate structure of the gypsum crystals is obvious in all samples (Figure 5e).



a



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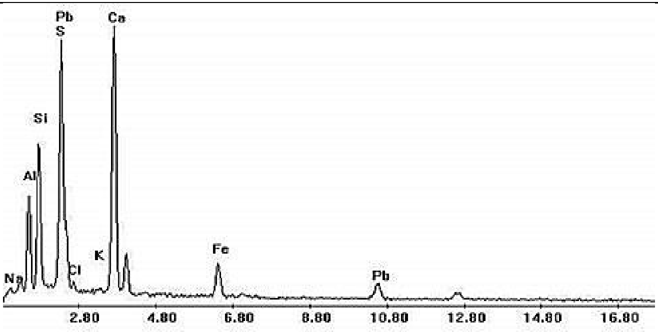

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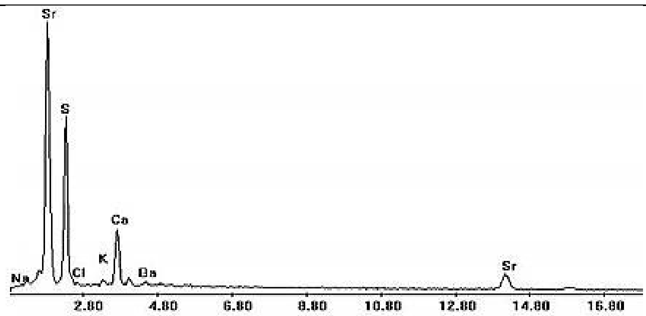

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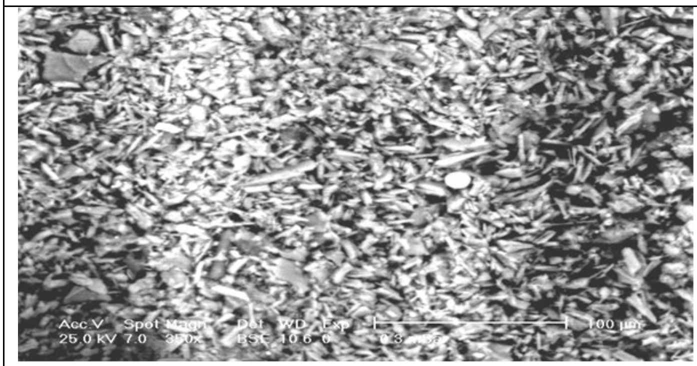

e

Figure 5 X-ray energy dispersion analysis by ESEM-EDX. a, Sample 1 (Gamali Yusef Mosque). b, Sample 2 (Dakhla in Tatar El-Higaziiya Mosque). c, Sample 3 (Bayt el-Kredlea museum, glass window the Byzantine room). d; Sample 4 (stained glass window in Zeinab Khatoon's house). e, the lamellate structure of the gypsum crystals of stained-glass windows in Zeinab Khatoon's house.

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MOISTURE EFFECT ON FUNGAL COLONIZATION

In a semi-simulation experiment, tested fungi failed to grow on the dry stucco mockup surface for an incubation period of 3months. After wetting the mockup surface, fungi grew, and the color deformation was evident in all samples. There is an electronic proximity between the polar water molecules and negatively charged stucco (Sharaf, 2002), enabling exposed stuccos to catch water even from moist air or from precipitation. Moreover, the growth of fungi like *A. niger* was evident as it formed a dense black layer that covered the entire surface of the stucco mockup (Figure 6a). Similarly, the fungal consortium (*A. flavipes, A. niger, A. flavus, A. fumigutas, A. ustus, P. canescens, P. chrysogenum*) of sample 1 (Gamali Yussef Mosque) densely covered the entire surface of the stucco model (Figure 6b). It was also observed that the growth of *A. flavus* in the recessed parts of the stucco mockup was more condensed than the rest of the surface mockup (Figure 6b). A similar result was noticed in the growth of the fungal consortium (*A. flavipes, A. niger, A. flavus, Alt. alternata, Penicillium* sp., *A. clavatus*) of sample 3 (Bayt El-Kredlea Museum) with a dark brown mat covering the entire mockup surface (Figure 6c). Additionally, a brown deformation characteristic to *A. flavus* growth covered a large part of the model surface and hid its original stucco color (Figure 6d). In figure 6 (e, f), an extensive fungal growth was apparent. The penetration of fungal hyphae among the lamellate crystals of the stucco material was obvious. Moreover, with a prolonged incubation time, the density of fungal growth exerted pressure on the stucco surface resulting in the decomposition of the surface layer and the falling away of some of its parts. In the same manner, all tested fungi produced a similar effect. Hence, enough moisture creates favorable conditions for the germination of fungal spores to carry out biodegradation. Therefore, moisture is the crucial factor in a fungal establishment on stucco surfaces, where the stucco material, as an inorganic substance, cannot support fungal growth alone.

In the monument environment, the source of moisture and nutrients was due to the poor sanitation network in the area and the use of toilets attached to the walls of mosques. Water leaks in soil transferred to the walls of archaeological buildings and their decorative elements, as indicated in Figure 7. In this respect, it is stated that sewage leakage in sub-surface water in Cairo is considered a significant factor in the damage to many archeological foundations (Awad, 2003). Daily entrance of visitors without supervision is viewed as another source of moisture (each person can produce 50 ml of moisture per hour), CO_2 and temperature. Nevertheless, anthropological factors cannot be ignored where studied monuments are in densely.

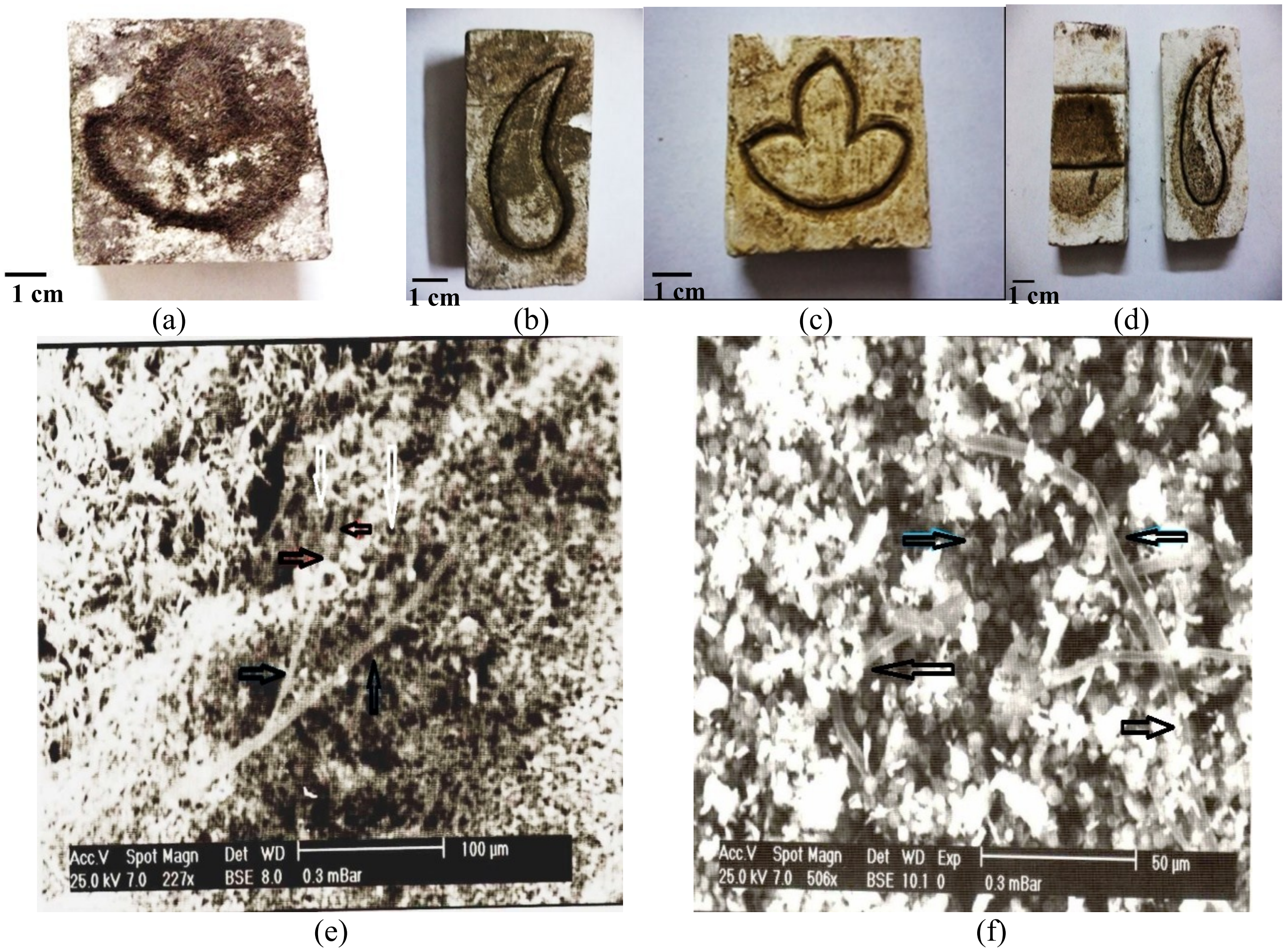

Figure 6 Semi-Simulation experiment (squares mockup 5 x 5 cm, 2.5 x 2.5 cm, rectangles 2.5 x 5 cm): (a) A. niger growing densely on the surface of a stucco mockup, (b); the dense growth of fungal consortium (from the mihrab wall decorations in the Gamali Yusef Mosque) on the surface of the stucco, (c); growth of A. flauvs on the plaster mockup surface and (d); condensed growth of fungal consortium in the hollow parts of plaster mockup representing stained-glass windows of the Bayt El-Kredlea museum. ESEM micrographs showing the hyphae of A. flauvs (e) and A niger (f) pass between the surface granules (arrowed).

Populated areas such as the Al-Gamaleya area. Some monuments, like mosques, are open to citizens without supervision and thus are exposed to many damage factors. For example, some vendors use parts of the monuments to display their goods or even throw garbage bags next to the monuments. Some residents spray water near the mosque walls, especially in the summer. In addition, some citizens attempt to restore parts of the monuments without referring to responsible authorities, which in the future leads to the destruction of the monument. A case in point is the use of cement in the restoration of walls. Other damage factors include the large scale dealing with the monuments by hand or writing and drawing on them. Thus, it is difficult to control the environmental conditions of open-air museums (Luo et al., 2015), which make the exhibits in constant contact with the external environment and the daily changes in temperature through the opening and closing of windows and doors which are the most important causes of fungal infections (Sterflinger, 2010).

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Figure 7 Tatar El Higaziya Mosque: a; The inner southern wall of the courtyard of the Mosque, b; The upper right part of the mosque's mihrab wall, showing High humidity in the inner walls of the mosque and the deep cracks salt efflorescence and wrong restoration by native people (arrows).

An additional reason for deterioration appears in figure 7 due to the water passage through the porous materials generating salt precipitation, crystallization and efflorescence (seen in a salty white layer over the moist region in many sample areas) and the production of a considerable pressure after hydration and that occupies larger spaces that lead finally to cracking, detachment and material loss (Adamiak et al., 2016). Moreover, wet-to-dry cycles, frequent heating and cooling stimulate chemical reactions that will eventually weaken the metal structures and lead to cracking, loss of materials, reducing the mechanical capacity and deterioration over time (Griffin, Indicator & Koestler 1991; Reyes et al., 2010; Emaraa & Korany, 2015). These porous surfaces are usually suitable passages for inhabitant hyphae (Verrecchia, Dumont & Rolko 1990). Efficient strategies can be implemented to reduce the deterioration of stucco carvings by increasing the archaeological awareness of the individual who deals with the monuments directly or during his presence in the surrounding environment. In addition, the carvings with high porosity stucco should be protected from contact with water regularly by any means. Thus, controlling environmental conditions, continuous cleaning and microbiological monitoring are essential for preventing contamination (Sterflinger & Piñar 2013). Since pollutants cannot be monitored in the air, maintaining museums and mosques regularly will reduce the risk of fungal infection.

BIODETERIORATION MECHANISM

Fungi are the main reason for the biodeterioration of archeological materials, as illustrated in Figure 8. Fungal damages appear as changes in the color of the stucco to gray, yellow or light brown as a result of its complications with the surface of the gypsum granules, dust and pollutants, which hide the fungal damage beneath them in many places of prominent and deep decoration. Since the fungal metabolites are colored, the injured part of the patina is transformed from white to other darker colors, as seen after removing a fungal layer from stucco mockups patina surface and as previously reported (Scheerer, Ortega-Morales & Gaylarde 2009; Sterflinger & Piñar 2013). The gray color is owing to using inappropriate mounting material for restoration by native people. Moreover, the growing fungi produce a plethora of secondary metabolites such as organic acids (oxalates, citric and malate) that cause metal plate breakdown in the growth environment and produce mechanical forces that facilitate the displacement of the matrix grains and lead to the disintegration of the substrate (Burford, Fomina & Gadd, 2003; Hoffland et al., 2004; Jain et al., 2009; Saba, Qui˜nes-Bola˜s & Batista 2019). Fungal organic acids bind to hydrogen ions and cause mineral hydrolysis and the formation of new compounds (Gadd, et al., 2002; Hoffland et al., 2004; Kamal et al., 2007; Mostafa et al., 2018; Saba, Qui˜nes-Bola˜s & Batista 2019). Furthermore, evolved CO2 (human respiration) can react with this strong acid and moisture, generating carbonic acid in the substrates that expectantly produces a dissolution pattern on the mineral structure of ancient stucco. Moisture with the physical and chemical components of archeological stucco activate specific chemical reactions which facilitate biodeterioration processes. Moreover, fungi can, to some extent, extract their metal requirements from stucco, where they were reported to effectively dissolve gypsum in the laboratory (Saba, Qui˜nes-Bola˜s & Batista, 2019), and they produce a specific "tunnel" in feldspar granules in retherm Podzol (Hoffland et al., 2004), that elucidate the penetration of the hyphae of A. flavus and A. niger among surface gypsum granules of synthetic mockups (Figure 5d).

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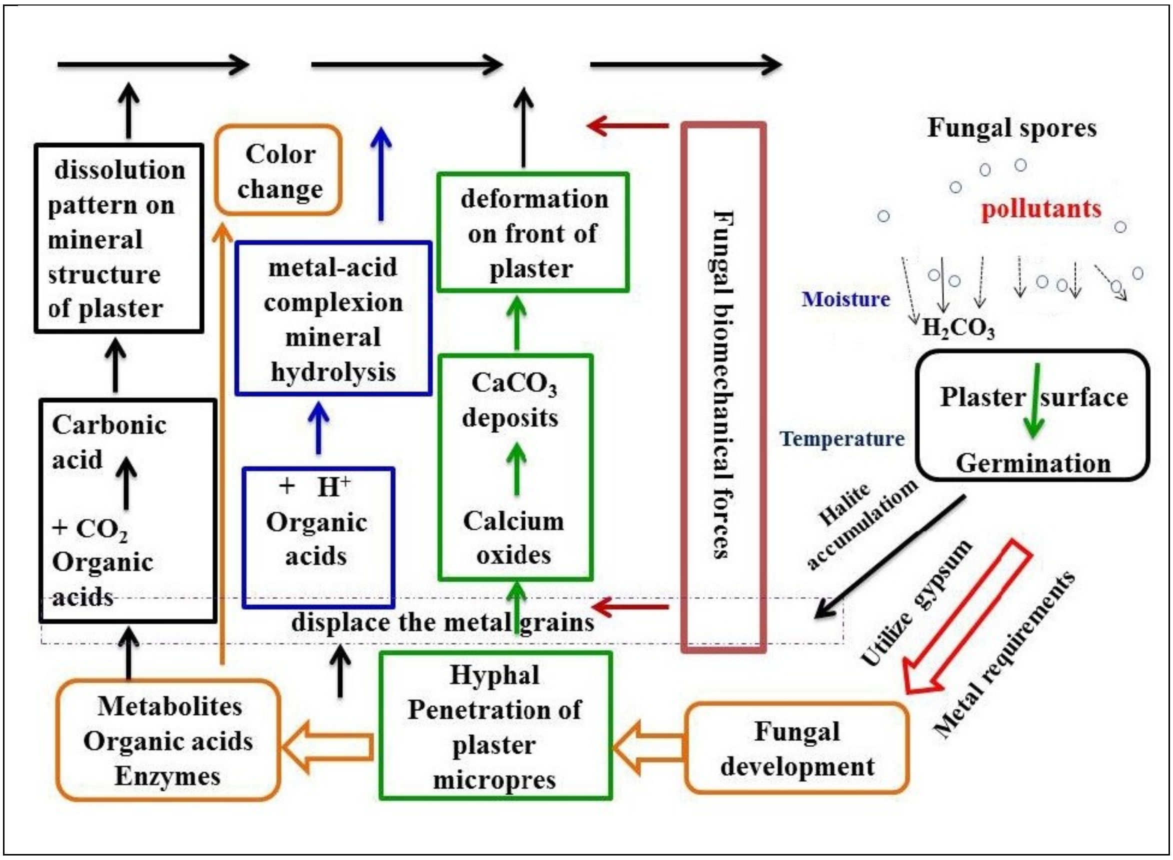

Figure 8 Proposed biodeterioration mechanisms of stucco carvings.

CONCLUSIONS

The structure and components of some historical stucco carvings are analyzed. The characteristic lamellate form of the gypsum crystals is evident in all samples. Calcium and sulphur elements, as two of the essential elements in the gypsum complex, were pronounced. Other impurities (copper, lead, Aluminum, Sodium, Chloride, Silicon, iron, Strontium & Barium) were frequently detected. The hemi-hydrated phase of gypsum (bassanite) was recorded in some samples. The high percentage of sand and halite in some samples indicates that they may have been used in the original stucco mixture. Halite is directly or indirectly involved in ancient stucco decay. Fungi are frequently isolated from the studied samples. The semi-simulation experiment confirmed that moisture content is the most potent factor for a fungal establishment observed by SEM micrograph penetrating the plaster mockup and wandering among its grains. Fungi cause cracks, holes and voids of stucco mockups structure and exert a mechanical pressure that may dislocate the matrix grains and lead to the fragmentation of the substrate in addition to changes in their aesthetic view. Thus, archeological mosques and houses are considered giant incubators providing a suitable and attractive environment for the growth of fungi, with the help of humidity combined with other factors. In the future, various materials will be applied to isolate the surface of plaster carvings from multiple damaging elements.

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