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A review of polycyclic aromatic hydrocarbon-induced microbial deterioration of mediterranean heritage and conservation strategies

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The Mediterranean region, distinguished by its cultural heritage and industrialization, suffers from significant pollution levels of polycyclic aromatic hydrocarbons (PAHs) due to frequent petroleum-related activities such as petroleum extraction and consumption. Being dominant pollutants, PAHs cause significant deterioration of historic buildings and materials. This review comprehensively explores PAH-induced heritage deterioration, especially PAH-induced microbial deterioration, and explores current solutions and future perspectives for cultural heritage preservation.

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic pollutants recorded in different environmental ecosystems, such as atmospheric, terrestrial, and aquatic ecosystems. These organic pollutants consist of only carbon and hydrogen atoms forming two or more conjugated benzene rings arranged in different manners, which are linear, angular, and cluster arrangements Fig. 1[1,2]. There are two classes of these PAHs based on molecular mass that are low molecular weight PAHs (LMW-PAHs) and high molecular weight PAHs (HMW-PAHs)[3]. LMW-PAHs consist of two or three fused aromatic rings. While HMW-PAHs consist of four, five, and/or six fused aromatic rings. LMW-PAHs are more bioavailable than HMW-PAHs because their smaller molecular masses penetrate biological tissues more easily than HMW-PAHs. Consequently, LMW-PAHs are considered more toxic than HMW-PAHs. However, HMW-PAHs can cause chronic pollution for a longer time than LMW-PAHs because of their high sorption capacity to the solid entities in the environment, such as fly ash particles in the atmosphere, soil particles, and sediments in water bodies[4,5]. In conclusion, PAHs are identified as persistent organic pollutants, especially HMW-PAHs, by the United States Environmental Protection Agency (U.S. EPA)[6]. Generally, PAHs are introduced into ecosystems as a result of the incomplete combustion of organic substances[7,8]. Such an incomplete combustion process occurs naturally or anthropogenically. The natural incomplete combustions occur in volcanic eruptions and forest fires[8]. The anthropogenic processes occur during the incomplete combustion of energy materials such as coal, oil, gas, or even organic waste to get energy[7,9,10]. Moreover, accidents during the extraction and transportation of energy materials such as petroleum oil are other potential anthropogenic pollution sources[10]. Petroleum oil consists of PAHs along with other organic and inorganic compounds. In the end, the released PAHs into ecosystems can be spread fast from polluted areas to others, causing secondary pollution. For instance, PAHs introduced into the air through incomplete combustion of energetic materials can spread with winds from polluted locations to non-polluted ones over long distances before being adhered to suspended particulates in the air or precipitated on surfaces of soil, water, and vegetation[7,11]. In addition, evaporation of the volatile PAHs (LMW-PAHs) occurs after crude petroleum spills onto waterbodies or soil surfaces, leading to air pollution surrounding the polluted locations[12]. Because of urbanization and the energy demand increase, the anthropogenic causes of PAH pollution have become more dominant pollution causes than natural ones.

Figure 1 presents the 3D stick model representations of the 16 PAHs designated as priority pollutants by the U.S. EPA[13]. The PAHs are categorized into LMW PAHs (up to three aromatic rings) and High Molecular Weight (HMW) PAHs (four or more aromatic rings). The chemical structures were sourced as SDF files from PubChem[14] and rendered using PyMOL[15] in 3D stick format, highlighting atom-specific color coding: hydrogen (white) and carbon (gray). Each PAH is labeled with its common name and PubChem CID for reference.

The Mediterranean Sea and ambient atmosphere can be considered sinks of anthropogenic PAH pollution since the 1960s due to human activities related to petroleum oil extraction and consumption. As evidence, Table 1 lists some huge spills of crude petroleum oil into the Mediterranean

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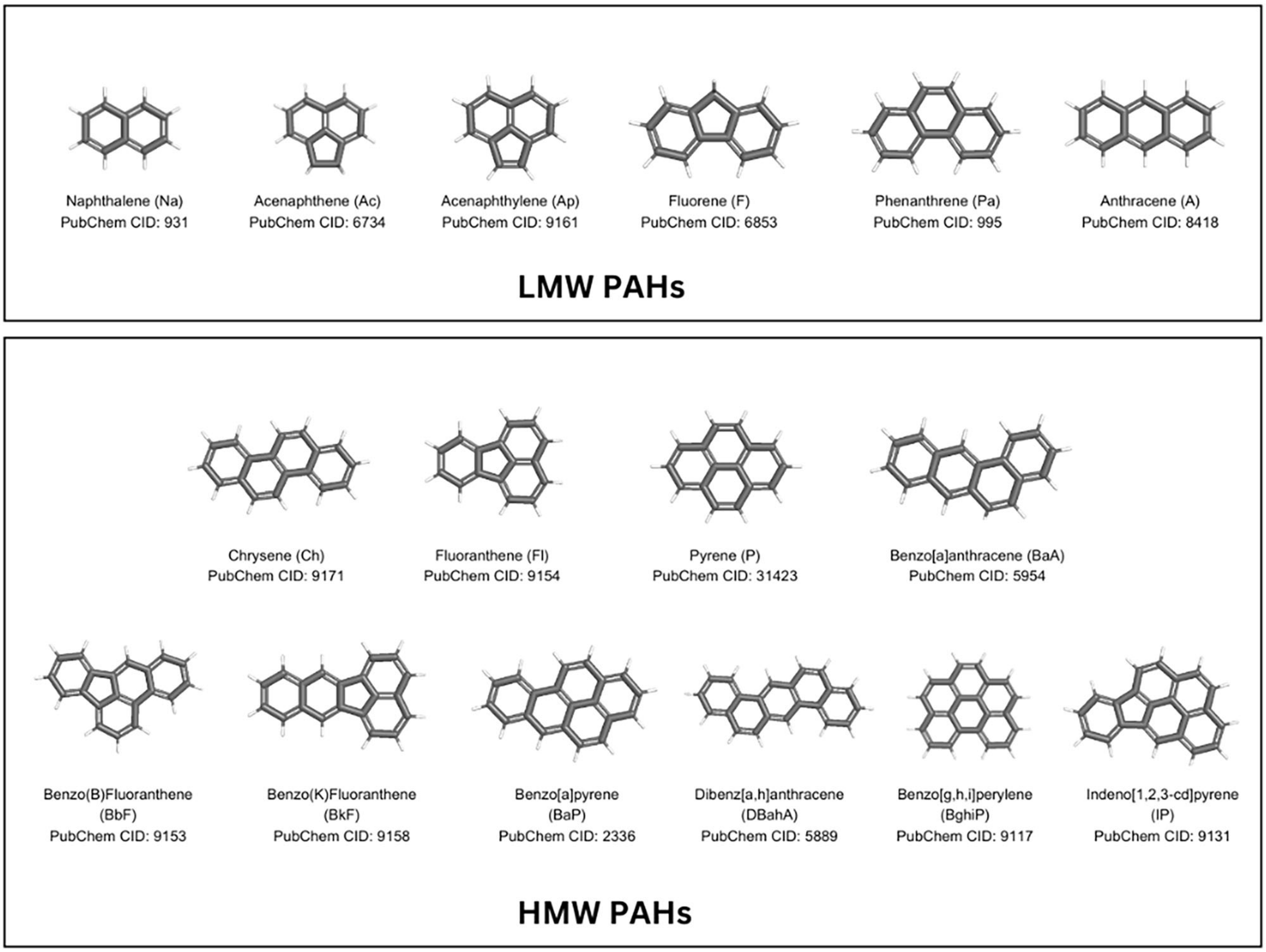

Fig. 1 | 3D Stick model representations of the 16 U.S. EPA priority pollutant polycyclic aromatic hydrocarbons (PAHs). The final layout and annotations were designed using www.canva.com.

Table 1 | Lists of catastrophic spills of massive amounts of crude petroleum oil, a source of PAHs, into the Mediterranean Sea in different Mediterranean cities from different countries

Spill	Location	Year	Spilled oil (tonnes)	Reference
Fina Norvege	Sardinia, Italy	1966	6000	116
Marlena	Sicily, Italy	1970	15,000	116
Ellen Conway	Port of Arzew, Algeria	1976	31,000	116
Al Dammam	Agioi Theodoroi, Greece	1976	15,000	116
URSS 1	Bosporus Strait, Turkey	1977	20,000	116
Kosmas M.	Asbas, Antalya, Turkey	1978	10,000	116
Messiniaki Frontis	Crete, Greece	1979	16,000	117
MT Independenţa ("Independence")	Istanbul City area and the Sea of Marmara, Turkey	1979	64,000	117
Juan Antonio Lavalleja	Port of Arzew, Algeria	1980	37,000	117
MV Haven	Genoa, Italy	1991	144,000	118,119
Castor off Nador	Morocco	2000	9900	120
Jiyeh power plant	coast of Lebanon	2006	15,000–30,000	116

Sea. Disastrously, there are more than 380,000 tonnes of crude petroleum oil spilled into the Mediterranean according to the listed disasters in Table 1 occurred between 1966 and 2006. Moreover, Table 2 lists many PAHs recorded in different environmental matrices (e.g., sediments, air, water) at different Mediterranean cities in different countries, resulting from oil exploration, extraction, or consumption. It is worth mentioning that PAHs' pollution can hinder the implementation of sustainable developmental goals (SDGs), especially in heavily polluted areas like the Mediterranean region. For instance, PAHs' pollutions have negative impacts on six SDGs, which are goal 2; zero hunger, goal 3; good health and well-being, goal 6; clean water and sanitation, goal 8; decent work and economic growth, goal 14; life below water and goal 15; life on land. In brief, PAH pollution threatens the food security issue as these toxic compounds are bioaccumulated within animals' and plants' tissues, leading to their death and hence food resource

Table 2 | Some locations in the Mediterranean region with polluted environmental matrices (air, water, sediment, and soil) with different levels of PAH pollution, indicating which PAH compounds were recorded in these locations

Location "Mediterranean region"	Environmental sample	ΣPAHs pollution level	PAHs compounds	Reference
Limassol, Cyprus	Air	0.12–4.02 ng/m^3	BbF, BeP, IcdP, Bghi, Ant, Cor	121
Alexandria-Manzallah, Egypt	surface sediments	13,156–34,852 ng/g	Σ_{16}PAHs	122
Traffic areas in Cairo, Egypt	Leaves of trees planted in the study areas	8,261–18,223 mg/kg	DahA, Bghi, BbF, Fluo, IcdP, Phe, BaP, Chry, Flu, Ace, BkF, BaA, Pyr, Ant, Acy, Nap	123
Alexandria, Egypt	Air	322.57–723.49 ng/m3	BkF, BaP, IcdP	124
Heraklion, Greek	Air	0.15–9.19 ng/m^3	BbF, BeP, IcdP, Bghi	121
Sicily Channel, Italy	surface seawater	43 ng/L	Flu, Phe, Fluo, Pyr, Chry, BaP	125
The Campania Region, Italy	surface water	10.1–567.23 ng/L	Acy, Ace, Flu, Phe, Ant, Fla, Pyr, BaA, and Chry, BbF, BkF, BaP, DahA, Bghi, IcdP	126
	suspended particulate matter in water	121.23–654.36 ng/L		
	sediment	331.75–871.96 ng/g		
Urretxu, Spain	Air	0.55–8.56 ng/m3	BaP, Nap, Phe, IcdP, Bghi	127
Azpeitia, Spain	Air	0.55–9.15 ng/m3	BaP, Nap, Phe, IcdP, Bghi	127
Oludeniz Lagoon, Turkey	surface sediments	1.85 mg/kg	Nap, Ace, Flu, Phe, Ant, Flu, Pyr, BaA, Chry, Bb/kF, BaP, IcdP, DahA, Bghi	128
Istanbul, Turkey	Air	85.6 ng/m^3	Acy, Ace, Flu, Phe, Ant, Fluo, Pyr, BaA, Chry, BbF, BkF, BaP, IcdP, DahA, Bghi	129
	Soil	684 mg/kg	Fluo, Phe, Pyr, Chry, BbF, BkF	

Ace acenaphthene, *Acy* acenaphthylene, *Ant* anthracene, *BaA* benzo[a]anthracene, *BaP* benzo[a]pyrene, *Bb/kF* benzo[b/k] fluoranthenes, *Bghi* benzo[g, h, i] perylene, *BbF* benzo[b]fluoranthene, *BkF* benzo[k]fluoranthene, *BeP* benzo[e]pyrene, *Chry* chrysene, *Cor* coronene, *DahA* dibenzo [a,h]anthracene, *Fluo* fluoranthene, *Flu* fluorene, *IcdP* indeno [1,2,3-cd]pyrene, *Nap* naphthalene, *Phe* phenanthrene, and *Pyr* pyrene.

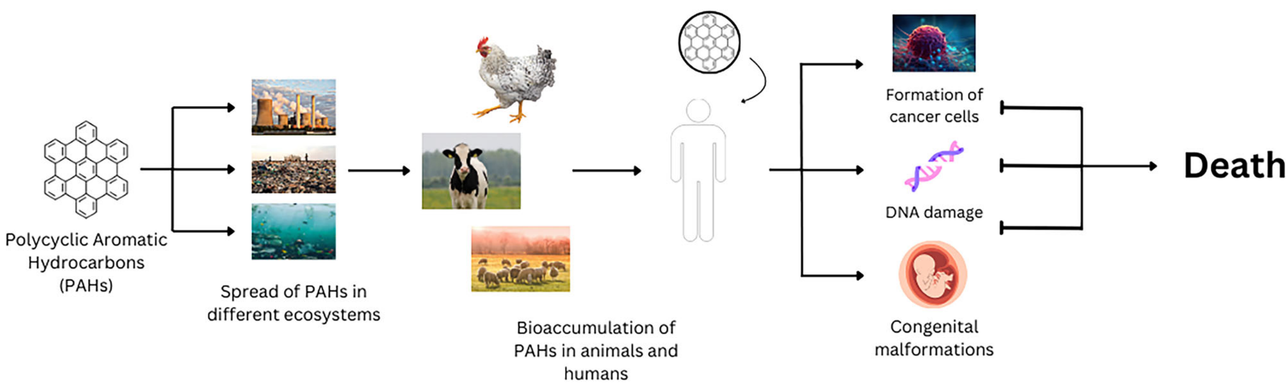

Fig. 2 | The polycyclic aromatic hydrocarbons (PAHs) distribution and life cycle. The diagram is designed with www.canva.com.

reductions[16,17]. Moreover, many dramatic health issues for humans arise due to the genotoxic[18], carcinogenic[19], and teratogenic[20] effects induced by exposure to PAH pollution Fig. 2. If such PAH pollution is not eliminated or even reduced, the polluted ecosystems lose many bioresources that are significant for human beings. In addition to bioresource loss, human beings lose their heritage cultures and hence, lose one of their economic sources as well. In this review, we discuss the deteriorative effects of PAH pollution on heritage cultures in terms of historical buildings and materials in the Mediterranean region. In addition, the possible deteriorative mechanisms induced by PAHs pollution, particularly PAH-microbial-induced deteriorations, applied solutions, and possible future aspects.

Distribution of PAHs in different environmental matrices, which are atmospheric, terrestrial, and aquatic ecosystems. Consequently, PAHs are bioaccumulated in the food chain starting from the edible plants, entering the livestock to invade human bodies. In addition to food chain bioaccumulation, PAHs can invade the human body directly through inhalation of PAH-polluted air. As a result, many dramatic effects take place on different levels, starting from genotoxicity, cytotoxicity, and tissue and organ malfunctions, which can lead to death.

Aim

This review thoroughly explores the pollution caused by PAHs and their negative effects on cultural sites in the Mediterranean. It examines the specific types of damage caused by PAHs, such as discoloration, material degradation, and destabilization of the structure of historical sites and artifacts. It has emphasized a number of ways through which PAH contamination in ancient sites can be reduced, such as the addition of a monitoring system to record the extent of PAH contamination, the use of bio-based and eco-friendly protective coatings, and the advancement of new bioremediation methods. It also points to important research gaps concerning the long-term impact of PAH exposure on ancient artifacts. These gaps are related to (i) the necessity of metagenomic studies on the microbiomes of various heritage materials is underscored to comprehend the impacts of PAHs and to formulate effective conservation strategies, and (ii) the importance of conducting metabolomic studies to explore the interactions between the microbiota of heritage materials and PAHs. This review hypothesized that there is a relationship between the concentration and type of PAHs and the behavior of the heritage material microbiota, which may lead to damage, a topic not extensively addressed in the literature.

Methodology

This review aims to identify the scientific gaps that were previously not tackled in existing literature regarding the PAH-driven microbial weathering of heritage monuments. To discover it, we searched with a single database of scientific literature, Google Scholar, with specific keywords: PAH, atmospheric PAH pollution, Mediterranean region, microbial corrosion of the archeological sites, multi-omics and PAH pollution, extracellular microbial metabolites and PAH, archeological sites, bio-restoration of heritage monuments, and archeological monuments. We deemed relevant literature reviews for every one of the keywords based on the title and abstract. The direction, along with the contents, of the reviews was selected through the screening of subtitles of all the collected reviews in order to define previously uncontested matters. The flowchart illustrates the main steps of the literature search and the corresponding results of relevant literatures Fig. 3. The search process for each keyword is divided into four stages: identification, screening, eligibility, and inclusion, through which the literatures are filtered from 253 results to 100 results finally.

PAH-induced deterioration of the historical monuments

Generally, the deterioration of a material is physical and chemical damage to its properties induced by physical, chemical, and/or biological factors. It is worth mentioning that air-borne pollutants in the Mediterranean region, such as PAHs, can deposit from air onto the historical surfaces' monuments, causing cultural and economic disasters due to the induction of physical and biological deteriorations.

Physical PAHs-induced deterioration of historical materials

Based on the PAHs' molecular weight emitted into the atmosphere, LMW-PAHs are emitted as a gaseous phase while HMW-PAHs are emitted as a particulate form[21]. Both PAH types have a high tendency, particularly HMW-PAHs, to be deposited or adsorbed on the building surfaces rather than being suspended in the atmosphere due to their high hydrophobicity[22]. These deposited PAHs are considered precursors of black-colored material known as soot[23]. Knowing that such soot in turn combines with dust, forming a black crust which is one of the most significant threats to the architectural heritage[24]. So, many studies analyzed the chemical composition, especially the PAHs composition profile, and the deteriorative effects of black crusts on many historical buildings' surfaces.

For example, Al-Quds mosque, located in the Roches Noires, Casablanca, Morocco, has historical value since the early 20th century[25]. Ozga et al. collected the black crusts from the surface of Al-Quds Mosque and analyzed them by attenuated total reflection/Fourier-transform infrared spectroscopies. They found that the most dominant components of such black crusts on Al-Quds mosque are automobile exhausts containing PAHs. Similarly, in Italy, PAHs are detected in the black crusts collected from the Monumental Cemetery of Milan and other historical buildings of Palermo (Italy)[26] and from the stones of the temples of Agrigento (Italy)[27]. Such studies also correlated the atmospheric concentration of PAHs and their deposition proportions into the black crusts of these historical buildings. They found that the composition profile of PAHs observed in these black layers/crusts of the historical buildings exhibited a resemblance to those analyzed in the polluted ambient air[28]. Consequently, these findings substantiate the assertion that air pollution with PAHs exerts a noticeable influence on the deterioration of historical buildings' materials through black crust accumulation[28]. Additionally, some studies focus on examining the harmful impacts of PAH pollution within the indoor environments of heritage museum buildings, such as a heritage building built at the end of the 19th century, situated in the Municipality of Beius, Bihor County, Romania[29].

Microbial PAHs-induced deterioration of the historical materials

In addition to being precursors for soot and black crust formation, PAHs are carbon sources for many microorganisms as well[30]. However, PAHs are still toxic, particularly oxy-PAHs resulting from PAHs degradation[31], to other microorganisms[32,33]. This means that PAHs in the black crusts on the historical buildings have effects on the community composition and the metabolic activities of microflora inhabiting these surfaces, leading to microbial-induced materials' deterioration. Knowing that the microbial-induced deterioration of materials is considered the most severe biological deterioration type. Since the microorganisms are characterized by resilience under extreme environmental conditions, a high reproduction rate leads to the acceleration of the material destruction, production of a wide range of different extracellular metabolites, and hydrolytic enzymes destroying different surface types such as stones, concrete, metallics, paintings, ceramics, mummies, and books[34,35]. Therefore, we summarize a group of potential material-destroying microbial factors which are (i) the microbial biofilm, (ii) extracellular metabolites (organic, inorganic, and salt metabolites), and (iii) extracellular enzymes and their production patterns' relations with PAHs pollution.

Deteriorative effects of the microbial biofilms

Generally, the microbial biofilm consists of an extracellular polymer matrix (EPM) in which the microbial cells and their extracellular metabolites and enzymes are embedded and secreted, respectively. EPM is responsible for historical material deterioration through direct and indirect mechanisms. The direct deteriorating effects of EPM are due to its mechanical stress on the stone minerals through its shrinking and swelling cycles inside the pores and cracks of the buildings[36,37]. Meanwhile, EPM indirectly deteriorates the historical monuments by sticking microbiota onto such surfaces, enhancing its expansion on surfaces and protection against unfavorable conditions[38,39]. Furthermore, EPM acts as a reactive interface in which the corrosive metabolites and enzymes are secreted from the surface-stuck microbiota, facilitating their deteriorating effects on heritage materials such as bio-leaching, dissolution, blistering, scaling, and decaying[40].

Moreover, there are many microbial metabolites secreted into EPM, causing heritage materials' deterioration. According to our knowledge, these corrosive extracellular metabolites can be classified into three major groups, which are organic and inorganic substances and salts consisting of organic and inorganic ions as listed in Table 3 and Table 4, respectively.

Deteriorative effects of the extracellular microbial organic metabolites. The extracellular organic microbial metabolites are further classified into organic acids and pigments. Many microorganisms, fungi, lichens, bacteria, and archaea, secrete many organic acids on the historical materials' surfaces of limestone, stone frescoes, and mural paintings such as acetic acid[35], citric acid[35], constitic acid[41], fumaric acid[35], lactic acid[35], malic acid[35], oxalic acid[35,41,42], stictic acid[41], and succinic acid[35] Table 3.

Such secreted organic acids have deteriorative effects on historical materials. As evidence, there are many identified fungal, bacterial, and lichen genera causing deterioration of different types of historical materials via secretion of different organic acids such as *Acremonium sp., Aspergillus sp., Fusarium sp., Penicillium sp., Trichoderma sp., Rubrobacter sp., Streptomyces sp., Dirina sp.*, and *Lepraria sp* Table 3. The organic acids-based deterioration mechanism of historical materials is known as the leaching process. The leaching process involves metals' dissolution that is involved in the materials' chemical structures, then free metal-ions precipitation forming non-soluble complexes onto materials' surfaces, causing scaling, blistering, and flaking of the surfaces[43]. For instance, oxalic acid dissolves calcium ions of calcium carbonate in limestones, producing calcium oxalate precipitate covering the surfaces of stones, forming crusts[44]. Some heterotrophic organisms flourish in chemical microenvironments (consisting of hydrocarbons, metals, and carboxylic acids), potentially influencing the distribution of chemical species themselves, as exemplified by Clostridium sp. in low-sulfate samples[45] since it uses sulfates as final electron acceptors under anaerobic conditions, reducing sulfate to sulfide[46].

Bio pigments secreted from some microorganisms participate in the microbial deterioration of the historical monuments. As evidence, microorganisms colonizing the stone monuments' surfaces cause esthetically

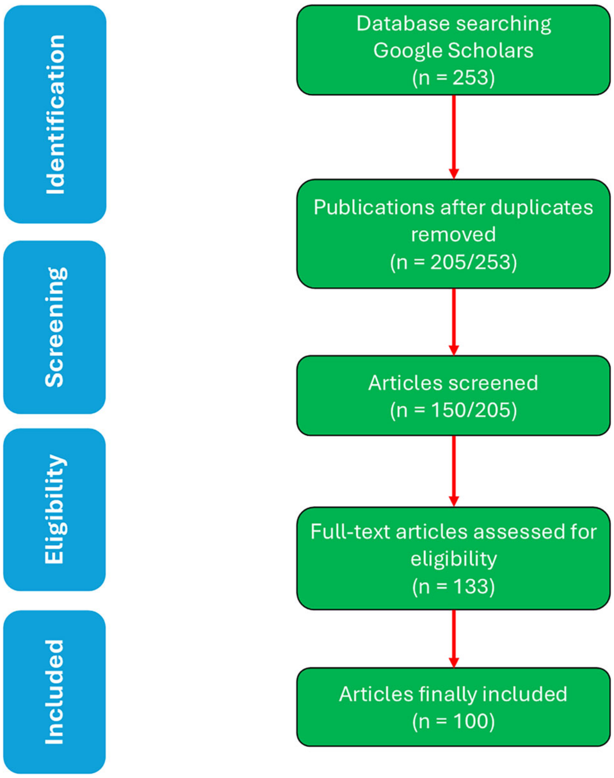

Fig. 3 | The flowchart indicates the main stages of literature collections and selections and the corresponding results of relevant literature. The stages are identification, in which all literature containing a keyword is collected from databases such as Google Scholar; screening, in which the duplicated literature is removed; eligibility, in which the collected literature is further filtered based on the review aim; and inclusion, which is the last stage and aims to select the most suitable literature to be involved in the review. The symbol (*n*) indicates the number of literature at each stage, illustrating the filtration process to get the most suitable literature for the review.

unappealing staining using biogenic pigments[47,48]. Moreover, as listed in Table 4, Microbially influenced staining is due to photosynthetic pigments such as carotenoids and Salinxanthin, melanin and melanoidin-based pigments produced by some fungi and pigmented bacteria such as *Rubrobacter sp*. and *Streptomyces sp*[49,50].

Deteriorative effects of the extracellular microbial inorganic metabolites and salts

Chemoautotrophic microorganisms play a key role in the monumental materials' deterioration through the secretion of inorganic acids and salts, which have chemical deteriorative effects on the historical materials as listed in Table 4. According to many previous studies[51,52], the deteriorative mechanism of inorganic acids such as nitrous acid, nitric acid, sulfurous acid, and sulfuric acid is through the formation of water-soluble nitrate and sulfate salts. Consequently, these salts interact with the acid-sensitive components of the stone materials[53,54].

For instance, the nitrifying archaea and bacteria are involved in the nitrogen cycle by oxidation of ammonia gas (NH_3) and nitrogenous oxides pollutants in the atmosphere, yielding nitrous and nitric acids. As a result, there are significant amounts of such corrosive inorganic acids deposited on the monuments' surfaces and hence materials' deterioration, especially stones[55,56]. *Nitrobacter sp*., *Nitrosomonas sp*., *Nitrosospira sp*., and *Nitrosovibrio sp*. are among the nitrifying bacteria that deteriorate historical materials such as stones, especially limestones[41]. Similarly, the Sulfur-oxidizing bacteria such as *Acidiphilium sp*[41]., and *Sulfurovum sp*[41]. oxidize Sulfur-containing aerosol pollutants such as hydrogen sulfide gas (H_2S) or elemental Sulfur, producing extremely corrosive sulfurous or sulfuric acids, forming harmful black crusts and calcium sulfate or gypsum[54,57–59].

Deteriorative effects of the extracellular microbial enzymes. In addition to the metabolite-induced microbial deterioration of the historical materials, there is another microbial deterioration tool, the extracellular enzymes. Extracellular enzymes play a crucial role for microbes by enabling them to interact with their environment effectively. Since the production of extracellular enzymes by microbes is a vital process that aids in the breakdown of complex organic compounds like cellulose, lignin, and chitin, as well as xenobiotics, including PAHs[60,61]. Therefore, the secretion of extracellular enzymes by microbes serves as a key strategy for nutrient acquisition, adaptation to diverse environments, and survival. Based on the substrate types, there are many classes of secreted microbial enzymes into the microbial biofilm, such as amylases[50], cellulases[50,62,63], collagenase[64], endoglucanase[64], exoglucanase[64], gelatinases[50], β-glucosidase[64], and proteases[62,64]. These extracellular enzymes have deteriorative effects on different historical materials such as paper/books and old manuscripts, ancient textiles, wooden coffins, and audio-visual materials Table 5. These enzymes are released by many bacterial and fungal genera. For instance, *Bacillus sp., Pseudomonas sp., Staphylococcus sp., Arthrobacter sp., Flavobacterium sp*., and *Streptomyces sp*. are isolated from deteriorated monumental materials such as papers, textiles, and wooden coffins.

Solutions for PAHs-induced deterioration of the archeological sites

PAH-induced deterioration of the archeological sites and historical materials is a complicated socio-environmental issue. Therefore, it is recommended to integrate a multifaceted approach to mitigate the deteriorative effects of urbanization and increasing rates of fossil fuel consumption, which increase PAH emissions. In this review, we propose four integrated strategies to address this issue: (i) implementing monitoring systems to estimate atmospheric PAH pollution levels, (ii) exploring the relationship between atmospheric PAH pollution and 'omics' insights into the microflora on historical materials, (iii) developing bio-based, eco-friendly materials to restore historical surfaces, and (iv) improving atmospheric PAH remediation techniques.

Establishment of the monitoring system for PAH pollution estimation

The first step towards effective PAH pollution removal is to identify the types of pollutants and their level or concentrations. Hence, the most proper treatment methods can be selected to remove or at least decrease the pollution level and consequently decrease its negative impact. Moreover, the determination of pollution type and level is mandatory to evaluate the efficiency of the treatment method.

Many analytical techniques have evolved to determine the environmental pollution with PAHs. Examples of such techniques are Chromatographic techniques, such as high-performance liquid chromatography in conjunction with fluorometric probes, photodiode-array, ultraviolet or mass spectroscopy detector, gas chromatography in conjunction with flame ionization or mass spectroscopy detector, and superficial fluid chromatography in conjunction with ultraviolet or mass spectroscopy detector, are among the conventional methods of detecting these compounds[65]. Fluorescence spectroscopy is a valuable technique for the detection of PAH pollution[66,67]. The technique utilizes the fluorescent behavior of PAHs, resulting in luminescence upon excitation by characteristic wavelengths[68]. The technique has several advantages in the sense that it is (i) very sensitive and selective to trace amounts of various PAHs in air samples; (ii) real-time measurable levels of PAHs; and (iii) non-destructive and rapid analytical operation[69,70].

However, all these previously mentioned analytical methods are considered sophisticated ones. Many concerns should be considered during the analysis of these techniques. One of the major obstacles to PAH analysis by chemical methods is sample preparation. The sample preparation is a major obstacle in the chemical analysis of PAHs due to the volatility and lipophilicity of these compounds, which can significantly change the sample

Table 3 | List of organic acids, depsides, pigments, and metal chelators secreted by microorganisms causing deterioration of many types of historical materials in many countries

A. Organic acids				
Organic substance	**Microorganism**	**Archeological monument**	**Material**	**Reference**
Acetic acid	*Fusarium solani Penicillium oxalicum*	Buak Krok Luang and Kham Temples, Thailand	Mural paintings	35
Citric acid	*Aspergillus aculeatinus* *Aspergillus fumigatus* *Aspergillus niger* *Aspergillus piperis* *Penicillium citrinum* *Penicillium hetheringtonii* *Penicillium oxalicum* *Trichoderma aethiopicum* *Trichoderma harzianum*	Buak Krok Luang and Kham Temples, Thailand	Mural paintings	35
Constictic acid	*Lepraria lobificans*	Southern Spain	Mortars	41
Fumaric acid	*Aspergillus niger* *Aspergillus piperis* *Penicillium hetheringtonii* *Penicillium oxalicum* *Trichoderma aethiopicum*	Buak Krok Luang and Kham Temples, Thailand	Mural paintings	35
Lactic acid	*Aspergillus aculeatinus* *Aspergillus fumigatus* *Aspergillus niger* *Aspergillus piperis* *Fusarium solani* *Penicillium citrinum* *Penicillium oxalicum* *Trichoderma aethiopicum* *Trichoderma longibrachiatum*	Buak Krok Luang and Kham Temples, Thailand	Mural paintings	35
Malic acid	*Aspergillus piperis* *Fusarium solani* *Penicillium citrinum* *Trichoderma aethiopicum* *Trichoderma longibrachiatum*	Buak Krok Luang and Kham Temples, Thailand	Mural paintings	35
Oxalic acid	*Acremonium sp.* *Aspergillus niger* *Aspergillus piperis* *Dirina massiliensis* *Penicillium sp.* *Penicillium oxalicum*	Basarabi Churches Ensamble, Romania The house of the ancient hunt, Italy Buak Krok Luang and Kham Temples, Thailand The house of the ancient hunt, Italy Buak Krok Luang and Kham Temples, Thailand	Limestone Stones and frescoes Mural paintings Stones and frescoes Mural paintings	35,41,49
Stictic acid	*Lepraria lobificans*	Southern Spain	Mortars	41
Succinic acid	*Aspergillus aculeatinus* *Aspergillus fumigatus* *Aspergillus niger* *Aspergillus piperis* *Penicillium oxalicum*	Buak Krok Luang and Kham Temples, Thailand	Mural paintings	35
B. Pigments				
Carotenoids	*Rubrobacter sp.* *Rubrobacter radiotoleran* *Rubrobacter taiwanensis* *Rubrobacter xylanophilus*	Europe	Murals and stones	50
Melanin	*Streptomyces sp.* *Streptomyces rochei*	Giza and Luxor, Egypt	Tomb paintings and limestone	62
Salinxanthin	Salt-tolerant bacteria and archaea	Europe	Stones	50

compositions from collection to analysis. In addition to the sample preparation concern, prior experience with the technique and data analysis, calibration problems with the accurate quantification of multi-component mixtures, their high cost to employ for routine monitoring, and the in-situ applicability of such techniques are still other significant obstacles[71].

So, it is mandatory to establish other alternatives for PAH analysis that overcome or fill the technical gaps when using chemical analysis. Such alternatives are biosensors that are based on biological concepts[72]. In the biosensors, there are two main parts which are (i) the sensor which consists of the biological agent responsible for sensing the target analyte (PAHs in this review) in the sample, and (ii) the signal transducer that is responsible for converting the signal pulse from the sensor to be readable data to know the type and amount of the sensed PAH molecules. Regarding the bio-based sensor, there are many different biological agents exploited in such an approach, such as proteins (e.g., proteins through immunoassays) and nucleic acids (e.g., Deoxyribonucleic acid (DNA)[72] through aptamer construction). The signal transduction component can be either optical, electrochemical, colorimetric, or piezoelectric transduction methods.

Omics and PAHs-induced deteriorations of historical materials

Although there are many previous studies illustrating the deteriorative effects of the microbial extracellular metabolites and enzymes, as in Tables 3,4 and 5, it is still unclear what the relation between PAH pollution and these metabolites' and enzymes' production is. Therefore, it is essential to

Table 4 | List of inorganic acids and salts secreted by microorganisms causing deterioration of different types of historical materials in different countries

A. Inorganic acids				
Inorganic substance	**Microorganism**	**Archeological monument/country**	**Material**	**Referen-ce**
Nitric acid	*Nitrobacter sp*. *Nitrosomonas sp*. *Nitrosospira sp*. *Nitrosovibrio sp*.	LeiZhou Stone Dog, Leizhou Peninsula, China	Limestones and stones	41
Nitrous acid	*Nitrobacter sp*. *Nitrosomonas sp*. *Nitrosospira sp*. *Nitrosovibrio sp*.	LeiZhou Stone Dog, Leizhou Peninsula, China	Stones	41
Sulfuric acid	*Acidiphilium sp*. *Sulfurovum sp*.	LeiZhou Stone Dog, Leizhou Peninsula, China	Basalt sculptures and stones	41
Sulfurous acid	*Acidiphilium sp*. *Sulfurovum sp*.	LeiZhou Stone Dog, Leizhou Peninsula, China	Basalt sculptures and stones	41
B. Salts				
Calcium oxalate	*Dirina massiliensis*	Sicily and Mainland, Italy	Dolomitic rocks	41
Nitrate anion	*Caloplaca xantholyta*	The Crypt of the Original Sin, Italy	Paintings	49

Table 5 | List of extracellular microbial hydrolytic enzymes involved in the biological deterioration of historical materials

Enzyme	**Microorganism**	**Archeological monument/country**	**Material**	**Reference**
Amylases	*Aspergillus flavus*	Indonesia	Paper/book and old manuscripts	50
Cellulases	*Aspergillus flavus* *Aspergillus niger* *Bacillus sp*. *Paecilomyces canoes* *Paecilomyces carneus*	Indonesia Egyptian Textile Museum - Egyptian Textile Museum The Egyptian Museum	paper/book and old manuscripts Ancient textiles Foxed paper documents Ancient textiles Wooden coffins	50,62,62,63
Collagenase	*Bacillus sp*. *Psuedomonas sp*. *Staphylococcus sp*.	-	Parchment	64
Endoglucanase	*Aspergillus sp*. *Cladosporium sp*. *Eurotium sp*. *Penicillium sp*.	-	Papers	64
Exoglucanase	*Aspergillus sp*. *Cladosporium sp*. *Eurotium sp*. *Penicillium sp*.	-	Papers	64
Gelatinases	*Aspergillus flavus*	Indonesia	paper/book and old manuscripts	50
β-glucosidase	*Aspergillus sp*. *Cladosporium sp*. *Eurotium sp*. *Penicillium sp*.	-	Papers	64
Proteases	*Arthrobacter sp*. *Aspergillus flavus* *Aspergillus niger* *Bacillus sp*. *Conidiobolus sp*. *Flavobacterium sp*. *Neurospora sp*. *Paecilomyces canoes* *Paecilomyces carneus* *Streptomyces sp*.	- Egyptian Textile Museum Egyptian Textile Museum - - - - The Egyptian Museum Egyptian Textile Museum- 	Audio-visual materials Ancient textiles Ancient textiles Audio-visual materials Audio-visual materials Audio-visual materials Audio-visual materials Wooden coffins Ancient textiles Audio-visual materials	62,62,64,64,64

understand such mysterious relationships. "Omics" can be considered key techniques that may explain many mysterious biological phenomena at the molecular level. Since various "Omics" approaches are employed to study environmental issues, such as the deterioration of heritage materials. For instance, metagenomics is utilized to identify microbial communities present in heritage materials, enabling researchers to understand their role in biodeterioration[73]. In a recent study by Yanyu Li et al., they utilized metagenomics to investigate the microbial diversity in the door walls of the Ji family's residential houses, as well as their biological functions and chemical cycles[74]. In addition to metagenomics, other approaches such as proteomics, metabolomics, and transcriptomics are employed to gain a comprehensive understanding of biodeterioration processes on heritage materials. Moreover, many studies employ a "multi-Omics" approach to determine the causes and mechanisms of biodeterioration, with the ultimate goal of identifying appropriate solutions. For example, in 2018 Roldán et al. conducted metabolomic and proteomic analyses in the Iberian Mediterranean

Basin to describe the bacterial communities colonizing rocks and identify the presence of organic binders within them[75]. The multi-omics contributes significantly in determination of the environmental factors inducing the microbial outbreak on the ancient wall paintings of the Maijishan Grottoes, China[76], the hypogeum of the Basilica di San Nicola at the Carcere Church in Rome[77], identification of fungal Communities associated with the biodeterioration of waterlogged archeological wood in a Han Dynasty Tomb, China[78].These studies demonstrate the power of "Omics" approaches to be utilized in the investigation of the heritage materials biodeterioration, providing valuable insights into the microbial communities involved, the deterioration mechanisms, and potential solutions to address this economic and cultural issue. However, according to our knowledge, there is no previous study focusing on "Omics" to explain the possible relation between PAHs pollution and the deteriorative microbial metabolites and enzyme production.

Bio-based and eco-friendly restoration materials for deteriorated historical surfaces

The restoration of historical artifacts is crucial for their integrity and longevity, saving their economic and cultural values. According to the principle of the restoration methods, there are physical, chemical, and biological restoration methods. The selection among these diverse methods is based on the geographical location, atmospheric conditions, and historical material[79].

Physical methods include different techniques such as (i) irradiation using gamma (γ)-, ultraviolet (UV)- and laser irradiation, (ii) changing temperatures, applying very low temperatures as freezing, refrigeration, and low-temperature plasma disinfection or very high temperatures[80]. The sacrificial anodes and liquid nitrogen (−196 °C) have been widely employed as physical restoration methods in the past to conserve historic iron artifacts on the seabed[81], and ultrashort (in picoseconds) pulse lasers to clean historic artifacts such as the 19th-century military gold braid[82]. Chemical methods typically involve the use of chemicals, especially inorganic nanomaterials, and nanoparticle suspensions[83–86]. Add to this, some commonly used chemicals in the historical artifacts' treatment include distilled water, alkoxysilanes, phenols, amorphous silica, hydrogen peroxide, and organosilanes[87,88]. Physical and chemical preservation methods have shown effectiveness over the years, but they have several limitations regarding the environment and human health. The accumulation of chemicals and irradiation exposure pose threats to the workers owing to their toxicity, in addition to the possibility of the corrosion of the treated surfaces over time[87,89–91]. On the other hand, the biological restoration methods are characterized by being eco-friendly, relatively cheaper, and non-toxic without causing further material deterioration compared with the physical and chemical treatments[92–94].

In bio-restoration methods, either the organism or its metabolites can be exploited to preserve the deteriorated historical materials. For instance, the plants participate significantly in bio restoration through their secondary metabolites and application on deteriorated surfaces, such as biocides and biosurfactants. Biocides are used to reduce or eradicate damaging microorganisms like fungi and bacteria by preventing microbial growth and hence stopping the biodeterioration of the historical heritage[95]. The most applied plant-based biocides in this aspect are essential oils, flavonoids, phenolics, and anthocyanins[96]. On the other hand, biosurfactants such as saponin, sophorolipid, and betaine are applied to remove the unwanted corrosive microbial metabolites or compounds causing surface deterioration, such as sulfates and nitrates, from stone surfaces[97]. To enhance the biosurfactants' effects, these biosurfactants are usually combined with certain extracellular microbial enzymes (EMEs). Although EMEs are causing historical material deterioration, others have important preservative roles[98]. These enzymes have also been used because of their high selectivity and effective removal of undesired layers without damaging the artifact material. For example, glucose oxidase can remove biofilms from stone without harming the surface itself. Chitinase enzymes can reduce fungal and bacterial growth on woody materials, and hence remove deteriorating biofilm without damaging the artifact[99]. Although many microorganisms induce the microbial deterioration of historical heritage materials, there are other organisms that cause the consolidation of such valuable materials. As evidence, bacterial species, e.g., *Pseudomonas, Pantoea, and Cupriavidus* species, induce calcium carbonate precipitation, which can help to strengthen and protect the stone material[100].

Improvement of atmospheric PAH remediation techniques

Urbanization has led to the accumulation of PAHs in ecosystems, which are remediated through natural attenuation processes[101,102]. However, these processes have long-term effects on effectively restoring polluted ecosystems and can lead to contaminant dispersion in other ecosystems. High PAH pollution levels in the atmosphere resulting from crude petroleum oil spills pose health and economic issues, necessitating the development of fast and cost-effective clean-up technologies.

Several established remediation techniques have recently been developed to clean up many contaminants, including PAHs. Remediation technologies are categorized into physical, chemical, and biological types based on scientific principles[103]. Physical and chemical treatments are traditional remediation technologies. Unfortunately, these traditional cleanup technologies have many drawbacks, such as high implementation costs and environmental risks. Implementing these approaches is highly energy-intensive and environmentally detrimental because of the secondary pollution generated while remediating initially contaminated sites[104,105]. Therefore, it is imperative to develop eco-friendly, sustainable, and cost-effective restoration solutions, such as biological remediation technologies that have become increasingly favored over traditional methods for environmental cleanup[106].

The removal of volatile organic contaminants (VOCs), including PAHs, from the atmosphere is a major air pollution concern[107]. Biofiltration is one of the remediation technologies for polluted airstreams with VOCs. Biofiltration is based on the ability of microorganisms, bacteria and fungi, to degrade a wide spectrum of organic pollutants in an economic and eco-friendly manner[108–110]. In a biofilter, polluted air passes through porous media such as peat, soil, bark, compost, wood chips, or polystyrene spheres[111–114], resulting in the entrapment of pollutants within the porous media. These pollutants are subsequently oxidized or transformed into biomass through the activity of microorganisms that are pre-immobilized on the packing material[110].

The efficiency of atmospheric remediation from VOCs using biofilters depends on many factors, including the microbial consortia, biofilter design, operational parameters, and pollutant characteristics. The microbial composition significantly affects the biofilter performance. Fungi have the potential capacity to remove PAHs, especially those with higher molecular mass, such as benzo[α]pyrene, while bacteria have higher degrading activities for lower molecular weights of PAHs, such as phenanthrene[115]. The choice of packing material, one of the biofilter design parameters, is crucial for the performance of biofilters, affecting microbial growth, pollutant removal, and costs. Particle size and material type (organic or inorganic) influence the mechanical properties, especially when dealing with high air flow rates. Porous materials with rough surfaces are beneficial as they provide more space for microorganisms to grow and improve pollutant transfer[115]. The moisture content, pH, and temperature of the packing material must be optimized to decontaminate the polluted air better. The filter beds should contain adequate moisture content, maintaining the decontaminating activities of the fixed microbes without causing anaerobic conditions due to over-moisture or drying due to less moisture content[115]. Also, pH and temperature are critical for the optimization of the microbial enzymatic activities responsible for PAH degradation[115].

Conclusion

The conclusion presents the main points, emphasizing the danger that PAHs pose to the cultural heritage of the Mediterranean, especially for historical buildings and materials. The principal findings are as follows: (1)

PAHs directly induce soot and black crust formation, altering the appearance of historical buildings; (2) microbial communities acting on heritage materials enhance deterioration through metabolite secretion; and (3) there is a potential relationship between PAH levels and microbial metabolic activity, indicating a sophisticated deterioration mechanism. To overcome these challenges, we suggest: (1) establishment of advanced PAH monitoring systems; (2) application of omics-based technologies for root cause analysis; and (3) designing bio-based systems or compounds for PAH sequestration and mitigation. These findings and proposed solutions pave the way for more efficient preservation methods, emphasizing the need for a multidisciplinary approach to safeguard the Mediterranean's priceless cultural heritage.

Future perspectives

In this part, we address scientific gaps for interested scientists to fill in future research work. According to our work, we investigated the gap in research in archeological microbiology and toxicology, particularly, that is related to PAHs' pollution. We asked ourselves many questions without answers in the previous work according to our research, as follows:

- What is the impact of PAHs on the alpha and beta diversity of microbes in archeological sites? And is this impact different on biotic material from abiotic?

  After investigating the concentrations of PAHs in the air of different archeological sites and their impact on microbial diversity, we can find solutions to conserve these materials by microbiome engineering techniques. Moreover, we can reverse the effects of PAHs by biochemical engineering techniques. Therefore, we need to conduct a metagenomic study on different heritage material microbiomes.
- Does microbial diversity affect the deterioration of heritage materials, or are the PAHs the effectors?

  We did not find any previous work investigating the impact of metabolites secreted from heritage materials microbiota on PAH degradation. Therefore, we need to carry out a metabolomic study to investigate the effect of these microbes on PAHs.
- Is there a relationship between levels of PAHs and the corrosive microbial metabolites on heritage materials?

We claim that there is a relationship between the concentration and type of PAHs and the behavior of the microbiota of heritage materials, causing this damage. Some of these metabolites are also secreted from the microbiota of heritage materials, such as oxalic acid, malic acid, citric acid, and acetic acid. For future studies, a certain workflow is recommended to answer these questions. The workflow is summarized in designing the following stages; (1) a systematic research to quantify polycyclic aromatic hydrocarbon (PAH) concentrations in various archeological sites with standardized sampling techniques and analytical protocols, (2) in situ experiments in archeological sites for long-term monitoring of the microbial diversity by metagenomic analysis in relation to PAH concentrations, (3) strategies to determine the impact of PAH pollution on microbial metabolites responsible for material degradation, and (4) inoculation of possible probiotic microorganisms capable of mitigating PAH-induced deterioration in heritage materials.

Data availability

All data or results mentioned in this review are available and retrieved from online available literature.

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Conceptualization, A.A.M. Writing—original draft preparation, M.T.Y., F.O.A., J.M., N.G.A., N.M.E. Writing—review and editing, R.M.H. and A.A.M. Supervision, R.M.H. and A.A.M. All authors have read and agreed to the submitted version of the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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