International Biodeterioration & Biodegradation 143 (2019) 104721

Contents lists available at ScienceDirect

International Biodeterioration & Biodegradation

journal homepage: www.elsevier.com/locate/ibiod

Controlling biodeterioration of cultural heritage objects with biocides: A review

Mian Adnan Kakakhel[a,1], Fasi Wu[a,b,1], Ji-Dong Gu[c], Huyuan Feng[a], Khadim Shah[d], Wanfu Wang[a,b,*]

[a] MOE Key Laboratory of Cell Activities and Stress Adaptations, School of Life Sciences, Lanzhou University, Lanzhou, 730000, PR China
[b] National Research Center for Conservation of Ancient Wall Paintings and Earthen Sites, Department of Conservation Research, Dunhuang Academy, Dunhuang, Gansu, 736200, PR China
[c] Laboratory of Environmental Microbiology and Toxicology, School of Biological Sciences, The University of Hong Kong, Pokfulam Road, Hong Kong SAR, PR China
[d] Institutes and Key Laboratories, Chemistry Department, Tsinghua University, Beijing, 100084, PR China

ARTICLE INFO

Keywords:
Cultural heritage
Biodeterioration
Biocide
Nanoparticles
Essential oils

ABSTRACT

Biodeterioration is when living organisms chemically or physically change or alter the appearance of materials objects. Organisms can colonize and destroy valuable cultural heritage. New advances in biotechnology and applied microbiology provide important information on conserving cultural heritage. Various physical and mechanical methods have previously been used, but they are incapable of preventing the growth of organisms entirely. Organic biocides, particularly commercial formulations, do not last long because they can be utilized as a nutrient source by indigenous microflora after these microflora are exposed to biocides and develop resistance. Therefore, inorganic nanoparticles have a better chance to protect cultural heritage. Silver (Ag_2O) and titanium (TiO_2) oxides are effective against biofilm, and nanoparticles of zinc oxide (ZnO) are effective antimicrobial agents. This new generation of biocides is much smaller in size and extremely active to damage DNA or RNA. In addition, green biocides from natural sources offer an alternative to chemical ones, having low toxicity compared to chemically synthesized biocides. Future research on biofilm control technologies may contribute to a broader understanding of and new perspectives on a future generation of biocontrol agents and methods with the potential for sustainable development.

1. Introduction

Throughout world history, humans have been erecting monumental complexes and buildings for centuries using durable stones. The Mediterranean region was a main marble-producing area. Romans in the first century B.C.E founded the Baia as an important cultural site. Until the third century, it was an important destination for the imperial family and Roman aristocracy. Marbles was used extensively for decoration and for sculptures in antiquity (Ricca et al., 2015). Belfiiore et al. (2016) conducted a study over provinces of statuary marbles from the Roman archaeological site of Villa dei Quintili and found that architectural elements, sculptures and structures were made of marbles during the Roman Empire. Tykot et al. (2002) suggested that the “dolomite test” can be used to determine whether or not a marble sculpture piece is from Cape Vathy, Thasos in Mediterranean region. At the start of the sixth century, marble began to be extracted from different areas around the world to serve as building material and became common in a very short period of time. Marble and limestone inspired ancient Greek and Roman architects to construct the Acropolis and the Colosseum, respectively. In a similar way, Khmer civilization mainly used sandstone to build temples and monuments extensively in Southeast Asia (Liu et al., 2018). Nowadays, these stone temples and monuments are completely exposed to the ambient conditions of tropical climates and show visible colonization and deterioration by flora and microbial communities. The activities of microbial communities on cultural heritage works are significant causes of damage through chemical or mechanical processes (Ciferri et al., 2002; Warscheid and Braams, 2000). This kind of damage by biota of flora and microbiota is known as “biodeterioration,” which refers to any biological activity that affects the appearance and integrity of the materials (Allsopp et al., 2011). The term “biodeterioration” can also be defined as the unwanted alteration of various materials, mainly caused by small living organisms

---

[*] Corresponding author. MOE Key Laboratory of Cell Activities and Stress Adaptations, School of Life Sciences, Lanzhou University, Lanzhou, 730000, PR China.
E-mail address: wwanfu@hotmail.com (W. Wang).

[1] Co-first author, contributed equally to this work.

https://doi.org/10.1016/j.ibiod.2019.104721
Received 26 March 2019; Received in revised form 17 June 2019; Accepted 18 June 2019

Fig. 1. Biodeterioration of the world heritage sites. **A:** Biological deterioration of Leshan Giant Buddha at Sichuan Province, China. **B:** Microbial brown spots on wall paintings in Mogao Caves of Dunhuang, China; **C:** Biodeterioration of sandstone sculptures of North Grottoes in Gansu Province, China; **D:** Biodeterioration of the Khmer Smile at Bayon Temple, Angkor Thom, Cambodia. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

(microorganisms) like bacteria, archaea, fungi, and lichen (Allsopp et al., 2011). "Cultural heritage" refers to archaeological monuments, sites and other historic areas. The presence and activity of archaea, bacteria, algae, lichen, fungi, sometimes animals and higher plants often cause biodeterioration of stone relics and ancient wall paintings and are thus major threats to the long-term preservation of this valuable cultural heritage (Fig. 1).

Fungi play both positive and negative roles in our daily life. They are pathogens that synthesize mycotoxins and most importantly, have the potential to cause biodeterioration for both inorganic stone and concrete (Mitchell and Gu, 2000; Sabev et al., 2006; Meng et al., 2017), and also high-strength engineering polymeric materials (Gu, 2003; Gu et al., 1996, 1998; Koestler, 2000). The damage of cultural heritage objects is a complicated process, caused mainly by the chemical and physical processes set in a motion by activities of the organisms involved. Microbial growth on the surface of materials may cause the cracking, powdering and displacement of building materials, which results in the weakening and eventual complete destruction of monuments (Farooq et al., 2015; Liu et al., 2018). Biodeterioration has been discussed widely, but its negative impacts have been underestimated mostly due to few investigations on the mechanisms involved in the past. It was once believed that chemical processes are the main factors responsible for decay, but now, perceptions have changed, and it has become generally accepted that bacteria, archaea, and fungi cause serious destruction to cultural heritage such as mummies, books, and paintings through their enzymatic activity, metabolic processes, and corrosion/damage (Sterflinger et al., 2013). Under the moist conditions of tropical climate, the water/moisture contents are high, promoting a successful growth of biofilm and plants in stone monuments and buildings (Liu et al., 2018). Biodeterioration not only affects cultural heritage and books but also occurs underwater. In past decades, study on degradation underwater have increased (Ruffolo et al., 2017a,b). A study conducted by Bruno et al. (2016) presented a project named CoMAS. In this project, new materials have been developed to prevent biological colonization to better preserve cultural remains underwater. A study carried out by Ruffolo et al. (2017a,b) about the conservation of underwater archaeological stone materials at the Baia park (Naples, Italy) showed a great decrease in biological colonization on the surfaces of specimens that had been treated with synthesized and tested formulations.

Among the biological organisms responsible for biodeterioration, fungi play a key role in the biodeterioration of organic materials (Gu et al., 1996, 2003) and monuments (Meng et al., 2016, 2017) given their complex metabolic activities, which includes the production of organic and inorganic acids (Farooq et al., 2015). It is suggested that *Fusarium* sp. are capable of producing organic acid to attack concrete (Gu et al., 1998). As cultural heritage is very important for preserving our past and giving us insight into our past and our future, cultural heritage preservation is thus essential to retaining historical, artistic, and scientific value. Biotechnology and applied microbiology are valuable tools in the conservation of cultural heritage. In most cases of biodeterioration, biocides have been used to eliminate and inhibit the growth of microorganisms. Nevertheless, these biocides may cause damage to the quality of heritage artifacts.

As we know that in many cases biodeterioration is mostly caused by microorganisms, we normally use biocides to kill or destroy these microorganisms. Biocides are largely used in healthcare way. Antiseptic use of Quaterniry ammonium compound, phenol, aldehydes and alcohol are a clear landmark of biocide as disinfection (Maillard et al., 2005). For the conservation of artworks, the biocidal means through which different microorganisms are destroyed are known as "biocides". Biocides are used to stop and reduce the spreading of microorganisms (Griffin et al., 1991). Some biocides are inorganic and, due to their saline nature, are soluble in water, while others are organic and less soluble or insoluble in water (Unger et al., 2012). Biocides can be divided into four different groups: 1) pesticides, 2) disinfectants, 3) preservatives, and 4) other biocidal products (Ashraf et al., 2014). The Angkorwat site in Cambodia is an important authentic site that records the history of the Khmer civilization. It became a UNESCO World Cultural Heritage site in 1992, after which different countries became involved with the protection and conservation of the site. At this site, a large number of microbes can be observed on the sandstone's surface over time. These microbes are responsible for the color development on the surface, and the activity of some selective groups such as sulfur-oxidizing bacteria, AOB and AOA, that are responsible for biodeterioration of the sandstone integrity through mineral dissolution and acid production (Bartoli et al., 2014; Gu and Katayama, 2017; Meng et al., 2017; Kusumi et al., 2014; Liu et al., 2018; Zhang et al., 2018).

This review is intended to summarize the advancement of cultural heritage biodeterioration and protection countermeasures by focusing on the application of antimicrobial technologies such as green nanoparticles, plants essential oils, organic and inorganic biocides that appears to be the most convenient and effective way for the conservation of cultural heritage. Some biocides failed either because of the environmental hazards and toxicity or microbial communities utilizing biocides as carbon or nitrogen sources. In the future, more attention should be given to modulation of the ambient environmental conditions and the materials, including several factors such as nanosized, synthesis method, pressure time, temperature, pH, and environmental humidity. The quality and quantity of nanoparticles is influenced by proximity. Therefore, minimization of the colonization and initial growth of microorganisms provides more effective protection than application of additional external chemicals.

2. Living microflora involved in biodeterioration

The presence of microorganisms that deteriorate monuments and stone artwork has received a great attention in recent years. *Aphanocapsa* sp., *Aphanotheca saxicola* and *Calothrix marchica* have been identified in the deterioration of marble and limestone (Madeco et al., 2009). These photoautotrophic organisms grow easily and produce colored pigments on heritage objects (Macedo et al., 2009). One recent review specifically pointed out that water is a critical factor in evaluating and assessing microbial colonization and destruction of Angkor sandstone architectures (Liu et al., 2018). The establishment of a microbial population on stone heritage objects is initiated by phototrophic or autotrophic cyanobacteria, algae and lichens followed by heterotrophic bacteria and fungi (Hu et al., 2013). Polymeric materials (e.g., polysaccharides and proteins) are secreted by these microorganisms to create a prominent and complex layer of biofilm (Schaefer et al., 2001). Biofilm is formed by different microorganisms on the external surface, which leads to trapping of water inside the sandstone. When water cannot move in and out of the sandstone freely, emergence of salting effects, powdering, and delamination will occur and destroy the underlying sandstone (Liu et al., 2018). Biofilms that lead to biodeterioration form on external surfaces as well as inside stone or rock. According to de los Rios et al. (2004), microorganisms can occupy cavities or cracks in stone, classified as cryptoendolithic and chasmoendolithic respectively. This colonization leads to cracking, which penetrates the rock. Several types of epilithic and endolithic microorganisms are usually observed on stone monuments.

2.1. Deterioration caused by bacteria and cyanobacteria

Microorganisms are present on exposed surfaces of murals, stones and sculptures, and then cause color change or attack the underlying materials chemically through metabolites (Abdel-Haliem et al., 2013). Cyanobacteria and lichen are phototrophic microorganisms and are considered to be the main colonizers of stone monuments under both humid and semi-humid conditions (Crispim et al., 2005). Phototrophic bacteria derive their energy from sunlight, while autotrophs derive energy from redox reactions, while hydrogen sulfide (H_2S) is oxidized by sulfur-oxidizing bacteria to form sulfuric acid (H_2SO_4) (Lepidi et al., 1973). The nitrification process is carried out by ammonia-oxidizing bacteria as well as by ammonia-oxidizing archaea (Cao et al., 2013; Meng et al., 2016). In this process, the ammonia is oxidized to nitrate by using the enzyme ammonia monooxygenase under aerobic conditions (Meng et al., 2016, 2017). Chemolithoautotrophic bacteria, sometimes present on stone surfaces, release different types of acids such as nitric acid, sulfuric acid, and nitrous acid, which change the local pH and cause deterioration (Crispim et al., 2005). Vijayakumar et al. (2014) pointed out that many taxa of cyanobacteria were found on artworks and monuments in temperate and tropical regions. The most prevalent was *Lyngbya,* followed by *Oscillatoria*, *Phormidium*, *Chroococcus*, *Microcytis*, *Aphanocapsa*, *Gloeocapsa,* and *Plectonema*. Proteobacteria, Actinobacteria, Acidobacteria, and Chloroflexi were the most dominant groups, and the process of nitrification was faster and more active at Beng Mealea, which might contribute to severe biodeterioration on sandstone at the Royal Palace in Angkor Thom and Beng Mealea (Zhang et al., 2018).

2.2. Biodeterioration caused by fungi

Fungi are heterotrophic microorganisms and thus require organic materials for their growth. Various laboratory tests have shown that fungi oxidize manganese (Petersen et al., 1988). Fungi cause decay of cultural monuments due to their pigmentation and patterns of colonization (Sterflinger et al., 2013). Farooq et al. (2015) carried out a study in Pakistan on the microbial deterioration of the stone memorials of Dharmarajiika to reveal that 19 species of fungi belonging to 13 genera were present on the monument's surface. The prevalent species of fungi were *Penicillium frequentans*, *Rhizopus oryzae*, and *Aspergillus fumigatus*. It was noted that these fungal species produced different organic acids such as gluconic acid, oxalic acid, succinic acid, and acetic acid. Spores of fungi are always present in the heritage-attached environment. Wang et al. (2011, 2012) analyzed airborne fungi in the Mogao Grottoes of Dunhuang, China and found that *Cladosporium* spp. were the most prevalent species, closely followed by *Aspergillus*, *Penicillium* and *Fusarium*. A more recent study by the Dunhuang Academy showed that the fungi affiliated to the order of Sordariales and the family of Trichocomaceae were dominant in mural samples, mostly due to the poor museum preservation conditions and conservation intervention afterwards (Duan et al., 2018). Nevertheless, the fungus *Aspergillus allahabadii* isolated from surfaces of Angkor Thom stone may have a basic role in eliminating of microbial biofilms, as different kinds of enzymes can be produced by these microbial species to exploit many kinds of organic substrate. This is a potential key step toward biological control of biodeterioration without using toxic chemicals (Hu et al., 2013).

2.3. Biodeterioration caused by Lichen

Lichens are biodeteriogens responsible for the damage of outdoor sculptures and sites over time. The mycobiont, which comes in direct contact with the substrate, is the key in biodeterioration caused by lichens. For the most part, the lichen family existing on cultural properties does not differ greatly from those species that exist on nearby rocks, and due to the limited samples taken from monuments, many studies of lichen-stone collaborations were therefore performed on samples from nearby rocks.

Epilithic lichens develop thalli on stone surfaces and these lichens penetrate their hyphae variably into the substratum. The ability of lichens to alter the substrate is connected both to the physio-chemical features and physiological differences (Salvadori and Municchia, 2016). Lichen-caused biodeterioration mainly occurs by way of chemical (organic acid, lichen substances and interaction of CO_2) and physical processes (hyphal penetration and pressure exerted by thalli) (Salvadori and Municchia, 2016). Lichens are responsible for both the abiotic weathering mechanism and also the protection of the rock (Naylor et al., 2002; Carter and Viles, 2003). When colonization is initially begun by lichens, the movement of inorganic carbon starts from the atmosphere and becomes attached to sandstone. Along with this, the phosphorous, nitrogen, and sulfur also combined with the biomass community. However, after the decomposition of biomass P, N and S were released from organic form to inorganic acid (Liu et al., 2018).

Sohrabi et al. (2017) detected 28 different lichen species on the Pasargadae monument; from these, both endolitithic and eplithic lichens contributed to the biodeterioration of stone and heritage sites. Municchia et al. (2018) evaluated biodeterioration activity of the lichens in the Cave Church of Üzümlü (Cappadocia, Turkey), and found a dramatic loss of the stone matrix and a dense network of fungal hyphae within the rock. Thus, proper assessment and biocidal and physical optimization must be carried out to treat lichens on heritage objects. Despite the damaging effects of lichens, certain species of lichens may actually help protect stone objects. A study was conducted by Concha-Lozano et al. (2012) on the protective effects of limestone buildings. In this study, patinas were analyzed using XRD and SEM-EDX, including both the weathered and unweathered sides of gypsum contents. An old query of weathering forms indicated that colonies of lichen (*Verrucaria nigrescens* and *Caloplaca auranita*) have filled the pores of limestone with a dense network of lichenized fungal hyphae. This study showed that a stone can be made waterproof by these endolithic organic matters and can act as a barrier for sulfate contamination. This suggests that some species of lichens may contribute to the protection of some cultural monuments.

2.4. Biodeterioration caused by archaea

For the study of bio-erosion of cultural monuments, it is necessary to examine the community of microorganisms and their activity (Zhang et al., 2018). The archaea cause damage that is more severe than expected (Scheerer et al., 2009). The archaea related to the nitrogen cycle can produce inorganic acid, which leads to damage of cultural monuments (Meng et al., 2017). A species of the archaea *Halobacillus trueperi* has been known to take part in carbonate mineralization (Rivadeneyra et al., 2004). The nitrification process is carried out by ammonia-oxidizing bacteria and ammonia-oxidizing archaea, but archaea were more abundant than bacteria at the Temple of Angkor (Meng et al., 2016). Another study used the Ion Torrent next-generation sequencing platform in Poland to detect three archaea phyla present on historic building materials that may be involved in the biodeterioration of materials (Adamiak et al., 2018). It can be concluded that archaea are more ubiquitous in causing biodeterioration as compared to other microorganisms, but the information on this group of microorganisms is limited due to previous negligence of the study of them.

3. Countering biodeterioration for heritage conservation

Conserving culturally relevant buildings and historic sites provides opportunities for future generations to honor their legacy and achievements (Steinbauer et al., 2013). However, improper preservation practices and complexes condition of artifacts present a large issue for preserving cultural heritage. Currently, science and technology often relate to the arts in many ways; the analytical approaches of biotechnology undoubtedly play important roles in conserving historical heritage (Fernandes et al., 2006). However, in the process of heritage conservation and biodeterioration management, only products and methods deemed suitable by available scientific knowledge may be used. The control methods for biodeterioration are classified as 1) physical, 2) mechanical and 3) biochemical. Biochemical methods, either broad spectrum or narrow spectrum, are used more commonly than physical or mechanical methods for conservation of heritage (Allsopp et al., 2011).

3.1. Mechanical methods

The use of mechanical methods for microflora removal and cultural heritage protection has been described and tested in various research projects. The “mechanical method” for treating biodeterioration refers to techniques most commonly used for the removal or displacing of biodeteriogens; this may be done manually or by using other physical tools such as vacuum cleaners, scalpels, and spatulas. However, such methods used in the past have failed because they are unable to fully inhibit microbial growth. The substrates where the microflora exists can indeed be damaged by these mechanical tools. It is difficult to remove the lichens without a strong peeling effect, so physical removal is not advised (Municchia et al., 2018). The use of laser techniques, which shows the improvement of mechanical methods, which leads to weakening of the lichen. Although the combined use of mechanical tools and laser tools provides satisfactory results, it is still unable to remove the thalli deep inside the stone (Rivas et al., 2018). It has been suggested that lasers are only able to remove or eliminate the cortexes of these thalli.

3.2. Physical methods

Many physical methods such as low frequency electrical systems, heat, and Ultra-violet (UV) radiation have been applied to conserve heritage sites (Scheerer et al., 2009). UV radiation has been used to counter the growth of fungi, algae and bacteria, while gamma radiation has been used to sterilize microflora. However, the effects of using gamma radiation are not long-lasting, and UV radiation has proved dangerous for the user. Pfendler et al. (2018) compared the effects of treating biofilm with biocides and UV-C. The use of biocides resulted in an incomplete removal of biofilm, while the UV-C method combined with necrotic organic matter can be harmless for the environment and may also help to remove the biofilm.

Gamma radiation, on the other hand, induces a chemical reaction in cellulose molecules, which leads to the breakdown of molecule linkage (Adamo et al., 2001). A higher frequency can be used to combat wood-destroying insects. Heat can be used as a disinfectant (Gaylarde et al., 2011). Another laser technique has been in development over the last couple of years. Its diagnostics seem promising, but this technique is at present still unable to completely remove microorganisms from artwork (Salimbeni, 2006). Er:YAG is a laser that is promising tool to conserve the cultural monuments. Pereira-Pardo et al. (2017) performed a study wherein the Er:YAG laser was used to remove lime wash, but failed to remove all of the lime wash. Ethanol with water was applied for a few minutes, but still the laser was not completely successful. This laser technique can be considered successful in removing biofilm from monuments because traditional chemical and mechanical cleaning is inefficient.

Salimbeni (2006) mentioned that the marble statue was cleaned with the Q-switch ND: YAG laser combined with third harmonic UV. Another study conducted by Sanz et al. (2017) in Spain investigated rock samples on which lichens such as *Candelariella vitellina, Aspicilia viridescens, Rhizocarpon disporum* and *Protoparmeliopsis muralis* were present. A laser study was then carried out, which showed that the lichen species were not completely removed by the laser application. The upper cortex was only partially removed, indicating that laser techniques cannot completely remove lichen colonies, but can only somewhat damage the colony. Rivas et al. (2018) showed that using the laser alone not only hardly removes the lichens but also can damage the mineral, mostly biotite. The process of cleaning cultural monuments is always a tough challenge. Many techniques have been used; some are promising but have side effects. Atmospheric plasma torches have been used by Voltolina et al. (2016) and tested on cleaning cultural stones. Acrylic polymers, epoxy and hydrophobic siloxanes were chosen to test the efficiency, but a siloxane-based coating cannot be fully removed by this technique.

3.3. Biocides treatment

Different compounds like aldehydes, phenol, amides, acids, and alcohol are used as biocides. Among these, however, only a very limited number of biocides are long-lasting. Most of the biocides are ultimately ineffective at removing microbes and may eventually lead to a new wave of microbes on the affected surfaces after the microbes develop a resistance. Furthermore, the details of how biocides instigate antimicrobial activity are not fully understood (Singer et al., 2010), although McDonnel et al. (1999) provides a brief discussion of the mechanisms involved. According to this study, some biocides target the cell envelope and protein crosslinking. Quaternary ammonium compounds target the cytoplasmic membrane and damage the phospholipid bilayer. Some silver compounds target DNA and interrupt enzymes. Effective biocides have been shown to have the ability to self-sufficiently enter microbial target cells, thus suppressing or killing the living microbes internally. Some chemicals are used as permeation-promoters, which make the cell membrane porous, allowing biocides to enter the target cell more easily (Young et al., 2008). Biocides may be organic or inorganic. Inorganic biocides are longer-lasting while organic biocides deteriorate quickly. Therefore, many studies champion the use of inorganic nanoparticles (La Russa et al., 2014).

One way on which biocides operate is by suspending the metabolic action of microbes, thus altering the proper functioning of microbial cells and causing cell death. Biocides are divided into two kinds with regards to their mode of action, which cause either a non-oxidizing or an oxidizing effect. The most commonly used oxidizing biocides are

Table 1
Some characteristics of the major biocides.

Biocide	Results	Efficiency	Drawbacks	Durability	Reference
Algophase	3%	Stable	Harmful with organic solution	Very Durable	Urzì et al. (2007)
Anatase	0% Growth after application	High Stable	Less effective in Long UV Rays	Best Durable	Fonseca et al., 2010
Glifene SL	Stop growth	Stable	Acute Eco-Toxic		Favero-Longo et al., 2017

hydrogen peroxide, bromine, chlorine, and ozone. Despite their effectiveness, oxidizing biocides have some negative effects as well. The most used non-oxidizing biocides are quaternary ammonium compound, acrolein, and aldehyde di-amines and amines (Martin-Sanchez et al., 2012; Ashraf et al., 2014). When comparing oxidizing and non-oxidizing biocides, a better result may be observed with oxidizing biocide simply because the non-oxidizing biocide cannot penetrate the lipid membrane of a bacterial cell. When it comes to the advantage and limitations of the applied biocides, Anatase was considered highly durable and controlled the growth of biodeteriogens in an effective manner; Glifen SL is good for controlling the biological growth but toxic to the environment (Table 1).

4. Major biocides and their applications

4.1. Application history

Stone artwork and archeological monuments have been studied by researchers from around the world in order to inhibit their biodeterioration. Ranalli et al. (2003) tried to preserve stone monuments by using a bioluminescent low-light imaging technique, while Urzì et al. (2007) used various biocides in combination with water repellant compounds like rhodorsil RC80, hydrophase superficial, and hydrophase malte to achieve results showing complete elimination of microbial growth. Martin-Sanchez et al. (2012) applied biocides (Devor Mousse and Parmetol DF12) to the Lascaux Cave subterranean environments in France, resulting in a succession of microbial communities over time. The different kinds of biocides and nanoparticles that have been used in conservation of cultural heritage sites and artworks are shown in Table 2.

Undoubtedly, various biocides have been used for microbial control for many years. Some of them are organic while others are inorganic. It is worth mentioning that Silva et al. (2015) conducted a study on secondary metabolites of bacterial species. The metabolite extract was applied on different fungal species, showing great antifungal activity. This is a natural solution to stop the growth of fungal species.

4.2. Case studies

Some authors have discussed the treatment of cultural heritage and stone artwork by living microflora from a historical perspective (Griffin et al., 1991). The Lascaux Cave was discovered in 1940 and subsequently opened for visitors. In the early 1960s, a green biofilm caused by algae formed in the caves, causing a series of biocides to be used at this site. In 2001, a fungal growth in the caves was identified as *Fusarium solani.* Biocidal treatment was subsequently undertaken, but after the treatments, new black stains appeared, worsening in 2007. It was concluded that melanized fungi were mostly responsible for the biofilm formation (Bastian et al., 2009).

Another study was further conducted by Martin-Sanchez et al. (2012) to identify the community of fungi, characterized by black stains, in the Lascaux Cave where an outbreak of *Fusarium solani* occurred. To check the antifungal effectiveness of biocide, two samples from the black stains were collected before and after the biocidal treatment respectively. The biocide used, which was named “Devor Mousse,” consisted mainly of chemicals such as quaternary ammonium, benzalkonium chloride, 2-octyl-2H-isothiazol-3-one, and Parmetol. After four months of biocide application, a new outbreak of black stains occurred, which progressively began to occupy the cave. Biocides were applied to control the outbreak, but mostly without success over time

Table 2
Major biocides that were widely used for inhibition of biological deterioration on cultural heritage.

Name of Biocide	Active Components	Some References
Algophase R	2,3,5,6-tetrachloro-4-methylsulfonyl-pyridine	Urzì et al. (2007)
Anatase	Titanium dioxide (TiO2)	Fonseca et al. (2010)
Biotine R	N-octyl-isothiazolinone (3–5%) þ 3-iodoprop-2-ynyl N-butylcarbamate (10–25%)	Ruffolo et al. (2017a)
Clove Extract	Eugenol	Jeong et al. (2018)
Colloidal silver	Nanosilver	Gutarowska et al. (2012)
DES®	2-phenylethoxy-quinazoline	Silva et al. (2016)
DesNovo R	Essential oil and benzalkonium chloride	Stuper et al. (2014)
Devor Mousse R	Quaternary ammonium and benzalkonium chloride	Martin-Sanchez et al. (2012)
Polymyxins R	Non-ribosomal synthesized secondary metabolites	Banat et al. (2014)
Glifene SL	N-(phosphonomethyl) glycine; 30–40%	Favero-Longo et al. (2017)
Kimistone Biocide	Didecyldimethylammonium chloride	Pinna et al. (2012)
Lichenicida 464	4,5-dichloro-2-octyl-4-isothiazolin-3-one þ 3-iodo-2-propynyl N butyl carbamate þ 2-octyl-4-isothiazolin-3-one þ benzyl alcohol	Favero-Longo et al. (2017)
Metatin N58-	Dimethyl benzyl ammonium bromide	Ranalli et al. (2003)
Mora Poultice	Ammonium/Sodium Bicarbonates	Mora et al. (1984)
Panacide	Dichlorophene	Silva et al. (2016)
Parmetol DF12	Isothiazoline Derivatives	Martin-Sanchez et al. (2012)
Polybor TR	Borates/Boric acid	Richardson et al. (1973)
Prenentol R	Ammonium quaternary compound	Paulus (2012)
Prepared nanoparticles	ZnO	El-Feky et al., 2014
Preventol R180	Alkyl dimethyl benzyl ammonium Chloride; 80%), isopropyl alcohol (2%)	Vannini et al. (2018)
Rocima™ 103	Alkyl-benzyl-dimethylammonium	Urzì et al. (2016)
Sanosil S003	Silver nitrate and hydrogen peroxide	Savković et al. (2016)
Secondary Metabolites	Lipopeptide	Silva et al. (2015)
Sinoctan PS	4,5-Dichloro-2-octylisothiazolinone, Iodopropynylbutyl	Favero-Longo et al. (2018)
Ucarcide R	di-aldehyde product	Rakotonirainy et al. (2007)

(Gu, 2003). Slight success was noted, but the application of biocide also caused some negative effects, especially ecotoxicologically. Before the treatment, *Ochroconis lascauxensis* was dominant. This dominance was confirmed by the clone library to be 97%. After the application of biocide, the *O. lascauxensis* was reduced to 52.3%, but the fungal community became more dominant, comprising 96.6% Ascomycota and 3.4% Basidiomycota. The samples collected in 2010 certainly differed from those collected in 2008 and 2009. *O. lascauxensis* sp.*, Aspergillus* sp., *Trichderma* sp., *Cladosporium* sp., *Alternaria* sp., *Rhodotorula* sp. and *Gymnascella* sp. were most abundant in the 2008-09 samples. The Herpotrichiellaceae family was most abundant in the samples collected in 2010. Now a question arose: if a biocide was used as an antifungal and antibacterial agent, then how had a new outbreak occurred in the form of new black stains? It was then discovered that the biocides applied had contained organic carbon and nitrogen in their commercial formulations (Gaylarde et al., 2011; Gu, 2003). The *A. nepalense, Ochroconis* spp., and Herpotichiellaceae families that were abundant in the cave had metabolized the organic additives present in the biocide. This may have been the main cause of the new black spots.

Conservators and scientists in Asian countries have gained an understanding of deterioration and management of cultural heritage more recently. The first international symposium on conservation of cultural heritage was held on 20–22 August 2014 in Dunhuang, China (Wu et al., 2017). The seasonal dynamics, diversity, and molecular description of bacteria and fungi transported by air in the caves of the Mogao Grottoes have been monitored in recent years (Wang et al., 2011, 2012). This site has been called “the art gallery of the world” and was named a UNESCO World Heritage Site in 1987. Ma et al. (2015) found out that most of the culturable strains separated from wall paintings showing indications of biodeterioration at this site were impervious to various stresses, making them potentially responsible for the damage to cave paintings; thereafter, more research on activity detection and methods to prevent the growth of these microorganisms are urgently needed in such similar heritage sites (Fig. 2).

5. Development of biocides and new requirements

5.1. Trends in biocide application

Scientists have used different forms of biocides in many forms to treat cultural heritage sites and stone monuments, with varying levels of efficacy. This section will discuss whether organic biocides or inorganic nanoparticles are more effective and environmentally friendly. Another question that will be discussed in this section is why organic biocides are being replaced by inorganic nanomaterials. Organic biocides are effective, but sometimes can be deactivated by other organic materials present in the soil. More recent research has focused on the efficiency, persistence, and environmental benefits of organic biocide products. Two commercial biocides, Biotine T (with the active principle 2-octyl-2H-isothiazole and Quaternary Ammonium Salts) and Preventol RI80 (didecyldimethylammonium chloride, 2-octyl-2H-isothiazole), were used against lichen thalli to check the physiological recovery of the thalli. A great physiological alteration was noted after the application of biocides. Preventol R180 was found to be more effective and created quick physiological alterations (Vannini et al., 2018). The commercial biocides Des 50 and Biotin T were used against non-pathogenic bacteria; both of these biocides are more effective against gram positive as compared to gram negative (Dresler et al., 2017). Jeong et al. (2017) found through laboratory testing and on-site application that the antifungal potential of the substance Eugenol extracted from natural medicinal clove was more effective. It showed great anti-fungal activity and caused a momentous decrease in microbial activity. Vegetable essential oil fungicides are promising and environmentally friendly.

Fig. 2. Microbial deterioration of wall paintings and its control. **A:** Whitish fungal outbreak investigation and samples collection at the World Cultural Heritage Site Maijishan Grottoes, Tianshui, China; **B** and **C:** Soft fiber papers infiltrated with screened biocides for controlling of brownish microbial spots causing deterioration on wall painting of Mogao Grottoes, Dunhuang, China.

However, nanoparticles (NPs) have received more attention for the restoration and conservation of cultural heritage sites and stone monuments. NPs have both catalytic and photocatalytic activity (Ortego-Morales et al., 2018). Pure and mixed TiO_2 (TiO_2 + AgNPs) were used by Ruffolo et al. (2017a,b) to prohibit microbial colonization on a stone surface in Italy. This study results revealed that the usage of the TiO_2-based nanocomposite may extend the efficacy of organic biocides for the treatment of microbial colonization, but reduction in efficacy of TiO_2-based nanoparticles has been observed when there is a higher concentration of water. At present, the synthesis of nanoparticles, which show great results when applied to different kinds of pathogens, needs improvement. It is therefore concluded that preference should be given to silver nanoparticles on environmental strains. This work should be continued in future due to the current lack of adequate experimentation. An advantage of these nanoparticles is that they are longer-lasting against bacteria. However, sometimes the nanoparticle can also be affected if the surface has more humidity and atmospheric deposition. A technique must be developed to monitor the potential risks of this behavior. The silver nanoparticles can be synthesized by chemical and physical processes, biological method for synthesis are also in stages of development (Ali et al., 2018).

For the restoration of heritage objects, coating containing nanoparticles has multidimensional features to guard stone objects against environmental degrading agents. During this situation, monitoring the concentration of nanoparticles is an important issue because this monitoring over time can give an understanding of its durability. A study was conducted by Macchia et al. (2017) upon a theoretical background to develop an instrumental method to meet this challenge to fulfill and monitor the durability of nanoparticles concentration. In this model, it is possible to determine and monitor the coating thickness and concentration of nanoparticles. A study considered by Sierra-Fernandez et al. (2018) about synthesis methods, the application of nanotechnology for the conservation of stone monuments, and characterization techniques for nanomaterials noted that it is very important to consider human health. TiO_2 NPs are widely used in different fields, but some aspects of their use in the conservation of cultural heritage are still unresolved. Crupi et al. (2018) conducted a study evaluating the effects of different amount of nanoparticles in the features of coating. Neutron radiography revealed that there was a significant decrease in penetration into the limestone as the TiO_2 increased within the coating.

How can nanotechnology applications help with cultural heritage conservation? In conclusion, nanotechnology can serve a role in protection of cultural heritage through applications such as cleaning painted glass, protecting pigments, and destroying airborne pollutants.

5.2. The environmental impact of biocides

According to scientific methods, every chemical or biological component has positive and negative effects depending on either biotic or abiotic factors. For example, disinfectants are antimicrobial but are harmful to humans. Likewise, antibiotics are used to kill or suppress bacteria but can also cause re-emergence through antibiotic resistance.

This is also the case with biocides, which are very effective in preventing monuments from microbial attacks for a short period of time in the beginning, but are expected to cause further colonization and development by microorganisms difficult to eradicate. Some of the positive and negative applications of biocides are discussed below.

Contemporary agriculture requires the application of pesticides, but overuse of such pesticides can cause very dangerous environmental and ecological problems. Some pesticides are biodegradable, but others remain in the environment or ecosystem (Alavanja et al., 2004; Lee et al., 2008; Hancock et al., 2008). One study suggested that some pesticides may be the major cause of neurodegenerative disease, as they can halt the biological processes in mitochondria (Le Couteur et al., 1999). These negative effects of chemical biocides and pesticides have thus caused researchers to consider alternative, more environmentally friendly sources from which biocides can be obtained.

At present, the biocides available on the market have a specific mechanism of action that stops cell wall synthesis and causes DNA and RNA damage (Singer et al., 2010; Ashraf et al., 2014). Therefore, chemically synthesized biocides are highly toxic to humans, even affecting those who merely touch them. Conservators are among those most commonly exposed to toxic biocides (Varnai et al., 2011). Green plants may be a better alternative to chemical biocides, due to their low toxicity, ease of handling, and environmental suitability (Silva et al., 2016).

Many plants contain secondary metabolites that are important bioactive molecules and contain flavonoids, phenylpropanoids, and anthraquinones. (Rios and Recio, 2005; Svetaz et al., 2013). Fungi in the form of mushrooms and plants in the form of vegetables are served as foods; both groups can alter the activity of many enzymes like tyrosinase and melanine oxidase and can play vital roles in boosting health (Arrebola et al., 2015; He et al., 2015). Some plants and fungi are used as the most promising biocides and are environmentally and ecologically friendly.

A species of tree known as *Metopium brownie* belongs to the family Anacardicaceae and synthesizes eriodictyol, which contains naturally antifungal properties (de Rodríguez et al., 2006). Secondary metabolites extracted from *Calceolaria,* which belongs to the family Calceolariaceae, also show antifungal and antibacterial properties (Cespedes et al., 2015). A search over the last few decades for secondary metabolites of fungi led to the discovery of biochemical novelties and vigorous molecules, many of which had antimicrobial properties. *Hypholoma capnoides,* of the basidiomycetes family, produces *p*-anisaldehyde and 3-chloro-*p*-anisaldehyde, which has biological properties. *H. sublateritium* produces 3,5-dichloro-4-methox- ybenzyl alcohol and marasmal (Aqueveque et al., 2010).

A study conducted by do Rosário Martins et al. (2014) assessed the antimicrobial properties of oils extracted from the fruit and leaves of *Schinus molle*. The chemical compositions of these essential oils (EO) were analyzed using gas chromatography. The results showed that fruit and leaf EO were effective and displayed good antibacterial and antifungal properties. The leaf and fruit EO produced the same effects against *Fusarium oxysporum* and *Aspergillus* spp., but the leaf EO proved more effective against *Rhizopus* spp.

Another study conducted by Foksowicz-Flaczyk et al. (2008) aimed to use environmentally friendly biocides to treat bio-deteriorating textiles. They used mint oil (*Mentha piperita*) as a biocide. Treating the fungus *Aspergillus niger* with concentrated mint oil showed some levels of antifungal activity. The growth of *Aspergillus niger* was only slightly visible through a microscope but not to the naked eye. A similar study was conducted by Stupar et al. (2014) in which the plant's essential oil and benzalkonium chloride were used against fungal species isolated from the stone surface. This essential oil was taken from the plant family Lamiaceae. The tested fungi species were isolated from stone substrata (*Epicoccum nigrum* and *Bipolaris spicifera*). Among all the essential oils (EO), *O. vulgare* EO proved to be the most effective with regards to antifungal properties, with *L. angustifolia* EO coming in second and *R. offocinalis* EO coming third. Strong antifungal activity was shown by benzalkonium chloride against all fungal species. It was thus concluded that the high antifungal efficacy of *O. vulgare*, which was almost the same as that shown by benzalkonium chloride, may have been due to the presence of phenol carvacrol in the EO. Jeong et al. (2018) showed that Eugenol mixed with emulsifiers has great antifungal activity with a rate of almost 80% microbial reduction observed. All these studies suggest that essential oils may be effective, environmentally friendly alternative antifungal agents.

6. Conclusions and prospects

Microbes are both abundant and diverse. Though their impact on heritage sites may not be immediate, it is important to assess their impact over the long term to gain comprehensive understanding. A promising strategy for conserving stone monuments is thus to limit microbial colonization and further growth if possible. This can be achieved in two ways. The first is to develop a simple, rapid and novel method for evaluating and monitoring the microbial community on stone monuments. Many innovative methods are being used for synthesizing the nanomaterials and biocides. In green synthesis, microbial species and plant species have been used, but until now, mammalian cells have never been used to synthesize nanomaterials, so this might be considered a new direction for nanomaterials synthesis. The second is to close the presently existing gaps of information between microbial physiology and microbial community so that physiologically active microorganisms can be easily identified in the future. This review summarized the role of microflora in the biodeterioration process of cultural heritage sites. We outlined how chemicals secreted by some microorganisms lead to the deterioration of cultural heritage sites. We also summarized different methods used to mitigate and control the biodeterioration of cultural heritage sites. Using physical methods like UV-C radiation exposure mainly depends on the cultural heritage site. The association between physical methods for control of biodeterioration and microorganisms must be analyzed.

Biocides are one of the most effective methods to control microorganisms for cultural heritage conservation because of their acute toxicity to living microorganisms. In this review, we evaluated different types of biocides. Inorganic biocides are shown to be more effective than organic biocides. In the future, more studies should be conducted to consider inorganic nanoparticles such as ZnO, TiO_2, and Ag_2O. TiO_2 is the most efficient photocatalyst. The effect of employing these inorganic biocides is long-lasting. Nanoparticles have certain environmental benefits different from organics because they cannot be eliminated by biotransformation by microorganisms.

Green nanotechnology has been designed for direct application to environmental problems. Green nanotechnology is a sustainable option because it is ecofriendly and harmless to humans. The green synthesized biocide, which are derived from the *Bacillus* species, is important for its eco-friendliness. The *Bacillus* species produces many metabolites with antifungal activity. These metabolites must be assessed for their biocidal ability and lasting effectiveness in vitro and on site. Proper mechanical and physical methods should be combined with chemical control measures to ensure long-term conservation of cultural properties. Since the functions of microbial communities are still not fully understood, omics technologies can provide useful information for conservators. Moreover, continuous monitoring and control of environmental conditions, including temperature, relative humidity, water contents and pollutants, should be a high priority in overall prevention and conservation activities.

Acknowledgements

This study was supported by the China Gansu Province Science and Technology Plan (No. 18JR3RA004, 1604WKCA003), the National Natural Science Foundation of China (No. 31560160, 31500430), and the Light of West China Program of the Chinese Academy of Sciences. We also would like to thank Bridget Nicholas from Harvard University for English text editing.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ibiod.2019.104721.

References

Abdel-Haliem, M.E.F., Sakr, A.A., Ali, M.F., Ghaly, M.F., Sohlenkamp, C., 2013. Characterization of *Streptomyces* isolates causing colour changes of mural paintings in ancient Egyptian tombs. Microbiol. Res. 168 (7), 428–437. https://doi.org/10.1016/j.micres.2013.02.004.

Adamiak, J., Otlewska, A., Tafer, H., Lopandic, K., Gutarowska, B., Sterflinger, K., Piñar, G., 2018. First evaluation of the microbiome of built cultural heritage by using the Ion Torrent next generation sequencing platform. Int. Biodeter. Biodegr. 131, 11–18. https://doi.org/10.1016/j.ibiod.2017.01.040.

Adamo, M., Brizzi, M., Magaudda, G., Martinelli, G., Plossi-Zappala, M., Rocchetti, F., Savagnone, F., 2001. Gamma radiation treatment of paper in different environmental conditions: chemical, physical and microbiological analysis. Restaurator 22 (2), 107–131. https://doi.org/10.1515/REST.2001.107.

Alavanja, M.C., Dosemeci, M., Samanic, C., Lubin, J., Lynch, C.F., Knott, C., Barker, J., Hoppin, J.A., Sandler, D.P., Coble, J., Thomas, K., 2004. Pesticides and lung cancer risk in the agricultural health study cohort. Am. J. Epidemiol. 160 (9), 876–885. https://doi.org/10.1093/aje/kwh290.

Ali, I., Qiang, T.Y., Ilahi, N., Adnan, M., Sajjad, W., 2018. Green synthesis of silver nanoparticles by using bacterial extract and its antimicrobial activity against pathogens. Int. J. Biosci. 13 (5), 1–15. https://doi.org/10.12692/ijb/13.5.1-15.

Allsopp, Dennis, 2011. Worldwide wastage: the economics of biodeterioration. Microbiol. Tod. 38, 150–153.

Aqueveque, P., Anke, T., Saéz, K., Silva, M., Becerra, J., 2010. Antimicrobial activity of submerged cultures of Chilean basidiomycetes. Planta Med. 76 (15), 1787–1791. https://doi.org/10.1055/s-0030-1249853.

Arrebola, J.P., Fernández, M.F., Martin-Olmedo, P., Bonde, J.P., Martín-Rodriguez, J.L., Expósito, J., Rubio-Domínguez, A., Olea, N., 2015. Historical exposure to persistent organic pollutants and risk of incident hypertension. Environ. Res. 217 − 223. https://doi.org/10.1016/j.envres.2015.02.018.

Ashraf, M.A., Ullah, S., Ahmad, I., Qureshi, A.K., Balkhair, K.S., Abdur Rehman, M., 2014. Green biocides, a promising technology: current and future applications to industry and industrial processes. J. Sci. Food. Agr. 94 (3), 388–403. https://doi.org/10.1002/jsfa.6371.

Banat, I.M., De Rienzo, M.A.D., Quinn, G.A., 2014. Microbial biofilms: biosurfactants as antibiofilm agents. Appl. Microbiol. Biot. 98 (24), 9915–9929. https://doi.org/10.1007/s00253-014-6169-6.

Bartoli, F., Municchia, A.C., Futagami, Y., Kashiwadani, H., Moon, K.H., Caneva, G., 2014. Biological colonization patterns on the ruins of Angkor temples (Cambodia) in the biodeterioration vs bioprotection debate. Int. Biodeter. Biodegr. 96, 157–165. https://doi.org/10.1016/j.ibiod.2014.09.015.

Bastian, F., Alabouvette, C., 2009. Lights and shadows on the conservation of a rock art cave: the case of Lascaux Cave. Int. J. Speleol. 38 (1), 55–60. https://doi.org/10.5038/1827-806X.38.1.6.

Belfiore, C.M., Ricca, M., La Russa, M.F., Ruffolo, S.A., Galli, G., Barca, D., Malagodi, M., Vallefuoco, M., Sprovieri, M., Pezzino, A., 2016. Provenance study of building and statuary marbles from the Roman archaeological site of “Villa dei Quintili”(Rome, Italy). Ital. J. Geosci. 135 (2), 236–249. https://doi.org/10.3301/IJG.2015.19.

Bruno, F., Muzzupappa, M., Barbieri, L., Gallo, A., Ritacco, G., Lagudi, A., La Russa, M.F., Ruffolo, S.A., Crisci, G.M., Ricca, M., Comite, V., 2016. The CoMAS project: new materials and tools for improving the in situ documentation, restoration, and conservation of underwater archaeological remains. Mar. Technol. Soc. J. 50 (4), 108–118. https://doi.org/10.4031/MTSJ.50.4.2.

Cao, H., Auguet, J.C., Gu, J.D., 2013. Global ecological pattern of ammonia-oxidizing archaea. PLoS One 8 (2), 52853. https://doi:10.1371/journal.pone.0052853.

Carter, N.E.A., Viles, H.A., 2003. Experimental investigations into the interactions between moisture, rock surface temperatures and an epilithic lichen cover in the bioprotection of limestone. Build. Environ. 38 (9–10), 1225–1234. https://doi.org/10.1016/S0360-1323(03)00079-9.

Céspedes, C.L., Alarcon, J.E., Aqueveque, P., Seigler, D.S., Kubo, I., 2015. In the search for new secondary metabolites with biopesticidal properties. Isr. J. Plant Sci. 62 (4), 216–228. https://doi.org/10.1080/07929978.2015.1006424.

Ciferri, O., 2002. The role of microorganisms in the degradation of cultural heritage. Stud. Conserv. 47 (Suppl. 1), 35–45.

Concha-Lozano, N., Gaudon, P., Pages, J., De Billerbeck, G., Lafon, D., Eterradossi, O., 2012. Protective effect of endolithic fungal hyphae on oolitic limestone buildings. J. Cult. Herit. 13 (2), 120–127. http://doi.org/10.1016/j.culher.2011.07.006.

Crispim, C.A., Gaylarde, C.C., 2005. Cyanobacteria and biodeterioration of cultural heritage: a review. Microb. Ecol. 49 (1), 1–9. https://doi.org/10.1007/s00248-003-1052-5.

Crupi, V., Fazio, B., Gessini, A., Kis, Z., La Russa, M.F., Majolino, D., Masciovecchio, C., Ricca, M., Rossi, B., Ruffolo, S.A., Venuti, V., 2018. TiO2–SiO2–PDMS nanocomposite coating with self-cleaning effect for stone material: finding the optimal amount of TiO2. Constr. Build. Mater. 166, 464–471. https://doi.org/10.1016/j.conbuildmat.2018.01.172.

De los Rios, A.Galvan, V.Ascaso, C. In situ microscopical diagnosis of biodeterioration processes at the convent of Santa Cruzla Real Segovia, Spain. Int. Biodetr. Biodegr. https://doi.org/10.1016/j.ibiod.2004.03.020 54, 2–3, 113–120.

de Rodríguez, D.J., Angulo-Sánchez, J.L., Hernández-Castillo, F.D., 2006. An overview of the antimicrobial properties of Mexican medicinal plants. Adv. Phytomed. 3, 325–377. https://doi.org/10.1016/S1572-557X(06)03014-5.

Dresler, C., Saladino, M.L., Demirbag, C., Caponetti, E., Martino, D.F.C., Alduina, R., 2017. Development of controlled release systems of biocides for the conservation of cultural heritage. Int. Biodeter. Biodegr. 125, 150–156. https://doi.org/10.1016/j.ibiod.2017.09.007.

Duan, Y., Wu, F., Wang, W., Gu, J.D., Li, Y., Feng, H., Chen, T., Liu, G., An, L., 2018. Differences of Microbial Community on the wall paintings preserved *in situ* and *ex situ* of the Tiantishan Grottoes, China. Int. Biodeter. Biodegr. 132, 102–113. https://doi.org/10.1016/j.ibiod.2018.02.013.

El-Feky, O.M., Hassan, E.A., Fadel, S.M., Hassan, M.L., 2014. Use of ZnO nanoparticles for protecting oil paintings on paper support against dirt, fungal attack, and UV aging. J. Cult. Herit. 15 (2), 165–172. https://doi.org/10.1016/j.culher.2013.01.012.

Farooq, M., Hassan, M., Gull, F., 2015. Mycobial deterioration of stone monuments of Dharmarajika, Taxila. J. Microb. Exp. 2, 36. https://doi.org/10.15406/jmen.2015.02.00036.

Favero-Longo, S.E., Benesperi, R., Bertuzzi, S., Bianchi, E., Buffa, G., Giordani, P., Loppi, S., Malaspina, P., Matteucci, E., Paoli, L., Ravera, S., 2017. Species-and site-specific efficacy of commercial biocides and application solvents against lichens. Int. Biodeter. Biodegr. 123, 127–137. https://doi.org/10.1016/j.ibiod.2017.06.009.

Favero-Longo, S.E., Brigadeci, F., Segimiro, A., Voyron, S., Cardinali, M., Girlanda, M., Piervittori, R., 2018. Biocide efficacy and consolidant effect on the mycoflora of historical stuccos in indoor environment. J. Cult. Herit. 34, 33–42. https://doi.org/10.1016/j.culher.2018.03.017.

Fernandes, P., 2006. Applied microbiology and biotechnology in the conservation of stone cultural heritage materials. Appl. Microbiol. Biot. 73 (2), 291. https://doi.org/10.1007/s00253-006-0599-8.

Foksowicz-Flaczyk, J., Walentowska, J., 2008. Eco-friendly antimicrobial finishing of natural fibres. Mol. Cryst. Liq. Cryst. 484 (1), 207–573. https://doi.org/10.1080/15421400801904526.

Fonseca, A.J., Pina, F., Macedo, M.F., Leal, N., Romanowska-Deskins, A., Laiz, L., Gómez-Bolea, A., Saiz-Jimenez, C., 2010. Anatase as an alternative application for preventing biodeterioration of mortars: evaluation and comparison with other biocides. Int. Biodeter. Biodegr. 64 (5), 388–396. https://doi.org/10.1016/j.ibiod.2010.04.006.

Gaylarde, C.C., Morton, L.H.G., Loh, K., Shirakawa, M.A., 2011. Biodeterioration of external architectural paint films–a review. Int. Biodeter. Biodegr. 65 (8), 1189–1198. https://doi.org/10.1016/j.ibiod.2011.09.005.

Griffin, P.S., Indictor, N., Koestler, R.J., 1991. The biodeterioration of stone: a review of deterioration mechanisms, conservation case histories, and treatment. Int. Biodeterior. 28 (1–4), 187–207. https://doi.org/10.1016/0265-3036(91)90042-P.

Gu, J.D., 2003. Microbiological deterioration and degradation of synthetic polymeric materials: recent research advances. Int. Biodeter. Biodegr. 52 (2), 69–91. https://doi.org/10.1016/S0964-8305(02)00177-4.

Gu, J.D., Ford, T.E., Berke, N.S., Mitchell, R., 1998. Biodeterioration of concrete by the fungus Fusarium. Int. Biodeter. Biodegr. 41 (2), 101–109. https://doi.org/10.1016/S0964-8305(98)00034-1.

Gu, J.D., Ford, T.E., Mitchell, R., 1996. Susceptibility of electronic insulating polyimides to microbial degradation. J. Appl. Polym. Sci. 62 (7), 1029–1034. https://doi.org/10.1002/(SICI)1097-4628(19961114)62:7 < 1029::AID-APP8 > 3.0.CO;2-M.

Gu, J.D., Katayama, Y., 2017. A microbiological challenge in the protection of the sandstone Angkor monuments in Cambodia. IIC News Conserv. 63, 7–9.

Gutarowska, B., Skora, J., Zduniak, K., Rembisz, D., 2012. Analysis of the sensitivity of microorganisms contaminating museums and archives to silver nanoparticles. Int. Biodeter. Biodegr. 68, 7–17. https://doi.org/10.1016/j.ibiod.2011.12.002.

Hancock, D.B., Martin, E.R., Mayhew, G.M., Stajich, J.M., Jewett, R., Stacy, M.A., Scott, B.L., Vance, J.M., Scott, W.K., 2008. Pesticide exposure and risk of Parkinson's disease: a family-based case-control study. BMC Neurol. 8 (1), 6–18. https://doi.org/10.1186/1471-2377-8-6.

He, X., Jing, Y., Wang, J., Li, K., Yang, Q., Zhao, Y., Li, R., Ge, J., Qiu, X., Li, G., 2015. Significant accumulation of persistent organic pollutants and dysregulation in multiple DNA damage repair pathways in the electronic-waste-exposed populations. Environ. Res. 137, 458–466. https://doi.org/10.1016/j.envres.2014.11.018.

Hu, H., Ding, S., Katayama, Y., Kusumi, A., Li, S.X., de Vries, R.P., Wang, J., Yu, X.Z., Gu, J.D., 2013. Occurrence of Aspergillus allahabadii on sandstone at Bayon temple, Angkor Thom, Cambodia. Int. Biodeter. Biodegr. 76, 112–117. https://doi.org/10.1016/j.ibiod.2012.06.022.

Jeong, S.H., Lee, H.J., Kim, D.W., Chung, Y.J., 2018. New biocide for eco-friendly biofilm removal on outdoor stone monuments. Int. Biodeter. Biodegr. 131, 19–28. https://doi.org/10.1016/j.ibiod.2017.03.004.

Koestler, R.J., 2000. Polymers and Resins as Food for Microbes. Of Microbes and Art. Springer, Boston, MA, pp. 153–167. https://doi.org/10.1007/978-1-4615-4239-1_11.

Kusumi, A., Li, X., Osuga, Y., Kawashima, A., Gu, J.D., Nasu, M., Katayama, Y., 2013. Bacterial communities in pigmented biofilms formed on the sandstone bas-relief walls of the Bayon Temple, Angkor Thom, Cambodia. Microbes. Environ. 28 (4), 422–431. https://doi.org/10.1264/jsme2.ME13033.

La Russa, M.F., Macchia, A., Ruffolo, S.A., De Leo, F., Barberio, M., Barone, P., Crisci, G.M., Urzì, C., 2014. Testing the antibacterial activity of doped TiO$_2$ for preventing biodeterioration of cultural heritage building materials. Int. Biodeter. Biodegr. 96, 87–96. https://doi.org/10.1016/j.ibiod.2014.10.002.

Le Couteur, D.G., McLean, A.J., Taylor, M.C., Woodham, B.L., Board, P.G., 1999.

Pesticides and Parkinson's disease. Biomed. Pharmacother. 53 (3), 122–130. https://doi.org/10.1016/S0753-3322(99)80077-8.

Lee, H.S., Lee, K.S., Yu, K., Hong, S.Y., 2008. Expression of genes related to Parkinson's disease after paraquat treatment in Drosophila melanogaster. Pestic. Biochem. Phys. 92 (1), 19–23. https://doi.org/10.1016/j.pestbp.2008.05.002.

Lepidi, A.A., Schippa, G., 1972. Growth of a sulphide oxidizing bacterium estimated by various methods and a new method of sulphide determination. In: Colloque international sur la deterioration des pierres en oeuvre. 1er. International Symposium on the Deterioration of Building Stones, pp. 139–141.

Liu, X., Meng, H., Wang, Y., Katayama, Y., Gu, J.D., 2018. Water is a critical factor in evaluating and assessing microbial colonization and destruction of Angkor sandstone monuments. Int. Biodeter. Biodegr. 133, 9–16. https://doi.org/10.1016/j.ibiod.2018.05.011.

Ma, Y., Zhang, H., Du, Y., Tian, T., Xiang, T., Liu, X., Wu, F., An, L., Wang, W., Gu, J.D., Feng, H., 2015. The community distribution of bacteria and fungi on ancient wall paintings of the Mogao Grottoes. Sci. Rep. 5, 7752. https://doi.org/10.1038/srep07752.

Macchia, A., Mangini, F., Ruffolo, S.A., Muzi, M., Rivaroli, L., Ricca, M., La Russa, M.F., Frezza, F., 2017. A novel model to detect the content of inorganic nanoparticles in coatings used for stone protection. Prog. Org. Coat. 106, 177–185. https://doi.org/10.1016/j.porgcoat.2016.11.004.

Maillard, J.Y., 2005. Antimicrobial biocides in the healthcare environment: efficacy, usage, policies, and perceived problems. Ther. Clin. Risk Manag. 1 (4), 307–320.

Macedo, M.F., Miller, A.Z., Dionísio, A., Saiz-Jimenez, C., 2009. Biodiversity of cyanobacteria and green algae on monuments in the Mediterranean Basin: an overview. Microbiology 155 (1), 3476–3490. https://dx.doi.org/10.1099/mic.0.032508-0.

Martin-Sanchez, P.M., Nováková, A., Bastian, F., Alabouvette, C., Saiz-Jimenez, C., 2012. Use of biocides for the control of fungal outbreaks in subterranean environments: the case of the Lascaux Cave in France. Environ. Sci. Technol. 46 (7), 3762–3770. https://dx.doi.org/10.1021/es2040625.

McDonnell, G., Russell, A.D., 1999. Antiseptics and disinfectants: activity, action, and resistance. Clin. Microbiol. Rev. 12 (1), 147–179. https://doi.org/10.1016/S0266-7681(05)80179-2.

Meng, H., Katayama, Y., Gu, J.D., 2017. More wide occurrence and dominance of ammonia-oxidizing archaea than bacteria at three Angkor sandstone temples of Bayon, Phnom Krom and Wat Athvea in Cambodia. Int. Biodeter. Biodegr. 117, 78–88. https://doi.org/10.1016/j.ibiod.2016.11.012.

Meng, H., Luo, L., Chan, H.W., Katayama, Y., Gu, J.D., 2016. Higher diversity and abundance of ammonia-oxidizing archaea than bacteria detected at the Bayon Temple of Angkor Thom in Cambodia. Int. Biodeter. Biodegr. 115, 234–243. https://doi.org/10.1016/j.ibiod.2016.08.021.

Mitchell, R., Gu, J.D., 2000. Changes in the biofilm microflora of limestone caused by atmospheric pollutants. Int. Biodeter. Biodegr. 46 (4), 299–303. https://doi.org/10.1016/S0964-8305(00)00105-0.

Mora, P., Mora, L., Philippot, P., 1984. The Conservation of Wall Paintings. Butterworth & Co, pp. 350.

Municchia, A.C., Bartoli, F., Taniguchi, Y., Giordani, P., Caneva, G., 2018. Evaluation of the biodeterioration activity of lichens in the cave Church of Üzümlü (Cappadocia, Turkey). Int. Biodeter. Biodegr. 127, 160–169. https://doi.org/10.1016/j.ibiod.2017.11.023.

Naylor, L.A., Viles, H.A., Carter, N.E.A., 2002. Biogeomorphology revisited: looking towards the future. Geomorphology 47 (1), 3–14. https://doi.org/10.1016/S0169-555X(02)00137-X.

Ortega-Morales, B.O., Reyes-Estebanez, M.M., Gaylarde, C.C., Camacho-Chab, J.C., Sanmartín, P., Chan-Bacab, M.J., Granados-Echegoyen, C.A., Pereañez-Sacarias, J.E., 2018. Antimicrobial properties of nanomaterials used to control microbial colonization of stone substrata. In: Advanced Materials for the Conservation of Stone. Springer, Cham, pp. 277–298. https://doi.org/10.1007/978-3-319-72260-3_13.

Paulus, W., 2012. Microbicides for the Protection of Materials: a Handbook, first ed. Springer Science & Business Media, pp. 38.

Pereira-Pardo, L., Korenberg, C., 2018. The use of erbium lasers for the conservation of cultural heritage. A review. J. Cult. Herit. 31, 236–247. https://doi.org/10.1016/j.culher.2017.10.007.

Petersen, K., Kuroczkin, J., Strzelczyk, A.B., Krumbein, W.E., 1988. Distribution and effects of fungi on and in sandstones. In. Biodeter. 7, 123–128. https://doi.org/10.1007/978-94-009-1363-9_16.

Pfendler, S., Borderie, F., Bousta, F., Alaoui-Sosse, L., Alaoui-Sosse, B., Aleya, L., 2018. Comparison of biocides, allelopathic substances and UV-C as treatments for biofilm proliferation on heritage monuments. J. Cult. Herit. 33, 117–124. https://doi.org/10.1016/j.culher.2018.03.016.

Pinna, D., Salvadori, B., Galeotti, M., 2012. Monitoring the performance of innovative and traditional biocides mixed with consolidants and water-repellents for the prevention of biological growth on stone. Sci. Total Environ. 423, 132–141. https://doi.org/10.1016/j.scitotenv.2012.02.012.

Rakotonirainy, M.S., Caillat, L., Héraud, C., Memet, J.B., Tran, Q.K., 2007. Effective biocide to prevent microbiological contamination during PEG impregnation of wet archaeological iron-wood artefacts. J. Cult. Herit. 8 (2), 160–169. https://doi.org/10.1016/j.culher.2007.01.005.

Ranalli, G., Zanardini, E., Pasini, P., Roda, A., 2003. Rapid biodeteriogen and biocide diagnosis on artworks: a bioluminescent low-light imaging technique. Ann. Microbiol. 53 (1), 1–14.

Ricca, M., Belfiore, C.M., Ruffolo, S.A., Barca, D., De Buergo, M.A., Crisci, G.M., La Russa, M.F., 2015. Multi-analytical approach applied to the provenance study of marbles used as covering slabs in the archaeological submerged site of Baia (Naples, Italy): the case of the "Villa con ingresso a protiro". Appl. Surf. Sci. 357, 1369–1379. https://doi.org/10.1016/j.apsusc.2015.10.002.

Richardson, B., 1973. Control of biological growths. Stone Ind. 8, 22–26.

Rios, J.L., Recio, M.C., 2005. Medicinal plants and antimicrobial activity. J. Enthnopharmacol. 100 (1–2), 80–84. https://doi.org/10.1016/j.jep.2005.04.025.

Rivadeneyra, M.A., Párraga, J., Delgado, R., Ramos-Cormenzana, A., Delgado, G., 2004. Biomineralization of carbonates by Halobacillus trueperi in solid and liquid media with different salinities. FEMS Microbiol. Ecol. 48 (1), 39–46. https://doi.org/10.1016/j.femsec.2003.12.008.

Rivas, T., Pozo-Antonio, J.S., de Silanes, M.L., Ramil, A., López, A.J., 2018. Laser versus scalpel cleaning of crustose lichens on granite. Appl. Surf. Sci. 440, 467–476. https://doi.org/10.1016/j.apsusc.2018.01.167.

Ruffolo, S.A., De Leo, F., Ricca, M., Arcudi, A., Silvestri, C., Bruno, L., Urzì, C., La Russa, M.F., 2017a. Medium-term in situ experiment by using organic biocides and titanium dioxide for the mitigation of microbial colonization on stone surfaces. Int. Biodeter. Biodegr. 123, 17–26. https://doi.org/10.1016/j.ibiod.2017.05.016.

Ruffolo, S.A., Ricca, M., Macchia, A., La Russa, M.F., 2017b. Antifouling coatings for underwater archaeological stone materials. Prog. Org. Coat. 104, 64–71. https://doi.org/10.1016/j.porgcoat.2016.12.004.

Sabev, H.A., Barratt, S.R., Handley, P.S., Robson, G.D., Greenhalgh, Malcom, 2006. Biodegradation and biodeterioration of man-made polymeric materials. In: Gadd, G.M. (Ed.), Fungi in Biogeochemical Cycles. Cambridge University Press, Cambridge.

Salimbeni, R., 2006. Laser techniques for conservation of artworks. Archeometriai Muhely 3 (1), 34–40.

Salvadori, O., Municchia, A.C., 2016. April. The role of fungi and lichens in the biodeterioration of stone monuments. JJAP Conf. Proc. 7, 39–54. https://doi.org/10.2174/2210289201607020039.

Sanz, M., Oujja, M., Ascaso, C., Pérez-Ortega, S., Souza-Egipsy, V., Fort, R., de los Rios, A., Wierzchos, J., Cañamares, M.V., Castillejo, M., 2017. Influence of wavelength on the laser removal of lichens colonizing heritage stone. Appl. Surf. Sci. 399, 758–768. https://doi.org/10.1016/j.apsusc.2016.12.032.

Savković, Ž.D., Stupar, M.Č., Grbić, M.V.L., Vukojević, J.B., 2016. Comparison of anti-Aspergillus activity of Origanum vulgare L. essential oil and commercial biocide based on silver ions and hydrogen peroxide. Acta Bot. Croat. 75 (1), 121–128. https://doi.org/10.1515/botcro-2016-0011.

Schaefer, M.O., Gutzmer, J., Beukes, N.J., 2001. Late Paleoproterozoic Mn-rich oncoids: earliest evidence for microbially mediated Mn precipitation. Geology 29 (9), 835–838. https://doi.org/10.1130/0091-7613(2001)029%3C0835:LPMROE%3E2.0.CO;2.

Scheerer, S., Ortega-Morales, O., Gaylarde, C., 2009. Microbial deterioration of stone monuments—an updated overview. Adv. Appl. Microbiol. 66, 97–139. https://doi.org/10.1016/S0065-2164(08)00805-8.

Sierra-Fernandez, A., Gomez-Villalba, L.S., De la Rosa-García, S.C., Gomez-Cornelio, S., Quintana, P., Rabanal, M.E., Fort, R., 2018. Inorganic nanomaterials for the consolidation and antifungal protection of stone heritage. In: Advanced Materials for the Conservation of Stone. Springer, Cham, pp. 125–149. https://doi.org/10.1007/978-3-319-72260-3_6.

Silva, M., Pereira, A., Teixeira, D., Candeias, A., Caldeira, A.T., 2016. Combined use of NMR, LC-ESI-MS and antifungal tests for rapid detection of bioactive lipopeptides produced by *Bacillus*. Adv. Microbiol. 6 (10), 788–796. https://doi.org/10.4236/aim.2016.610077.

Silva, M., Rosado, T., Teixeira, D., Candeias, A., Caldeira, A.T., 2015. Production of green biocides for cultural heritage, novel biotechnological solutions. Int. J. Conserv. Sci. 6, 519–530.

Singer, H., Jaus, S., Hanke, I., Lück, A., Hollender, J., Alder, A.C., 2010. Determination of biocides and pesticides by on-line solid phase extraction coupled with mass spectrometry and their behaviour in wastewater and surface water. Environ. Pollut. 158 (10), 3054–3064. https://doi.org/10.1016/j.envpol.2010.06.013.

Sohrabi, M., Favero-Longo, S.E., Pérez-Ortega, S., Ascaso, C., Haghighat, Z., Talebian, M.H., Fadaei, H., de los Ríos, A., 2017. Lichen colonization and associated deterioration processes in Pasargadae, UNESCO world heritage site, Iran. Int. Biodeter. Biodegr. 117, 171–182. https://doi.org/10.1016/j.ibiod.2016.12.012.

Steinbauer, M.J., Gohlke, A., Mahler, C., Schmiedinger, A., Beierkuhnlein, C., 2013. Quantification of wall surface heterogeneity and its influence on species diversity at medieval castles–implications for the environmentally friendly preservation of cultural heritage. J. Cult. Herit. 14 (3), 219–228. https://doi.org/10.1016/j.culher.2012.06.003.

Sterflinger, K., Piñar, G., 2013. Microbial deterioration of cultural heritage and works of art—tilting at windmills? Appl. Microbiol. Biot. 97 (22), 9637–9646. https://doi.org/10.1007/s00253-013-5283-1.

Stupar, M., Grbić, M.L., Džamić, A., Unković, N., Ristić, M., Jelikić, A., Vukojević, J., 2014. Antifungal activity of selected essential oils and biocide benzalkonium chloride against the fungi isolated from cultural heritage objects. S. Afr. J. Bot. 118–124. https://doi.org/10.1016/j.sajb.2014.03.016.

Svetaz, L., Derita, M., Rodríguez, M.V., Postigo, A., Butassi, E., Castelli, M.V., Sortino, M., Petenatti, E., Zacchino, S., 2013. Antifungal compounds from Latin American plants. In: Antifungal Metabolites from Plants. Springer, Berlin Heidelerg, pp. 3–26. https://doi.org/10.1007/978-3-642-38076-1_1.

Tykot, R.H., Herrmann, J.J., van der Merwe, N.J., Newman, R., Allegretto, K.O., 2002. Thasian marble sculptures in European and American collections: isotopic and other analyses. ASMOSIA 5: interdisciplinary Studies on Ancient Stone. In: Herrmann, J., Herz, N., Newman, R. (Eds.), Proceedings of the Fifth International Conference of the Association for the Study of Marble and Other Stones in Antiquity, Museum of Fine Arts, Boston, 1998, pp. 188–195 London, 2002.

Unger, A., 2012. Decontamination and "deconsolidation" of historical wood preservatives and wood consolidants in cultural heritage. J. Cult. Herit. 13 (3) S196 − S202. https://doi.org/10.1016/j.culher.2012.01.015.

Urzì, C., De Leo, F., 2007. Evaluation of the efficiency of water-repellent and biocide compounds against microbial colonization of mortars. Int. Biodeter. Biodegr. 60 (1), 25–34. https://doi.org/10.1016/j.ibiod.2006.11.003.

Urzì, C., De Leo, F., Krakova, L., Pangallo, D., Bruno, L., 2016. Effects of biocide treatments on the biofilm community in Domitilla's catacombs in Rome. Sci. Total Environ. 572, 252–262. https://doi.org/10.1016/j.scitotenv.2016.07.195.

Vannini, A., Contardo, T., Paoli, L., Scattoni, M., Favero-Longo, S.E., Loppi, S., 2018. Application of commercial biocides to lichens: does a physiological recovery occur over time? Int. Biodeter. Biodegr. 129, 189–194. https://doi.org/10.1016/j.ibiod.2018.02.010.

Varnai, V.M., Macan, J., Ljubičić Ćalušić, A., Prester, L., Kanceljak Macan, B., 2010. Upper respiratory impairment in restorers of cultural heritage. Occup. Med. 61 (1), 45–52. https://doi.org/10.1093/occmed/kqq170.

Vijayakumar, S., 2014. Role of Cyanobacteria in biodeterioration of historical monuments –A review. BMR. Microbiol. 1, 1–13.

Voltolina, S., Nodari, L., Aibeo, C., Egel, E., Pamplona, M., Simon, S., Falzacappa, E.V., Scopece, P., Gambirasi, A., Favaro, M., Patelli, A., 2016. Assessment of plasma torches as innovative tool for cleaning of historical stone materials. J. Cult. Herit. 22, 940–950. https://doi.org/10.1016/j.culher.2016.05.001.

Wang, W., Ma, X., Ma, Y., Mao, L., Wu, F., Ma, X., An, L., Feng, H., 2011. Molecular characterization of airborne fungi in caves of the Mogao Grottoes, Dunhuang, China. Int. Biodeter. Biodegr. 65 (5), 726–731. https://doi.org/10.1016/j.ibiod.2011.04.006.

Wang, W., Ma, Y., Ma, X., Wu, F., Ma, X., An, L., Feng, H., 2012. Diversity and seasonal dynamics of airborne bacteria in the Mogao Grottoes, Dunhuang, China. Aerobiologia 28 (1), 27–38. https://doi.org/10.1007/s10453-011-9208-0.

Warscheid, T., Braams, J., 2000. Biodeterioration of stone: a review. Int. Biodeter. Biodegr. 46 (4), 343–368. https://doi.org/10.1016/S0964-8305(00)00109-8.

Wu, F., Wang, W., Feng, H., Gu, J.D., 2017. Realization of biodeterioration to cultural heritage protection in China. Int. Biodeter. Biodegr. 117, 128–130. https://doi.org/10.1016/j.ibiod.2016.12.002.

Young, M.E., Alakomi, H.L., Fortune, I., Gorbushina, A.A., Krumbein, W.E., Maxwell, I., McCullagh, C., Robertson, P., Saarela, M., Valero, J., Vendrell, M., 2008. Development of a biocidal treatment regime to inhibit biological growths on cultural heritage: BIODAM. Environ. Geol. 56 (3–4), 631–641. https://doi.org/10.1007/s00254-008-1455-1.

Zhang, X., Ge, Q., Zhu, Z., Deng, Y., Gu, J.D., 2018. Microbiological community of the royal Palace in Angkor Thom and Beng Mealea of Cambodia by illumina sequencing based on 16S rRNA gene. Int. Biodeter. Biodegr. 134, 127–135. https://doi.org/10.1016/j.ibiod.2018.06.018.