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Climate change and air pollution impacts on cultural heritage building materials in Europe and Mexico

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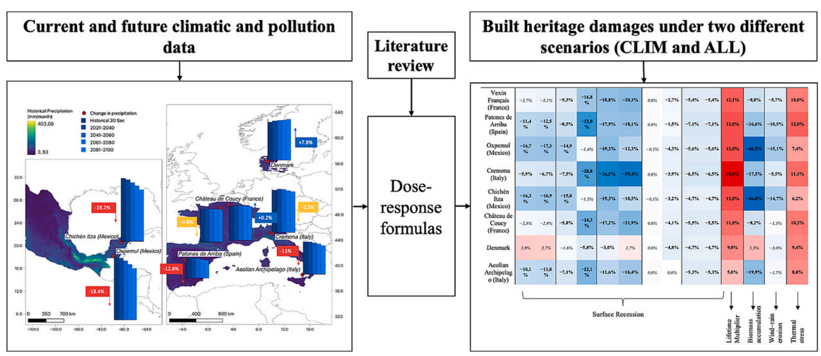
HIGHLIGHTS

- Application of dose-response functions for environmental data was performed.
- Damage decrease is projected due to future climate and pollution changes.
- Surface recession changes vary up to a 40 % difference across equations.
- Climate-induced damage was found to be predominant over the pollution-induced one.
- More campaigns are needed to determine real damage in different climate locations.

GRAPHICAL ABSTRACT



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ABSTRACT

Climate and air pollution have adverse effects on cultural heritage building materials. However, the quantified damage due to modeled changes in climate and air pollution is still poorly studied. Here, we review first the damage affecting these materials and the associated damage equations in the literature. Across all relevant studies (n = 87), we found only nine independent equations to estimate different damage categories, mainly limited to limestones. Then, by using current meteorological data and future bias-corrected CMIP6 climate and air pollution data at high resolution (1 km; historical and business-as-usual scenario) and applying these equations, we quantified the relative contributions of climate and air pollution changes on the building materials of eight cultural heritage sites of the European project Sustainable COnservation and REstoration of built cultural heritage (SCORE) from 2020 to 2100. On average across the sites, a significant decrease in damage is projected in surface recession (−10 % ± 10 %), biomass accumulation (−20 % ± 18 %), and wind–rain erosion (−7 % ± 6 %) in response to future climate and air pollution changes, except in the regions where precipitation substantially increases (Northern Europe). A large uncertainty in the relative magnitude of the damage to built cultural heritage materials was found for the same site, changes in surface recession vary up to a 40 % difference across the equations. Moreover, thermal expansion and lifetime multiplier equations project an increase in the related damage while all the other types of damage are significantly reduced. Finally, in general, but not systematically, climate-induced damage was found to be predominant over the pollution-induced one. Our results allow

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prioritizing cultural heritage maintenance decisions in regions where damage will further increase. Beyond simulated damages which are still limited use, we urge more campaign studies to determine real in situ damage in different climate locations to validate or build the best equations.

1. Introduction

The consequences of climate change have been reflected in different socioecosystems worldwide (Riahi et al., 2011). One of the poorly studied ones is outdoor built cultural heritage, which is directly exposed to atmospheric conditions and vulnerable to climatic variability. According to the latest IPCC report, many changes in the climate system are becoming more extensive in direct relation to global warming, and this is reflected in the increase, for example, in the frequency and intensity of hot extremes, heavy precipitation, and other impacts on the building systems (IPCC, 2023). In addition to these impacts, cultural losses affect adaptive capacities, and in this sense, cultural practices related to nature and cultural expressions associated with the construction of homes, communities, populations, and the use of natural and resistant materials are irrevocably lost.

In the 2021 IPCC-WGII report, there are only two general statements on cultural heritage, particularly the fact that "studies have described (…) climate services in support of cultural heritage" (IPCC, 2021). But in the 2022 IPCC-WGIII report, there are hundreds of occurrences of cultural heritage but none on built materials nor quantitative, for instance (IPCC, 2022), "Climate change destroys unique natural archives and important cultural heritage site," due to "flooding and sea level rise" and "erosion and saltwater." The authors point out that "More research is needed, however, particularly on cultural heritage, spiritually significant places, and in low-income countries" and "European cultural heritage in general and world heritage sites specifically lack adaptation strategies to preserve key cultural assets." (Masson Delmotte et al., 2021). This highlights the particular need for research on the impact of climate change on built cultural heritage, particularly in the context of increased extreme climate phenomena (Orr et al., 2021). Global concern has recently grown for protection strategies against climatic events that can deteriorate or destroy this heritage (UNESCO, 2006).

Moreover, the combined effects of changes in temperature, precipitation, atmospheric humidity, wind intensity, and the concentration of some atmospheric pollutants that are deposited by different mechanisms on the surfaces of buildings, sculptures, monuments, and any other construction can substantially damage the building materials (Sesana et al., 2021). On the basis of this concern and to determine the possible impact that environmental factors may have on cultural heritage, different authors have developed dose-response formulas to predict this impact, through damage quantification, and many others have applied these formulas in different past, current, and future conditions to predict damage under different scenarios. Sesana et al. (2021) classified the impact of climate change on cultural heritage into three main categories: (i) impact of gradual changes in climate on outdoor cultural heritage, (ii) impact of gradual changes in climate on indoor cultural heritage and collections, and (iii) impact of changes in the natural physical environment on cultural heritage. They showed that few studies have measured the impact but most of them just described climate-induced outdoor damage based on qualitative analysis.

Orr et al. (2021) concluded that research on the impact of climate on outdoor cultural heritage, as well as the study of heritage sites and individual buildings and monuments, has focused on Europe. The research methods used were mainly based on quantitative methods and the use of literature and secondary data, for example, the most used techniques in these studies were modeling (29 %), field or laboratory research (24 %), and remote sensing/GIS (15 %). They also identified an increase in the number of papers that identify barriers to reducing the impact of climate change on cultural heritage and incorporate this issue in mitigation and adaptation actions. These barriers from the technical perspective refer to (i) methodological barriers, (ii) lack of understanding of the complexity of the impact of climate change, and (iii) conservation challenges.

In addition, although there is considerable research on this, especially in Europe, there are serious concerns about the way in which internal policies and thus the management of cultural heritage currently tends to be handled (Valagussa et al., 2021). At present, "cultural heritage policies and laws tend to reflect climate change issues more than the other way round" (European Commission. Directorate General for Education, Youth, Sport and Culture, 2022). This indicates that a change in scientific research is necessary, but also a change in the application of policies and laws for the protection of cultural heritage.

Given those research gaps, here, we aim to perform a quantitative analysis of the impact of climate change on the weathering of cultural heritage materials using one of the most widely used methods in research on this subject: dose-response formulas. These formulas have been designed for certain specific materials such as marble and limestone (Bonazza et al., 2009a), which are one of the most employed stones in the construction of cultural heritage buildings.

First, we performed a state-of-the-art review of the existing environmental damage and the associated dose-responde formulas, when available. Then we analyzed the sensitivity of the dose-responde formulas to the variability in climate and air pollution factors under the business-as-usual SS5-RCP8.5 scenario proposed by the IPCC. Lastly, we critically assessed the current methodologies and models used to estimate the effects of climate and air pollution on built cultural heritage materials.

In consequence, this research aims to answer the following scientific questions.

- How is environmental damage to built cultural heritage currently quantified?
- What are the modeled responses of key built cultural heritage sites under climate and air pollution changes?
- According to quantified scientific evidence, what are the research recommendations for future modeling of the impact of the environment on built cultural heritage?

1.1. State-of-the-art quantitative and qualitative review of the impact of climate change on built cultural heritage

The present state of the art focused on analyzing the main damages that can be measured in materials such as limestone, marble, and other types of stones, commonly used for the construction of what is currently known as cultural heritage and that are very varied in SCORE sites, by means of dose-response formulas and whose structure has climatic parameters and/or parameters related to atmospheric contamination. Thus, considering the present review, the damages were grouped according to the main stressor.

1.1.1. Precipitation

One of the main reported causes of damage to built cultural heritage materials is precipitation (Sesana et al., 2021). An increase in precipitation could cause saturation of soils and overloading of gutters and downpipes and hence a higher risk of damp penetration in historical materials (Sabbioni et al., 2008). Increased precipitation can also cause increased deposition of contaminants on surfaces and contribute to their penetration into the pores of building material (Camuffo, 2019).

The effects of precipitation also include an increase in biological activity and, very importantly, although not considered in this work, an increase in flood risk, which undoubtedly harms the cultural heritage (Haugen and Mattsson, 2011). If we consider the increase in precipitation with an increase in the concentrations of atmospheric pollutants, it may increase the damage due to the deposition of pollutants that contribute to the corrosion of the material or a loss of material due to the increase of acid rain, which is directly related to the corrosion of the material (Sesana et al., 2021).

When talking about the impact of precipitation on cultural heritage, it is important to note that there are two ways for damage to occur: The first is direct damage such as mechanical scouring or soaking, and the second is indirect damage such as freeze–thaw cycles, chemical corrosion, and biological degradation (Wang et al., 2022). In other words, freezing can increase the internal stress generated by water inside the porous material and damage the cultural heritage structure (Grossi and Brimblecombe, 2007). The increase in precipitation, particularly in a warmer climate, can accelerate the surface recession of vitreous materials and carbonate stones (Grøntoft, 2011; Camuffo, 2019), and the fluctuations in precipitation and temperature can accelerate the microbial growth on the surfaces of stone or wooden materials (Bates et al., 2005). Heavy precipitation events, including stormwater, storm surge, and flood, can destroy fragile outdoor heritage sites (Fu et al., 2022), and finally, even normal precipitation has a slow but prolonged negative impact on the heritage structure through salt cycles and capillary damp (Curtis, 2016).

The damage caused by precipitation depends on water resistance, pore structure, material composition, and other characteristics of the materials (Wang et al., 2022). As for damage such as corrosion of stones, the recession of carbonate stone surfaces are due to precipitation and the deposition of atmospheric pollutants such as SO_2 and NO_x, as well as increased atmospheric CO_2 and increased concentrations of acid rain that directly impact the cultural heritage (Sesana et al., 2021).

Another mechanism that generates different types of damage such as biomass accumulation (Formula 7 in Table 1), algae decay, and lichen decay on stones is biological growth, which is related to precipitation, humidity, and temperature (Sesana et al., 2021). In general, the damage is caused by biological colonization consisting of visual occlusion and overlying plants, algae, molds, mosses, and lichens (Bonomo et al., 2020).

The microorganisms that play a potential role in the biodeterioration processes of inorganic materials are autotrophic and heterotrophic bacteria, fungi, algae, and lichens. Microbial biofilms interact with inorganic materials in several ways (Joseph, 2021): (i) physical deterioration, where the material structure is affected by microbial growth (e.g., physical or mechanical breaking); (ii) aesthetic deterioration due to fouling; and (iii) chemical deterioration and mineral and metal transformations due to excretion of metabolites or other substances such as acids that adversely affect the structural properties of the material (e.g., increase in porosity, weakening of the mineral matrix, dissolution of minerals and formation of biominerals, and biocorrosion of metals and alloys).

The so-called biomineralization or biologically induced mineralization is the process by which biological activity induces the precipitation and accumulation of minerals (Joseph, 2021). It is the result of the metabolism of organisms. The minerals produced by biofilms and lichens include iron hydroxides, magnetite, manganese oxides, clays, amorphous silica, carbonates, phosphates, oxalates, etc.

The measurement of this damage is done in a much more specific way considering the formation of microorganisms and the growth of biological materials on the built cultural heritage. These two phenomena depend on the climatic conditions of the area, as well as the pH of the rain, the exposure to light, and the physical characteristics of the material (Loli and Bertolin, 2018). However, some specific conditions or ranges of temperature, precipitation, and humidity may allow estimating biomass accumulation, as the growth rate depends on climatic conditions (Loli and Bertolin, 2018). Biomass accumulation, considered organic carbon accretion on surfaces, can also be estimated from temperature and precipitation using the function developed by Gómez-Bolea et al. (2012) (Ciantelli et al., 2018).

1.1.2. Relative humidity (RH)

Although the study by Sesana et al. (2021) does not mention RH as a separate factor from precipitation impacting damage to outdoor cultural heritage, it is very important to consider it because many dose-response formulas use this variable to predict damage. For example, the study by Grøntoft and Cassar (2020) established that salt deposition in limestone and changes in salt crystallization events due to fluctuations in RH result in more variations in stone aging than air pollution variations.

RH is a critical threat to heritage materials. It is important to consider its long-term variation when considering damage in the past and likely under a changing climate in the future. High humidity can promote metal corrosion and a wider range of effects by promoting the deposition of pollutants and chemical reactions (Brimblecombe, 2013). Humidity has an impact on indoor and outdoor cultural heritage, contributing to different types of damage, especially the growth of organic materials and the deterioration of certain materials such as wood and paper, which are mostly found inside museums, libraries, etc.

1.1.3. Air pollution

The line between material corrosion and surface recession is very thin. Many authors mention that the effect associated with gases, which increase the corrosiveness of the atmosphere, is material corrosion (Vidal et al., 2019). Also, most of the dose-response formulas (Formulas 1–4 in Table 1) use three main atmospheric pollutants for the measurement of stone corrosion.

- Sulfur dioxide (SO_2) and interaction with nitrogen dioxide (NO_2) and ozone (O_3) (Vidal et al., 2019): In many studies, SO_2 has been considered the most relevant contaminant to measure the deterioration of materials, especially the surface recession of stones and the corrosion of metals, coming from the combustion of fossil fuels and other materials containing sulfur. SO_2 can damage the materials when in contact with water, which turns into sulfate. In this sense, SO_2 also highly influences stone decay processes, especially for carbonate stones, as it tends to react with calcium carbonate, causing the formation of soluble gypsum crusts. This may cause not only stone surface recession but also soiling, mainly depending on the incidence of rainwater (Kucera and Fitz, 1995; Tidblad et al., 2012).
- Nitric acid (HNO_3) (Vidal et al., 2019): Although its atmospheric concentrations are significantly lower than those of SO_2, the HNO_3 levels have remained unchanged, while the SO_2 levels have decreased in the past years. HNO_3 is a strong acid with very hygroscopic salts and a high deposition rate. Some studies have reported corrosion rates and deposition velocities of HNO_3 on calcareous stones and under low RH, raising concern in warm and dry climates such as Southern Europe (Kucera and Fitz, 1995).
- Particulate Matter (PM) (Vidal et al., 2019): Particulate Matter, in general hygroscopic, increases the possibility of corrosion to occur. Particulates can also be corrosive when containing corrosive chemicals and considering the catalytic role of carbonaceous particles in the formation of nitric and sulfuric acid (Kucera and Fitz, 1995).

We can add carbon dioxide (CO_2) to the list, which, despite not being included in the dose-response formulas, is directly related to the increase or decrease in acid rain. CO_2 is not only a pollutant but also a climate-altering gas, among others; the CO_2 from the air that dissolves in rainwater makes the rain slightly more acidic [carbonic acid (H_2CO_3)]. If the rain becomes more acidic, it contributes to the corrosion of materials (Vet et al., 2014). Rainwater reacts with materials that are largely made of calcium carbonate ($CaCO_3$); the most common example is limestone, transforming the slightly soluble calcium carbonate into more soluble calcium bicarbonate ($CaHCO_3$). This is washed away, causing the material to be weathered, which is called the karst effect (Spezzano, 2021).

In the study of the effect of air pollution on stone elements, besides the physical mechanisms related to the abrasion caused by atmospheric particles, chemical alterations could be the main cause of pollution-related stone deterioration (Wang et al., 2022). To study this damage, the surface recession equation for Portland limestone has been used multiple times (Spezzano, 2021), see Formula 3 in Table 1.

The dose-response formulas use the annual averages of air pollutant concentrations and climatic parameters as input data and return the average annual values of corrosion. Metric corrosion rate can be expressed in terms of surface recession or corrosion depth R (μm), which is defined as the displacement of a point on the corroded surface of the material from its initial position on the non-corroded surface taken (the reference point). Corrosion rates are also expressed in terms of mass loss (g/m^2) (Spezzano, 2021).

An example of the application of the Portland limestone formula is found in the paper by De Marco et al. (2017). They modeled limestone recession in Italy for the years 2003, 2005, 2007, and 2010 and found that the recession in this period decreased because the concentrations of different pollutants decreased due to mitigation measures (i.e., reduction of atmospheric pollutants and greenhouse gases).

Different studies have shown that corrosion in outdoor built cultural heritage will decrease due to decreasing concentrations of atmospheric pollutants (De Marco et al., 2017; Brimblecombe and Grossi, 2010), but it is important to mention that these studies have mostly been conducted in Europe as mentioned by Sesana et al. (2021) in their systematic review. Additionally, Europe is the predominant region researching climate change and cultural heritage with 86 articles out of 165 (52.1 %) from 2016 to 2020 (Orr et al., 2021), where policies for climate change mitigation and adaptation have been more effective, which may not be the case in the archaeological heritage in Africa, Asia, and South America (Orr et al., 2021). This impact can be further studied considering that corrosion causes a major financial problem in infrastructure, construction, mechanical engineering, and cultural heritage.

Thus, it is mandatory to understand how atmospheric environment affects the materials of cultural heritage buildings (De Marco et al., 2017). Rainwater causes surface recession in carbonate stones through three processes (Bonazza et al., 2009a): (1) clean rain effect due to rain at pH ~ 5.6 in equilibrium with atmospheric CO_2 (karst effect), (2) acid rain effect caused by rain with additional acidity due to the presence of sulfuric and nitric acid, and (3) dry deposition of gaseous pollutants occurring between precipitation events. The consequent damage process causes surface recession, whose appearance closely depends on rain and wind direction, material microstructure, and surface geometry.

To measure surface recession, the Lipfert function is used because it recognizes the three mechanisms mentioned earlier (karst effect, acid rain effect, and dry deposition) and because it can be used with future data for different projections (Bonazza et al., 2009a); see Formula 1 in Table 1. In this case, when measuring each of the mechanisms separately, the karst effect has a greater impact on surface recession due to the structure of the formula where the karst effect has a greater coefficient than the acid rain effect, which decreased over the years, and the deposition of pollutants, whose concentrations have also decreased (Brimblecombe and Grossi, 2008).

Finally, regarding the impact caused by corrosion in general, the most common phenomenon observable on stone buildings and monuments exposed to atmospheric pollution is the accumulation of pollutants on their surfaces, causing the formation of dark color surfaces known as "black crusts" (Comite et al., 2021). These crusts are formed by gypsum where atmospheric PM is embedded. Grossi and Brimblecombe (2007) mention that the impact of atmospheric pollution on stone has been studied repeatedly, showing that one of the main impacts is the chemical deterioration of the stone and the blackening of the stone due to the deposition of this pollutant, a product of the combustion of fossil fuels.

1.1.4. Temperature

Thermoclastism is due to the differential thermal expansion and contraction of mineral grains and surface–subsurface stones in response to solar radiation on materials and causes granular disaggregation and material exfoliation (Bonazza et al., 2009b). Thermoclastism has been studied using fieldwork and laboratory experiments that generally simulate the meteorological conditions of the study area (Germinario et al., 2015). Continuous temperature fluctuations cause repeated cycles of thermal expansion and contraction, causing mechanical stress on the material, which varies depending on the type of rock and is reflected in the microcracking of the material.

Regarding the thermal stress that the material may suffer due to the variations in air temperature and other factors such as precipitation and RH together with the material properties, there is a function that calculates thermal stress for a uniform elastic medium confined in the horizontal direction and unconstrained in the vertical direction using the Young's modulus, the thermal expansion coefficient, and the amplitude of the thermal variation (see Formula 9 in Table 1).

This function uses constant values that can be found in the literature and the daily temperature value. At the same time, this function allows us to identify not only the thermal stress caused but also the future stress that could be caused using climate models and future temperature projections. The risk of damage is evaluated by comparing the calculated thermal stress and the maximum sustainable load of the material (Bonazza et al., 2009b).

1.1.5. Wind

Rain with a horizontal velocity component given by the wind is called wind-driven rain (Formula 8 in Table 1) (Blocken and Carmeliet, 2004), which is one of the main factors responsible for surface erosion (Erkal et al., 2012). It usually causes surface erosion of stone materials, collapse of and/or damage to buildings and archaeological structures, and wind abrasion (Sesana et al., 2021). Surface erosion is the most studied wind-driven impact as it is a progressive and cumulative type of impact.

The loss of surface materials can be quantified by a probabilistic approach to enable variation and uncertainty and defined as a probability function of value (cultural, social, and historical), hazard, vulnerability, and exposure (Erkal et al., 2012). In terms of hazard, considering the different IPCC climate change scenarios, some extremes associated with precipitation tend to be more frequent, and therefore, an increase in wind-driven rainfall can add to the other damage (Nik et al., 2015).

Regarding the materials' susceptibility, the impact has been calculated specifically for cultural heritage made with earth (Luo et al., 2019) and for limestone. Some studies consider not only the intensity of rainfall and wind speed but also temperature and RH as factors that contribute to higher or lower erosion of the material (D'Ayala and Aktas, 2016). The impact also depends on the location of the building depending on the wind direction at the study site (exposure), which contributes to the penetration of not only rainwater but also pollutants and particles that may be in the atmosphere, which is reflected in some parts of a building being more affected than the others (Martínez-Martínez et al., 2022).

Finally, although it is not considered as a direct damage, the lifetime multiplier (LM; Formula 6 in Table 1) is based on RH and temperature. It is defined as "the predicted lifetime of the material subjected to the environmental conditions and the predicted lifetime at standard conditions of 20 °C and 50%RH" (Loli and Bertolin, 2018). Considered as a chemical risk index: when $LM < 1$, the material will have a shorter lifetime (higher deterioration rate) than the standard conditions while for $LM > 1$ the material will last longer.

2. Methodology

2.1. Study area and associated climate

The project Sustainable COnservation and REstoration of built cultural heritage (SCORE) (http://www.score-project.net) seeks to analyze in a general way the development of new dose-response formulas and the modeling of climate change, to study in an integrated way the impact of climate change on the vernacular cultural heritage in Europe and the archaeological cultural heritage in Mexico. In the project, a total of eight places were studied, six in Europe and two in Mexico, which are listed and described in detail in the Supplementary Methods in the Supplementary Material.

The SCORE project focuses its research in two geographical and climatic areas: vernacular built cultural heritage in Europe and archaeological sites (850–1200 CE) in Yucatan Peninsula (Mexico). The main climate types (Arnfield, 2020) of the selected areas are very different: equatorial (A) in Latin America and warm temperate (C) in Europe. This allows us to study the behavior and durability of built cultural heritage materials in different climate conditions.

Vernacular built cultural heritage is much less protected than monuments by national and international legislation and conventions and therefore it is more vulnerable. It is exposed to climate conditions, increasing pollution (air, soil and water) and changes produced by men: architectural changes, material substitution, etc. Mexico has around 25,000 pre-Hispanic archaeological sites, this amount of archaeological built cultural heritage makes difficult their protection. In the Yucatan Peninsula the archaeological sites which not only compromises buildings but also the magnificent reliefs and mural paintings, were exposed over decades to the subtropical weather of the region (e.g. extreme heat and rainfall).

- The French Vexin (35 km northwest of Paris) is a rural area where different styles of architecture coexist and testify to an ancient occupation and different periods of construction. The main principal building material is limestone with lime mortars.
- The site of Patones de Arriba, 65 Km from Madrid, in the Northern highlands (Central System Mountain range), stands out for its diversity of cultural (historical, vernacular, ethnographic, industrial) and natural values. Traditional houses of Patones were made of slate, timber and Arab brick and tiles.
- Cremona (Italy) is a city located in northern Italy, in Lombardy. In this area from the Middle Ages until the 19th century we find carefully constructed brick masonry walls, built using earthen mortar. They co-exist alongside walls built with lime.
- Aeolian Archipelago (Sicilia) presents different emerged and underwater cultural heritage. The building materials are of local origin: foundations were built using blocks of lava rock, pumice for exterior walls and tuff stone for terrace flooring made with dry stone walls.
- Chateau de Coucy (France) located 100 km N-E of Paris: The castle and the entire fortress were among the largest fortified complexes in France, and it has been built with local limestones and lime mortar.
- Out of around 4 million buildings in Denmark, 9.000 are protected and 300.000 have been classified as worth preserving. A major part of these buildings is erected in fired clay bricks, where some of them are covered with plaster.
- The World Heritage site of the Pre-Hispanic City of Chichen Itza is in the southeast of Mexico in the North of Yucatan Peninsula and is the second most visited in Mexico. It and has been constructed with local limestone and lime.
- The mixed property of the Ancient Mayan City and Protected Tropical Forests of Calakmul (Oxpemul) is located within the Calakmul Biosphere Reserve (CBR) in the south of the state of Campeche, in the Yucatán Peninsula. The CBR is the second largest area of tropical rainforest in the Americas. It incorporates 38 recorded archaeological sites and numerous archaeological remains. Building materials are local limestones and lime.

2.2. Climate data

To apply the dose-response formulas for past and future climate data, we used monthly data at 30-s resolution (~1 km) of temperature (°C) and precipitation (mm/month) from the WorldClim dataset between 1990 and 2020. For the future climate data, we used the CMIP6-downscaled climate projections from 2020 to 2100, and bias correction was done with WorldClim v2.1 as baseline climate.

At each SCORE site, we extracted temperature (mean, minimum and maximum) and precipitation from the available bias-corrected general circulation models (GCMs) for the CURRENT (2020–2040) and FUTURE (2081–2100) periods, as inputs for the dose-response formulas (Section 2.4). We used the simulated climate under the SS5-RCP8.5 scenario describing a low climate mitigation future and business-as-usual socioeconomic measures.

Regarding the downscaling for the models, observed climate data were used to describe relationships between larger-scale climatic variables and local surface climatic variables. These relationships were then applied to the GCM output under the assumption that the GCMs perform best for the larger-scale climatic variables and that the relationships remain valid in a changing climate. The data used were initially produced by the projected change in a climatic variable. The projected changes are calculated as the difference between the GCM outputs for the baseline and for the target years, i.e., the future years for which the climate is to be simulated. These changes are interpolated to a grid with a high (~1 km) resolution.

2.3. Air pollution data

The current air pollution parameters needed for the application of Formulas 1–5 in Table 1 were extracted and averaged from daily measurements from 2006 to 2021 at each site (European Environment Agency, 2023; The Weather Company, 2023). As for future projections, the values obtained correspond to projected average values and the methodologies were based on the references cited for each compound and for each site: hydrogen ion [H^+] from Shah et al. (2020), sulfur dioxide (SO_2) from European Environment Agency (2023) and The Weather Company (2023), nitric acid (HNO_3) from Aksoyoglu et al. (2020) and Zheng et al. (2008), and particulate matter (PM_{10}) from European Environment Agency (2023) and The Weather Company (2023). Moreover, although carbon dioxide (CO_2) is not within the parameters of the dose-response formulas, it was used to calculate future projections of pH and its relationship to [H^+] and HNO_3 (Vet et al., 2014; Riahi et al., 2011). All the projected pollution changes are consistent with the future modeled climatic data under the SS5-RCP8.5 scenario. Then, considering the requirements of each response dose, the units were transformed, if necessary, to ensure the correct calculation of each one.

2.4. Dose-response formulas

Different authors have calculated the impact of climate change on cultural heritage using dose-response formulas that generally incorporate/consider climatic variables, such as precipitation, temperature (maximum, minimum or average), and wind speed, in addition to atmospheric pollution variables, such as the concentrations of SO_2, HNO_3, and H^+ as a function of rain pH, to determine the deterioration of different materials.

The different dose-response formulas are defined using the physicochemical relationship between the degradation of the material and the degradation factors such as environmental exposure (Jernberg, 2004). This construction was based on regression analysis to determine how environmental factors affect material degradation, i.e., how climatic variables such as precipitation, temperature, and wind speed among others, as well as nonclimatic variables such as the concentrations of atmospheric pollutants, generate damage to building materials. To establish these relationships, experimental studies exposed materials to certain environmental conditions and measured the degradation result.

To obtain the list of existing dose-response formulas, we performed a bibliographic review in Science Direct and Web of Science from 1995 to June 2023 using keywords and synonyms: quantification of damage, built cultural heritage, dose-response formulas, and impact of climate change on cultural heritage. After suppressing duplicates, n = 87 scientific articles (see Supplementary Table 1) were extracted after the first screening of title and abstract (all articles that do not relate to built cultural heritage or impact of climate and/or air pollution are discarded). After the first screening, we relied on extracting articles where damage quantification had been performed using a dose-response formula, especially but not exclusively for limestone, considering that it is the most used building material in the study sites. In this sense, the criteria for the exclusion and inclusion of articles were based on the following: (i) dose-response formulas involving climatic parameters such as precipitation and/or temperature, (ii) dose-response formulas involving atmospheric pollution parameters, (iii) application of the formula for limestone or a stone as a building material of cultural heritage, and (iv) studies on outdoor cultural heritage.

The formulas shown in Table 1 were extracted from this search (n = 9) using climatic and/or air pollution parameters. Formulas 1–5 correspond to surface recession in limestone, while Formulas 6–9 correspond to other types of damage generated in stones in general, siliceous stones, marble, and limestone. Note that several articles in Supplementary Table 1 use the same formulas. The climatic and air pollution parameters calculated according to the methodology were used for each of the dose-response formulas in Table 1.

Where in **red** we can find the parameters that varied over time and in **blue** those that remained constant.

2.5. Sensitivity of the formulas to climatic conditions

To analyze the sensitivity of the dose-response formulas to future climate changes, including precipitation, temperature, and wind speed, as well as the concentrations of some atmospheric pollutants, a comparison was performed when applying the formulas to two different scenarios (CLIM and ALL). In the first scenario (CLIM), the changes in damage are calculated in response to future climate change only (from the historical period to 2081–2100), keeping the parameters related to air pollution constant. In the second scenario (ALL), the changes in damage are calculated in response to both future air pollution and climatic parameters.

This allows us to identify whether the damage tends to decrease or increase and the relative contribution of environmental factors (air pollution versus climate) depending on the site of interest and the formulas used. Additionally, we assessed the robustness of the changes given the intermodel variability in the climatic parameters: a response is considered robust when the sign of anomaly is similar to at least 80 % of the climate model simulations.

3. Results and discussion

Considering the data obtained for each of the SCORE sites, a comparison was made between current precipitation (last 30 years until 2020) and future precipitation (2081–2100) to show the percentage of future change. The same was done for temperature (maximum, minimum or average), which is one of the two most used parameters in the dose-response formulas. Then, the simulated future changes in damage were calculated using the climate and/or air pollution data, according to the state-of-the-art dose-response formulas. Finally, we discuss the discrepancies across the dose-response formulas.

3.1. Future projections

Fig. 1A shows the ensemble-mean precipitation change between the historical period and the current (2021–2040), near-future (2041–2060), middle-future (2061–2080), and far-future (2081–2100) periods. We found a slight increase in precipitation in Chateau de Coucy in France (+0.2 %) and an increase in precipitation in Denmark (+7.8 %). The Vexin Francais Natural Regional Park in France (−0.8 %) and Cremona in Italy (−3.5 %) show a slight decrease in precipitation, while the Aeolian Archipelago in Italy (−11 %) and Patones de Arriba in Spain (−12.9 %) show a significant decrease in precipitation, and the two sites in Mexico, Chichen Itza (−18.2 %) and Oxpemul (−18.4 %), show a

Table 1
Review of the existing dose-response formulas for damage quantification associated with climate and/or air pollution change.

	Formula name	Application material	Climatic stressor	Pollutant related	Formula	References
1	Surface recession (μm/year)	Marble and Limestone	Precipitation	$[H^+]$ SO_2 HNO_3	$-\frac{dx}{dt} = 18.8{*}R + 0.016[H^+]R + 0.18\left(V_{dS}SO_{2(g)} + V_{dN}HNO_{3(g)}\right)$	(Lipfert, 1989)
2	Surface recession (μm/year)	Limestone	Temperature and Precipitation	$[H^+]$	$\Delta x = 2.7{SO_{2(g)}}^{0.48}exp(-0.18T)t^{0.96} + 0.019Rn[H^+]t^{0.96}$	(Tidblad et al., 2001)
3	Surface recession (μm/year)	Marble and Limestone	Relative Humidity and Precipitation	SO_2 $[H^+]$ HNO_3 PM_{10}	$R = 3.95 + 0.0059\,[SO_2]RH_{60} + 0.054\text{Rain}[H^+] + 0.078[HNO_3]RH_{60} + 0.0258\,PM_{10}$	(Kucera et al., 2007)
4	Mass Loss (g/m^2)	Limestone	TOW and Precipitation	$[H^+]$	$ML = 34.4 + 5.96TOW[SO_2] + 338Rain[H^+]$	(Jernberg, 2004)
5	Stone loss (mmol/L)	Limestone	Temperature	$[H^+]$	$\text{Stone loss} = \frac{0.16\left[1.0 - 0.015T + 0.0000922T^2\right]}{0.683} + 0.49[H^+]$	(Baedecker et al., 1992)
6	Lifetime Multiplier	Different types of stones	Relative Humidity, Precipitation and Temperature	N/A	$LM = \left\langle\frac{50\%}{RH}\right\rangle^{1.3}{*}e^{\frac{Ea}{R}\left(\frac{1}{T}-\frac{1}{293}\right)}$	(Silva and Henriques, 2015)
7	Biomass Accumulation (mg/cm^2)	Siliceous stones	Precipitation and Temperature	N/A	$B = e^{(-0.964+0.003P-0.01T)}$	(Gómez-Bolea et al., 2012)
8	Wind-driven rain load	Different types of stones	Precipitation, Wind Speed and Wind Direction	N/A	$R_{wdr} = \alpha{*}U{*}R_h^{0.88}{*}cos\beta$	(Lacy, 1977)
9	Maximum Thermal Stress	Marble	Temperature	N/A	$\sigma_T = \frac{E{*}\alpha{*}\Delta_T}{(1-\nu)}$	(Turcotte and Schubert, 2002)

R	Precipitation in m/year	TOW	Is the time of wetness (RH> 80%, T > 0º) as time fraction of a year
$[H^+]$	Hydrogen ion concentration in µmol/l evaluated from rain yearly pH	RH	Relative humidity
V_{dS}	Deposition velocity of SO_2 in cm/s	P (7)	Precipitation in mm
SO_2	SO_2 concentration in µg/m^3	α (8)	Adapted Wind Driven Rain coefficient in s/m
V_{dN}	Deposition velocity of HNO_3 in cm/s	U	Reference wind speed measured at the standard meteorological height of 10m
HNO_3	HNO_3 Concentration in µg/m^3	R_h	Unobstructed rainfall intensity
T	Surface air temperature in ºC	β (8)	Wind incidence angle between wind direction and the normal vector of the wall surface (0°)
t	Time in years	E	Young's modulus
RH_{60}	Is the measured relative humidity when RH > 60% otherwise 0%	α (9)	Thermal expansion coefficient
Rain	Amount of rainfall in mm or in m/year	ΔT	Actual amplitude of the periodic surface temperature variation
PM_{10}	Particulate matter concentration in µg/m^3	v	Poisson's ratio

more drastic decrease in precipitation. Note that except for precipitation projections in Chateau de Coucy in France and Denmark that are not significant (67 % and 75 % agreement, respectively), all temperature and other precipitation projections are significant which allows robust messages.

Fig. 1B shows a generalized increase in the maximum temperature in all the sites studied, particularly in Patones de Arriba in Spain (+6.9 °C) and Cremona in Italy (+6.6 °C). Although the increase in maximum temperature in Mexico is lower compared to that in Europe, the absolute maximum temperatures increase in Chichen Itza (+4.7 °C) and Oxpemul (+5.5 °C), reaching temperatures above 41 °C on an annual average.

3.2. Climate versus air pollution responses across the dose-response formulas

Accounting only for future climate change (CLIM), surface recession decreases in the future in most SCORE sites, except for Denmark, which is the only site that shows an increase in the surface recession of the material calculated using Formulas 1 and 3 (see the CLIM columns in Table 2). This can be explained by the increase in precipitation which is projected (75 % of the models show the same sign) only in Denmark across the sites (other sites show no changes or decrease in precipitation). This coincides with the different studies that show that an increase in precipitation can generate greater impact on the material, because it contributes to the deposition of contaminants (Sabbioni et al., 2008) and helps the penetration of these contaminants into the pores of the material (Camuffo, 2019).

Future decreases in the surface recession are usually more pronounced when air pollution changes are accounted for. This scenario of decreased air pollution impact was observed during COVID-19 where, by reducing atmospheric pollution, the impact on cultural heritage was reduced (Broomandi et al., 2022). This is true for Formulas 1–4 because Formula 5 does not use pollutant concentration values (Table 2).

The decrease in pollution-driven damage is due to a decrease in rain acidity because of the decrease in CO_2 (Vet et al., 2014). However, we found a large uncertainty across the surface recession dose-response formulas up to 40 % difference in the future change response (Table 2). Although Formula 4 shows the same trend, its change is much more noticeable between CLIM (no significant damage impact) and ALL (slightly significant decrease). First, we observe that scenario CLIM shows a minimum decrease compared to Formulas 1–3, and second, there is a large uncertainty between the formulas because although all of them show a decrease in damage for most of the SCORE sites, the values show a high variability among them. Formulas 6–9 were only applied to one scenario (CLIM) because these formulas do not use air pollution parameters. Concerning Formula 6 (LM), it is important to note that the two sites in Mexico and the site in Spain do not present any change between current and future deterioration, unlike in the other six sites where there is a significant increase in the deterioration of the material.

Biomass accumulation (Formula 7, Table 2) shows an overall decrease, except an insignificant increase in Denmark of 5.5 % (the latter mainly due to the increase in precipitation and RH). In Mexico, a decrease of ~47 % is simulated, which can be explained by the significant decrease in precipitation and the increase in temperature.

For wind–rain erosion (Formula 8, Table 2), the impact mostly tends to decrease, more in Mexico than in Europe, but remains constant in Denmark. Finally, the thermal stress, calculated in Formula 9, has a general tendency to increase in all the SCORE sites, with Spain and Italy being the sites with the greatest increase, followed by France, Denmark, and Mexico, with the impact being consistent with the study conducted by Bonazza et al. (2009a, b).

3.3. Discrepancies and limitations of the dose-response formulas

In general, we found a great discrepancy across the damage responses: LM and thermal stress (Formulas 6 and 9, Table 2) show a damage increase, while the surface recession, erosion, and biomass accumulation indicators (Formulas 1–5, 7, and 8) show a decrease. The changes are mainly driven by changes in precipitation or humidity factor in general, which is consistent with that by Wang et al. (2022). This indicates that across the environmental variables, water is the predominant worldwide factor for the deterioration changes of cultural heritage in the future.

On the one hand, previous studies about the impact of climate change and air pollution on cultural heritage tend to show that built materials would demonstrate increases in damage (Vidal et al., 2019). We show however that the state-of-the-art dose-response formulas tend to show an opposite trend in Europe at least. In the future, cultural heritage zones with increase in precipitation coupled with decrease in the concentrations of pollutants [due to the likely implementation of policies to reduce emissions and energy efficiency (Broomandi et al., 2022)] would certainly be less affected (De Marco et al., 2017). Regarding the contribution of climate change versus air pollution change to damage calculated using the surface recession dose-response formulas (Formulas 1–5 in Table 2), Fig. 2 shows in general (four cases out of five) that climate change has a higher contribution to damage than air pollution change. According to these results, warmer climates in Europe would present lesser damage because of drier climate

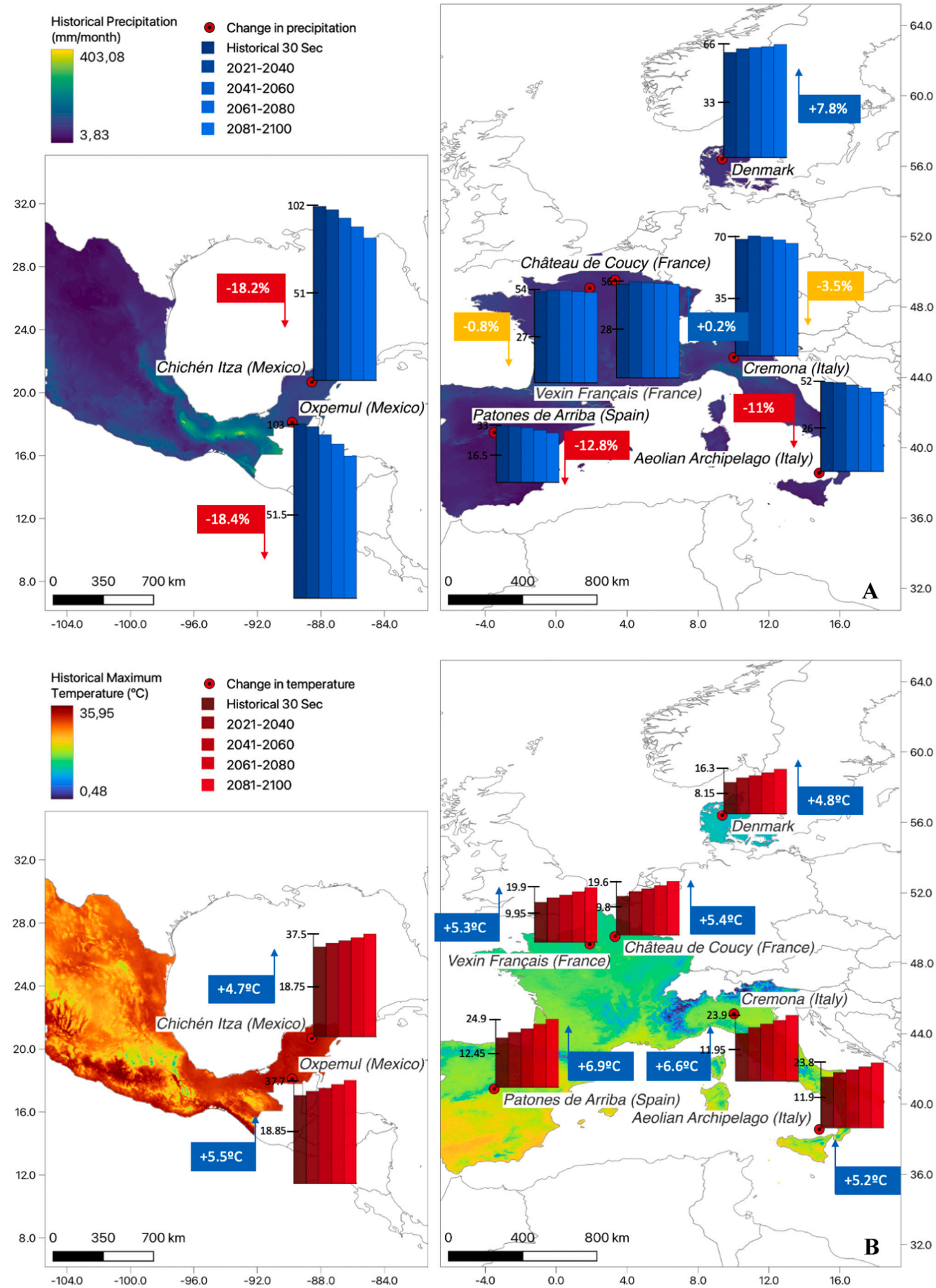

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Fig. 1. Changes in (A) mean precipitation and (B) maximum temperature from the historical period (1990–2020) to the current (2021–2040), near-future (2041–2060), middle-future (2061–2080), and far-future (2081–2100) periods in the SS5-RCP8.5 scenario. The underlying map presents the historical annual mean precipitation climatology in mm/month and the maximum temperature climatology in °C at 1 km resolution. For the precipitation and maximum temperature, the average of 24 and 22 bias-corrected CMIP6 models are shown in panel 1A and 1B, respectively. The red circles indicate the SCORE sites. The red, yellow, and blue arrows indicate a large decrease (< −10 %), a decrease (<0 %), and an increase (>0 % or > 0 °C) in precipitation and temperature, respectively between the historical period (1990–2020) and far-future (2081–2010). Historical 30 s or seconds (of a longitude/latitude degree) refers to about 1 km at the equator.

Table 2
Percentage difference of damages to materials for scenario 1 (columns CLIM), which applies the formulas varying only the climatic parameters while using a current and constant value for the concentrations of atmospheric pollutants, and for scenario 2 (columns ALL), which applies the formulas varying both the climatic and air pollution parameters. The darker **red** cells correspond to a damage increase, and the darker **blue** cells correspond to a higher damage decrease. Results in **bold** indicate values where >80 % of the models show the same sign of change. The absolute changes can be found in the Supplementary Table 2.

Dose-response formula	1		2		3		4		5		6	7	8	9
Scenario	CLIM	ALL	CLIM	ALL	CLIM	ALL	CLIM	ALL	CLIM	ALL	CLIM	CLIM	CLIM	CLIM
Vexin Français (France)	*−2,7%*	*−3,1%*	**−5,3%**	**−16,8%**	**−18,8%**	**−20,3%**	*0,0%*	**−2,7%**	**−5,4%**	**−5,4%**	**12,1%**	**−8,8%**	**−5,7%**	**10,0%**
Patones de Arriba (Spain)	**−11,4%**	**−12,5%**	**−8,5%**	**−32,0%**	**−17,9%**	**−18,1%**	*0,0%*	**−1,5%**	**−7,1%**	**−7,1%**	**13,0%**	**−16,6%**	**−10,5%**	**12,0%**
Oxpemul (Mexico)	**−16,7%**	**−17,3%**	**−14,9%**	*−1,4%*	**−19,3%**	**−12,3%**	*−0,1%*	**−4,3%**	**−5,6%**	**−5,6%**	**13,0%**	**−46,5%**	**−15,1%**	**7,4%**
Cremona (Italy)	**−5,9%**	**−6,7%**	**−7,5%**	**−28,8%**	**−36,5%**	**−39,4%**	*0,0%*	**−3,9%**	**−6,5%**	**−6,5%**	**18,8%**	**−17,5%**	**−5,5%**	**11,1%**
Chichén Itza (Mexico)	**−16,3%**	**−16,9%**	**−15,0%**	*−1,5%*	**−19,3%**	**−10,5%**	*−0,1%*	**−3,2%**	**−4,7%**	**−4,7%**	**13,0%**	**−46,8%**	**−14,7%**	**6,2%**
Château de Coucy (France)	*−2,3%*	*−2,9%*	**−5,8%**	**−24,3%**	**−17,2%**	**−21,9%**	*0,0%*	**−4,1%**	**−5,5%**	**−5,5%**	**11,0%**	**−8,2%**	*−1,1%*	**10,2%**
Denmark	*3,9%*	*3,7%*	*−1,6%*	**−5,8%**	**−3,0%**	*1,7%*	*0,0%*	**−4,8%**	**−4,7%**	**−4,7%**	**9,0%**	*5,5%*	*−3,0%*	**9,4%**
Aeolian Archipelago (Italy)	**−10,1%**	**−11,0%**	**−7,1%**	**−22,1%**	**−11,6%**	**−16,4%**	*0,0%*	*0,0%*	**−5,3%**	**−5,3%**	**5,0%**	**−19,9%**	*−1,7%*	**8,8%**
	Surface Recession										Lifetime Multiplier	Biomass accumulation	Wind–rain erosion	Thermal stress

(Southern Europe), so less acid rain–related damage. In this sense, future damage to built cultural heritage materials is generally reduced in sites where precipitation decreases or is constant.

The relative contribution of air pollution to materials' deterioration is lower than the direct contribution of rain, but it is important to consider emerging or coastal pollutants and study whether their presence could increase in the future (Enyoh et al., 2020; Comite et al., 2021) along with their effects on cultural heritage degradation. The impact of climatic extremes (droughts, floods, cold and hot extremes, and windthrows) on built cultural heritage is rarely studied even though they would increase in frequency, duration, and intensity in most parts of the world according to IPCC (2022). "How much some specific

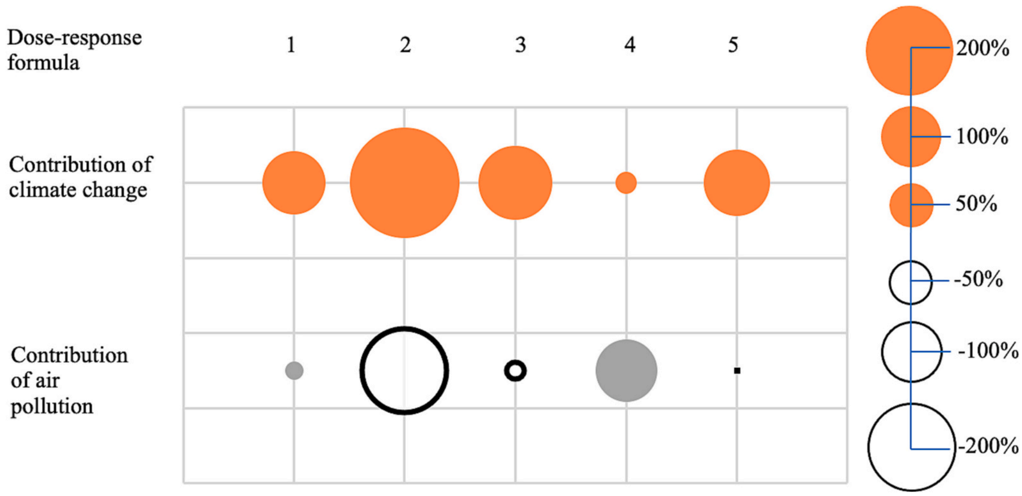

Fig. 2. Relative contribution of climate change (orange circles; units: %) versus atmospheric pollution changes (gray and white circles; units: %) to surface recession in limestone given the five different surface recession formulas. Contributions have been averaged among all sites. Note that the white circles with black border indicate a negative contribution, i.e., future damage is reduced in response to air pollution according to the formulas.

climatic extremes could affect much more building materials than several years of mean climate?" remains an open question in the literature, as rightly pointed out by Bonazza and Sardella (2023).

On the other hand, our study shows that the current dose-response formulas are strongly limited to study the long-term environmental impact on built cultural heritage. Several factors can restrict the representativity and validity of the current dose-response formulas for future prospects: site-specific affectations (tourism, architecture characteristics, and maintenance), nonlinear impact (one extreme event—thunderstorm, flood, pest, compound heatwave and drought, etc.—can affect more than years of mean climate), lack of long-term data (>1 year), and lack of real-time building integrity assessment (most facades or internal failures are not monitored continuously), among others. There is an urgent need to build more reliable formulas incorporating several types of damage, combining short- and long-term- climate change consequences (at least temperature, precipitation, and rainwater pH changes due to CO_2) and air pollution impact. Moreover, the dose-response formulas studied do not explain all the possible damage that could be caused (e.g., structural damage and microcracking of a stone), and our review shows that they are not accessible for several key materials (limestone is the overwhelmingly studied material). Given our results and state-of-the-art review, we recommend the following: (i) a quantitative compilation of monitoring campaigns worldwide to estimate long-term effects produced by the on-site climate and air pollution changes and (ii) a more comprehensive comparison of in-lab characterized damage versus observed on-site cultural heritage damage and measurements of damage to the site.

4. Conclusions

Large research gaps remain in the estimation of the future effects of climate and air pollution on cultural heritage. For the first time, we quantify here the combined effects of air pollution and climate changes on a multisite panel in a business-as-usual warming and air pollution scenario, using reviewed state-of-the-art dose-response formulas. Surprisingly, by applying these common formulas, we found that a significant decrease in damage is projected in built cultural heritage materials in most Europe and Mexico sites: decreases in surface recession (−10 % ± 10 %, on average), biomass accumulation (−20 % ± 18 %), and wind–rain erosion (−7 % ± 6 %) in response to future climate and air pollution changes, except in the regions where precipitation substantially increases (Northern Europe) by 2100.

Climate change is a worrying and growing phenomenon that could potentially increase damage and trigger more critical failure. However, the current dose-response formulas based on site-specific measurements of damage, climate, and air pollution, tend to show an opposite picture, but several limitations will be mentioned. A large uncertainty in the magnitude of the damage to built cultural heritage materials was found: For the same site, surface recession changes vary up to a 40 % difference across the equations. Greater amount of information that allows better modeling of the future climate in the medium and long term will be employed in future studies of built cultural heritage weathering evaluation. It should also be considered that the concentrations of pollutants used to quantify the damage to heritage materials, according to climate and air pollution projections, have decreased and could likely continue to decrease, mainly due to climate change mitigation and environmental policies. Moreover, we found that most of the existing studies focused only on limestone, which is the most employed stone in Europe but strongly limits any regional assessment of environmental-related damage to built cultural heritage. All these limitations on the current dose-response formulas introduce strong biases that future research should tackle.

Precipitation (and humidity) is a fundamental variable in the deterioration of cultural heritage; however, reassessment of the formulas must be prioritized, which is based on the current and future context of air pollution, climate change, and climatic extremes, for which new laboratory methodologies and field measurements must be implemented, for different materials and under future contamination scenarios.

Finally, considering the abundance of studies conducted in Europe, not only due to the large amount of cultural heritage that the continent possesses but also due to the centralization of this theme in the region, it is important to explore new regions around the world whose cultural heritage may be affected.

CRediT authorship contribution statement

Oscar Julian Esteban-Cantillo: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Resources, Validation, Visualization, Writing – original draft, Writing – review & editing. **Beatriz Menendez:** Conceptualization, Funding acquisition, Investigation, Project administration, Supervision, Writing – original draft, Writing – review & editing. **Benjamin Quesada:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Benjamin Quesada reports financial support was provided by European Commission. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Historical and future data for precipitation and temperature were provided by WorldClim (https://www.worldclim.org), also for historical data of the National Oceanic and Atmospheric Administration -NOAA in Climate Data Online (https://www.ncei.noaa.gov/cdo-web/). For air pollution data, please refer to the methodology where the source is specified for each parameter.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.170945.

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