



An assessment of the contribution of air pollution to the weathering of limestone heritage in Malta

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Abstract

Malta is known for its limestone megalithic temples of which many are inscribed on the UNESCO World Heritage List. A variation of this limestone was historically, and until very few years ago, a primary building material in Malta. The temples are subject to various environmental influences which until recently have led to several collapses due in part to serious stone surface and infill loss. As a protection measure, open-sided shelters have been built over three of these temples. This work assesses the degrading influence of air pollution (nitrogen dioxide, ozone, particle matter, sulfur dioxide, and acidity in rain) on the temples, in combination and comparison with the influence of other environmental factors (relative humidity, temperature, precipitation, moisture, sea salt, wind) and in this respect evaluates the potential protective effect of the shelters. The variation in air pollution weathering of limestone exposed outdoor in Malta was calculated by exposure–response functions from the ICP-materials programme and compared with measured values, and its contribution to the deterioration of the temples was evaluated. The difference between urban and rural locations in Malta, in the first year of atmospheric chemical weathering of limestone due to air pollution, was found to be about one micrometer loss of stone surface. This is probably less than the annual variations due to the influence of natural climatic factors, and small compared to the present annual variations in continental Europe. The deposition of sea salt and presence of salts on and in the limestone megaliths and changes in salt-crystallization events due to relative humidity fluctuations, inside and outside the shelters, will account for more of the variations in the first year of weathering of Globigerina limestone than variations in air pollution. The deterioration will also be related to temperature (including condensation events), wind parameters and rainfall, as well as ground water replenished from areas beyond the shelter.

Keywords Megalithic temples of Malta · Air pollution · Limestone weathering · Exposure–response function · ICP-materials

Introduction

This paper compares fresh limestone atmospheric weathering rates with regards to the presence of air pollution in different locations in Malta, and discusses the importance of this weathering, in relation to other environmental influences such as humidity and salts, and the importance of air

pollution for the preventive open sheltering which has been implemented for the megalithic temples of *Ħaġar Qim*, *Mnajdra* and *Tarxien* (Cassar et al. 2018). The importance for Maltese limestone buildings in general is also discussed.

The paper compares the measured recession of *Globigerina* and *Portland* limestone, with that expected in different locations in Malta with different amounts of air pollution. The comparisons are based on results from exposures of experimental samples, and on calculations by an air pollution exposure–response function (ERF) for the recession of *Portland* limestone in continental Europe, which are reported in the literature. The expected variations in the first year recession of *Portland* limestone due to atmospheric chemical weathering were first calculated from air pollution data available for Malta by the ERF, reported by

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the ICP-materials project (ICP 2018), as is explained in the Methods section.

The expected annual recession of Portland as compared with Maltese Globigerina limestone was then calculated from the reported weight loss on experimental samples co-located in England (Cabello-Briones 2015). To assess the relevance of the recession values of Portland limestone in Malta calculated by the ERF, for the recession of Maltese Globigerina limestone, the recession values calculated by the ERF were then compared with observed reported values for Maltese Globigerina limestone samples exposed inside and outside the protective shelters at the megalithic temple site of Ħaġar Qim (Cabello-Briones and Viles 2017). This information was further used to assess differences in limestone weathering in Malta due to anthropogenic air pollution. Differences between the European continental situation, which is the basis for the applied ERF, and the Maltese environment were then discussed. The paper thus assesses the possible contribution of atmospheric chemical weathering of limestone in relation to other different possible reported causes for the weathering of the temples (Becherini et al. 2016), and hence the influence of the sheltering in this respect. More complex long-term deterioration of limestone is not discussed.

The surface weathering of limestone buildings is a complex process, which depends on the interactions between characteristics of the stones, the structure of the stones, and the environment, including natural atmospheric causes, anthropogenic pollution and bio-growth. In addition to surface dissolution effects, similar to the “karst effect” on natural limestone, weathering generally occurs by chemical and physical deterioration processes associated with crust formation, physical stress due to wetting and drying, freeze–thaw cycles (not applicable to Malta), thermoclastism, salt crystallization, dissolution and leaching (Charola and Bläuer 2015; Sabbioni et al. 2010; Cassar 2010; Charola 2000). Such processes have been widely studied including in Malta where limestone was historically, and until very recently, used as a primary building material (Cassar et al. 2017; Cassar 2010; Rothert et al. 2007). Because of porosity and capillary uptake these processes will also act beneath the surface and weaken the limestones internally. The surface might become more compact due to moisture uptake and migration of the moisture outwards again by evaporative force (Rothert et al. 2007); depending on the exposition of the limestone in the cardinal directions to sun and weather. Air pollution is known to have an accelerating effect on the weathering of fresh limestone surfaces. In the long term, the air pollution will work together with other deterioration processes, and its effects will have repercussions on the overall behaviour of the limestone surface and building in a polluted environment (Brimblecombe 2003, 2014).

Clearly, there are large differences in the properties of Globigerina and Portland limestone. They consist of very different facies/micro-facies types. They can be very compact with low porosity–permeability characteristics or be very porous. For example, the porosity of Lower Globigerina limestone in Malta was reported to vary between 23.9 and 40.7% (Cassar et al. 2017) and the total porosity of Portland limestone from different strata in the Freestone succession in Bowers Quarry, Portland Island, UK, was found to vary between 15 and 26% (Dubelaar et al. 2003). Globigerina limestone is matrix-rich. Portland limestone can have a certain amount of matrix or can be a grain-stone without any matrix. These parameters will be reflected by the specific surface, permeability; and also by capillarity characteristics, which influence the moisture uptake of the stones and could be important for the uptake of marine spray (with NaCl).

The recession of other experimental samples of Globigerina and Portland limestone (other than those reported in the literature) could be somewhat different, and it is expected that the variation over a larger selection of the stones would be even larger. This gives some (undetermined) uncertainty in the results. Other uncertainties are discussed in the “Discussion” section. Nevertheless, it was evaluated that the experimental basis, of well documented recession values and variation for sets of Portland and Globigerina limestone samples, was sufficiently representative of the stones to provide realistic comparative results for the evaluation of the influence of air pollution on Globigerina limestone in Malta.

The intention of this work was not to report variation in the sensitivity of limestones depending on variation in their physical–chemical characteristics, but to compare the expected recession due to air pollution with that expected due to other environmental influences. For this purpose, it is most important to compare similar samples in all situations. However, exposure response functions, ERFs, exist only for Portland limestone, based on European continental measurements, and not for Globigerina limestone, and comparisons of simultaneously measured values for the recession of Globigerina and Portland limestone have been reported from the UK, but not from Malta. We considered, nonetheless that, it would be useful to apply the existing ERF for Portland limestone to assess the situation in Malta, and evaluate the results based on a reported experimental comparison of the recession of Globigerina and Portland limestone in the UK, and on the well documented differences in the climate and air pollution situation in Malta and continental Europe. There will be uncertainties in this discussion. However, no other such evaluation has, as far we know, been reported. We think that the evaluation presented here, of the expected magnitude of the air pollution influence as compared to other environmental influences, on the deterioration of Globigerina limestone and the megalithic temples in Malta provides useful added information. It should be noted that

Coralline limestone is also, although less, abundant in Malta. It has been much less used for building purposes than the Globigerina limestone. This work does not include an evaluation of the influences of air pollution on this stone, but it may be possible to assess this in the future by comparing the properties of the Globigerina and the Coralline limestones.

Methods

Watt et al. (2009:79) report an exposure–response equation for the first year weathering of a fresh Portland limestone surface in a multi-pollutant situation. The equation was derived from experimental exposures of stone samples, in three campaigns from 1987 until year 2002, at 46 non-coastal European stations in the ICP-materials program (ICP 2018; Tidblad et al. 2014) and the EU Multi-Assess project (Kucera 2005):

$$R = 4.0 + 0.0059 \times [\text{SO}_2] \times \text{RH60} + 0.054 \times \text{Rain} \times [\text{H}^+] + 0.078 \times [\text{HNO}_3] \times \text{RH60} + 0.0258 \times \text{PM}_{10} \quad (1)$$

where $[\text{HNO}_3]$ can be approximated by (CLRTAP 2014; Kucera 2005):

$$[\text{HNO}_3] = 516 \times e^{-3400/(T+273)} \times ([\text{NO}_2] \times [\text{O}_3] \times \text{RH})^{0.5} \quad (2)$$

where R is the surface recession (μm). The remaining parameters are annual averages of temperature (T, $^\circ\text{C}$), amount of precipitation (Rain, mm year^{-1}), concentration of H^+ ions in rain water ($[\text{H}^+]$, mg l^{-1}), the “relative humidity above 60%” (RH60, equal to RH minus 60 when $\text{RH} > 60$ and otherwise equal to zero, with RH being the relative humidity in %), and finally the concentrations in air ($\mu\text{g}/\text{m}^3$) of sulfur dioxide, $[\text{SO}_2]$, nitric acid, $[\text{HNO}_3]$, particle matter less than $10 \mu\text{m}$ in aerodynamic diameter, PM_{10} , nitrogen dioxide, $[\text{NO}_2]$, and ozone, $[\text{O}_3]$.

The values for the gaseous air pollution terms in Eqs. (1, 2) represent mainly anthropogenic emissions. The initial constant represents the background value for the weathering, suggested to be $3.2 \mu\text{m}/\text{year}$ (CLRTAP 2014), equal to about 10% of the average European weathering of the Portland limestone exposed in the ICP-materials program, and an additional unexplained variation. Kucera (2005) reports a linear time dependence for Eqs. (1, 2) over the first 4 years of the weathering, with slightly smaller reported constants of 3.1 plus the additional unexplained variation of $0.85 \mu\text{m}/\text{year}$ ($= 3.95 \mu\text{m}$ for the first year).

Calculations using Eqs. (1, 2) were performed in this work for urban, rural and coastal locations in Malta with

available air quality data, to assess the air pollution and possible climate change contributions with reference to the air pollution weathering of limestone. It must be stressed that Eqs. (1, 2) explains the influence of the climate factors by their combined effect with anthropogenic air pollution, and does not consider variations in some natural background influences, such as from windblown dust and sea salt, which are important in Malta. The climate also causes physical weathering and erosion by for example, precipitation, other forms of moisture, temperature, wind, their fluctuations and extremes, which are unrelated to air pollution, and which would be different in Malta and continental Europe. Comparison with the gradual weathering in the natural background are made in the “Discussion” section. Effects of extreme events are not discussed.

Equations (1, 2) were developed on the statistical basis of measurements from European continental stations. It could be argued that differences in the Maltese environmental data as compared to the continental European situation, disqualifies the use of Eqs. (1, 2) in Malta. We however think that calculations by Eqs. (1, 2) for Malta and a discussion of results and comparison with reported values for Maltese limestone, can contribute to the understanding of limestone weathering in Malta as well as contribute to future work. The discussion will also include uncertainties related to differences in the composition of the PM_{10} -particulate matter, including Sahara dust and salt aerosol, in Malta compared to continental Europe.

Locations and environmental data

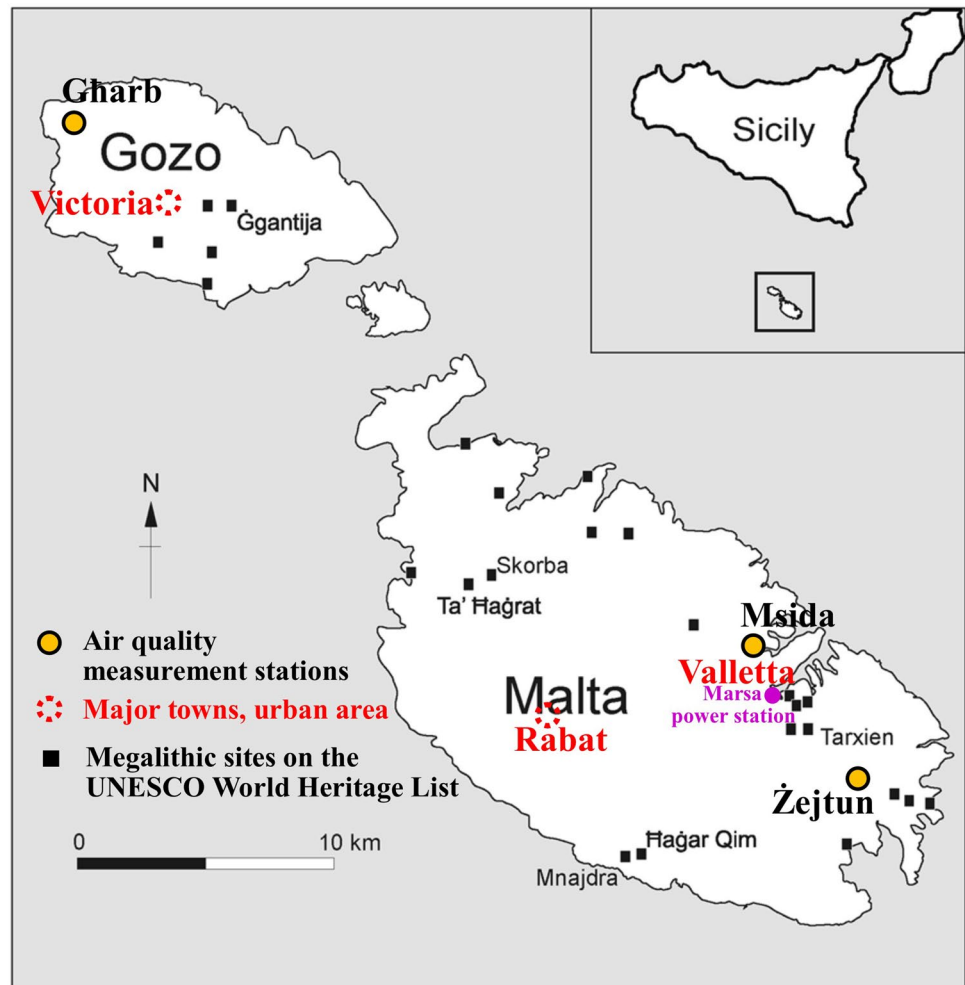
The map in Fig. 1 shows the air quality station locations in Malta for which the expected first year weathering of Portland limestone was calculated, by Eqs. (1, 2) and data from Table 1, the major towns and urban areas, and the locations of Prehistoric megalithic sites on the UNESCO World Heritage List. The major temple sites are named.

As can be seen from Fig. 1, most of the major megalithic sites are in rural locations, but with one of them, the temple site of Tarxien, in the general urban area of Valletta. Fig. 2 shows the sheltered temple of Haġar Qim.

Table 1 gives the input data used for the initial calculations of the expected weathering rates of Portland limestone, relative to noted locations of well-established air-quality measurement stations in Malta, shown on the map in Fig. 1.

The relative humidity (RH) value in Table 1 is close to the average for the norm of 1961–1990, reported by Galdies (2011), which varies from a maximum of 87% in January to a minimum of 61% in July, with the other monthly average, maximum and minimum values for the norm given on a graph in this report of Galdies. Malta is relatively small (the Maltese Islands encompass 316 sq km) and the average annual values for the climate: relative humidity (RH)

Fig. 1 Air quality station locations in Malta for which the expected first year weathering of Portland limestone was calculated, by Eqs. (1, 2) and data from Table 1. The major Maltese megalithic UNESCO sites and temples are named, and the location of the now decommissioned Marsa power station is given



temperature (T) rain amount (rain) and acidity in rain (H^+ = concentration of H^+ ions in water) were assumed to be similar for all the locations in Table 1. The values for the climate would vary between years, but there would be only slight changes from the norm period of 1961–1990; it was thus assumed that the values for 1961–1990 norm in Table 1 could represent “the present”. Continuous measurement data for nitrogen dioxide (NO_2) (2017), NO_x (NO_2 , + nitrogen monoxide, NO) (2018) and ozone (O_3) at the three stations (mentioned in Table 1) throughout the years 2017 (EEA-2 2019; ERA 2019) and 2018 (ERA 2019) (Fig. 3), showed slightly higher concentrations of ozone (O_3) in the spring/summer than winter. The observation of a seasonal trend was less certain for nitrogen dioxide (NO_2) and NO_x (continuous NO_2 values were not yet obtainable for year 2018), and PM_{10} -particulate matter, but there was some indication of lower concentrations in the summer than winter at the Msida and Żejtun stations, but not so for nitrogen dioxide (NO_2) (2017) and NO_x (2018) at the rural Għarb station.

The reason(s) for the high peak values for PM_{10} -particulate matter on 3rd and 25th March, 15th

April and 29th October (Fig. 3), was not determined here. They are in agreement for the three stations and seem to have the same explanation. The peaks do not correspond to low or high values for the average or maximum daily wind speed measured at the Malta International Airport Station, Luqa (Weather underground 2020). It is expected that they are related to the wind speed, direction and the emission sources: local anthropogenic emissions and possibly Sahara dust. The generally lower values of PM_{10} -particulate matter in Għarb than Msida and Żejtun is probably due to the dominant westerly winds on Malta, generally and in 2018 (Windy app 2020), which would ventilate and dilute local anthropogenic air pollution and bring in relatively more sea salt to Għarb and the western part of the Island than to Msida and Żejtun in the east. The in-blowing sea salt aerosol could be a significant part of the PM_{10} -particulate matter along the coasts. Further inland anthropogenic emissions would, generally, be a larger part of the PM_{10} -particulate matter. In the coastal city of Rotterdam in the Netherlands it has, for example, been found that sea salt contribute less to the

Table 1 Environmental data for parameters that influence limestone weathering (Eqs. 1, 2), for locations with different environmental situations in Malta

Environment, station				
	Urban traffic, Msida ^a	Urban background, Żejtun ^a	Rural, background Għarb ^a	Source
Parameter	Value			
Climate change multi-model mean projections for 2081–2100 relative to 1986–2005 under the RCP2.6 and RCP8.5 scenarios				
Delta T (°C)	2.375	2.375	2.375	(IPCC 2014)
Delta Rain (mm)	– 90	– 90	– 90	(IPCC 2014)
Present annual pollution and climate values				
SO ₂ (µg/m ³)	1.1	1.4	1.4	EEA 2019 ^b
RH, %	77	77	77	Measured in EU-Master project in 2005, in Rabat (Grøntoft et al. 2005)
T, °C	18.6	18.6	18.6	Malta climate norm, 1961–1990 (Galdies 2011)
Rain, mm	553	553	553	Malta climate norm, 1961–1990 (Galdies 2011)
H ⁺ (mg/l)	0.00093 (pH 6)	0.00093 (pH 6)	0.00093 (pH 6)	Typical Mediterranean background value. Reported for Casaccia, Italy, 2014 in the ICP-materials project. (Grøntoft and Ferm 2017)
PM ₁₀ (µg/m ³)	43.1	31.8	19.6	EEA 2019 ^b
NO ₂ (µg/m ³)	34.6	13.7	3.3	EEA 2019 ^b
O ₃ (µg/m ³)	56.9	73.1	95.5	EEA 2019 ^b

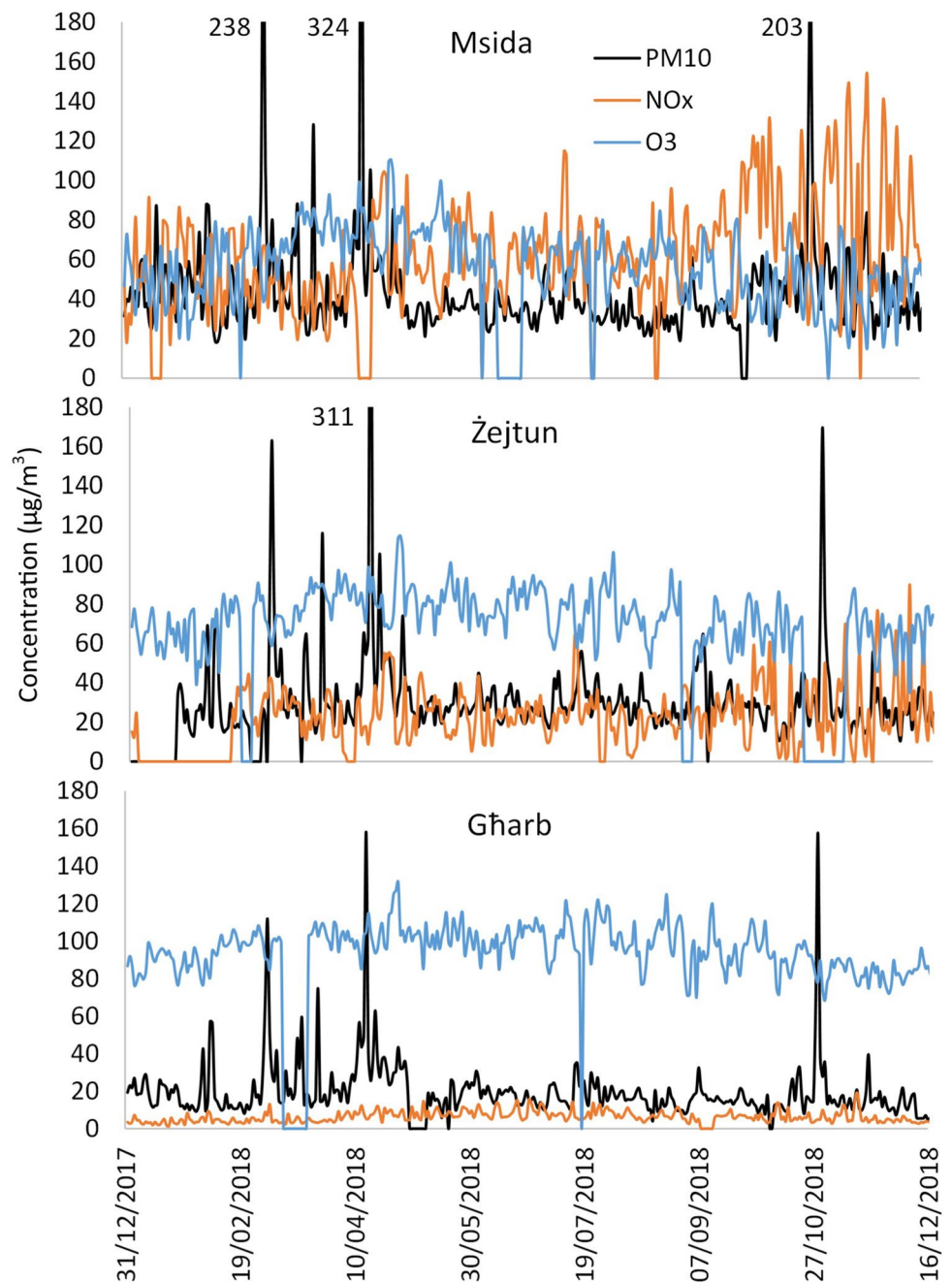
Annual averages

^aAir quality stations in Malta, which report values to the EEA (European Environmental Agency)^bEuropean Environmental Agency (EEA-1 2019)**Fig. 2** The sheltered temple Ħaġar Qim

PM₁₀-particulate matter than anthropogenic emissions, and that they are anti-correlated (Manders et al. 2009). It was however not the purpose of this work to perform time-series analysis of air quality data, which can be complex, but to document the air pollution levels in Malta and to discuss their contribution to the weathering of built heritage surfaces therein.

For comparison, the predicted limestone recession at the pollution values given by the most recent EU 2008 Air Quality Directive (AQD) (EEA-3 2019) in force and World Health Organizations (WHO) guidelines (WHO 2005) have also been calculated. The EU 2008 AQD and WHO guidelines determine annual average concentrations for PM₁₀-particulate matter = 40 µg/m³ (EU 2008 AQD) and 20 µg/m³ (WHO), and

Fig. 3 Average 24 h values for PM₁₀-particulate matter, NO_x (nitrogen dioxide, NO₂, + nitrogen monoxide, NO) and ozone (O₃) measured at the air quality stations of Msida, Żejtun and Għarb though the year 2018. The value for NO_x was calculated from reported values in ppb using the conversion factor for nitrogen dioxide (NO₂) (of 1.9). The values for some high peaks for the PM₁₀-particulate matter are given by the numbers near the peaks



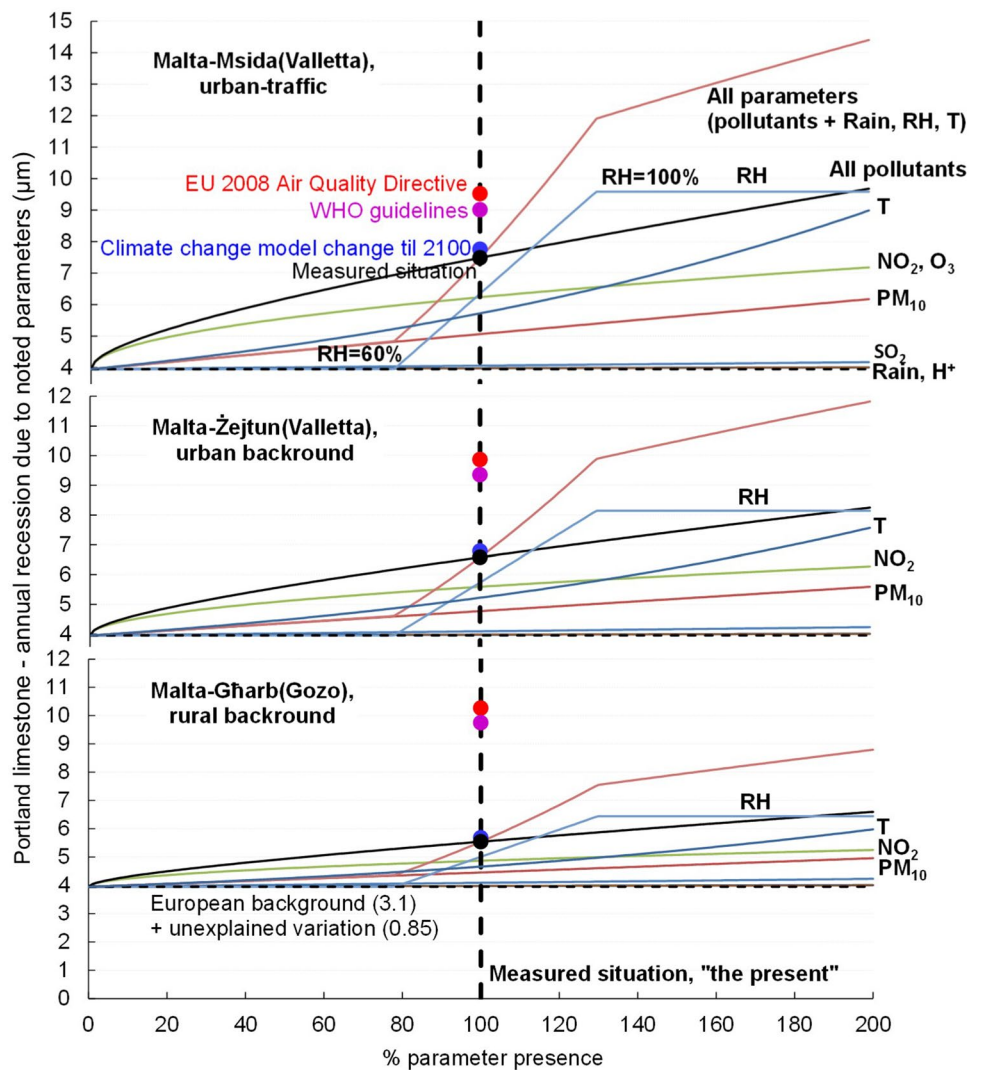
nitrogen dioxide (NO₂) = 40 µg/m³ for protection of health. As an annual mean health limit is not reported for sulfur dioxide (SO₂), the value of 20 µg/m³ for vegetation, given in the EU 2008 AQD, was applied for sulfur dioxide in both cases.

Results

Figure 4 shows calculated weathering rates according to Eqs. (1, 2), for Portland limestone theoretically exposed outdoors in Malta at the three air quality measurement

locations: Msida (urban traffic), Żejtun (urban background) and Għarb (rural background) (Table 1), as attributed to the noted single environmental parameters. The results for the measured “present” situation are represented by the vertical dotted line. The blue dot for “climate change”, shows that the predicted effect of the changes in the temperature and the amount of rain until year 2100 (given in Table 1), is only a very slight increase in the recession (when considering the simultaneous impact of all the pollutants). This prediction does not include prediction of changes in future levels of carbon dioxide (CO₂) and air

Fig. 4 Assessed weathering rates for Portland limestone theoretically exposed outdoors in Malta at three air quality measurement locations: Msida (urban traffic), Żejtun (urban background) and Għarb (rural background) (Table 1). The methodology from the ICP-materials project (Eqs. 1, 2) was applied. The blue dots show the predicted recession in year 2100, when including predicted changes in temperature and amount of rain before the year 2100 (Table 1). The red and pink dots show the estimated recession at the pollution limit values of the most recent EU 2008 Air Quality directive in force and the World Health Organization (WHO) guidelines. The theoretical influence of the presence of the single parameters is shown (see text in Sect. 4, Results). The upper part of the figure, for “Msida”, gives the combination and full description of all the curves and variables



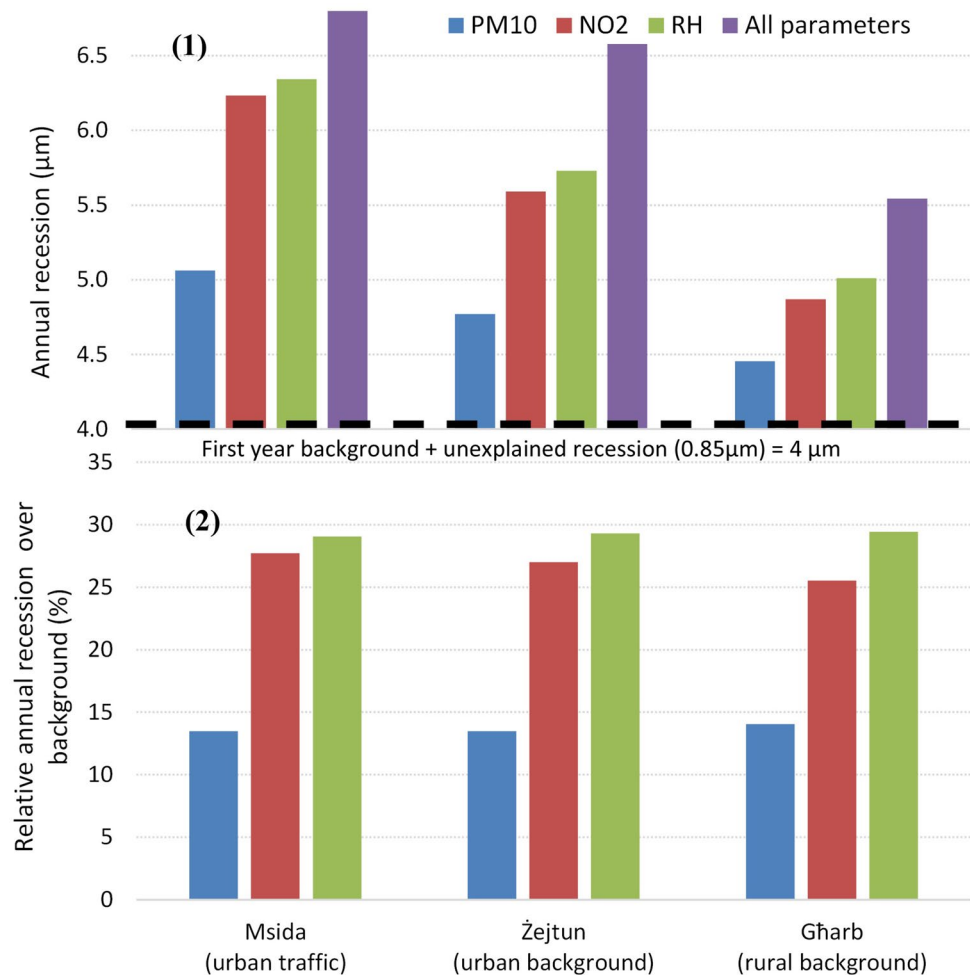
pollution, or extreme weather effects. Comparison with the red and pink dots shows that the expected recession due to all the present pollutants is today (2019) considerably lower when compared to what it would be at the pollution limit values of the EU 2008 AQD and WHO guidelines. The values on the curves for the separate environmental parameters show their theoretical single influence on the recession (according to Eqs. 1, 2) when their values change from 0 to 200%, with the values of the other parameters kept constant at the measured “present” situation (at 100%) (given in Table 1). The curve for O_3 is always similar to that of NO_2 due to the model formulation (Eq. 2). The curve for the temperature is from 0 °C to 37.2 °C.

Figure 5 shows the annual recession predicted by Eqs. (1, 2) and the assessed contribution to the recession due to the apparent single most influencing environmental parameters, particle matter (PM_{10}), nitrogen dioxide (NO_2) and relative humidity (RH), for the three locations. The values given by

the bars in the figure are also shown at 100% presence of that particular parameter in Fig. 4.

In Figs. 4, 5 it should be noted that the single environmental parameter effects are not additive, due to the exposure–response model formulation, and thus do not sum up to the total recession value. Figure 5 (1) clearly shows higher expected first year recession (for “All parameters”) at the urban traffic site (Msida), than in the urban background (Żejtun), and then in the rural background (Għarb) location. Similarly, the impacts of nitrogen dioxide (NO_2) and relative humidity (RH) at the stations decrease from the urban traffic site to the rural situation. With a hypothetical increasing relative humidity (RH), temperature (T) or nitrogen dioxide (NO_2) (above the values in Table 1) the influence of the relative humidity and temperature on the recession is however predicted to increase much more than that of nitrogen dioxide, until at $RH = 100\%$, its influence is expected to be about double that of nitrogen dioxide (NO_2) (Fig. 4). The influence of particulate matter (PM_{10}) is expected to be lower than

Fig. 5 Annual recession for Portland limestone obtained from exposure–response assessment (Eqs. 1, 2) based on data from three Malta locations with different environments (Table 1). **a** Absolute values (as shown at 100% in Fig. 4), and **b** the given parameter's share (%) of the total sum of the separate effects of all the environmental parameters in Eqs. 1 and 2



that of relative humidity (RH) and nitrogen dioxide (NO_2), and it decreases from the urban (traffic) to the urban background station, and then in the rural background. It should however be kept in mind that this evaluation, by Eq. (1), does not include the deterioration influences of sea salt in Malta (this will be discussed in the “Discussion” section). The impacts of sulfur dioxide (SO_2), rain and the acidity in rain (H^+) are low in all the three locations due to low values for sulfur dioxide and the expected relatively low acidity in rainwater (pH 6).

Discussion

It is a question how well Eqs. (1, 2), which were developed from European continental data, represent the situation in Malta, and how an assessment for Portland limestone would represent the ubiquitous Globigerina limestone used in Malta. Differences in the continental European and Maltese exposure environments and in the influence of the exposure environments on the weathering of Portland versus Globigerina limestone are considered below. The expected

weathering of limestone at the sites in Malta and European continental sites is then compared, and the potential environmental, including air pollution, influences on the surface recession of limestone inside and outside of the sheltered temples in Malta are discussed.

The composition of particulate matter (PM) in Malta as compared to continental Europe

Malta has significant, but in some cases declining, sources of anthropogenic dust emissions from quarrying, the construction industry, ship repair and servicing, power generation, road traffic and other sources. These include fine white dust from soft stone quarries and construction sites. In the area South-East of the Valletta harbour, where the Tarxien temples are located, elevated deposition of black dust, most probably coming from the now-decommissioned Marsa Power Station, has been measured in the past (Vella 2011). In addition, Malta has significant natural sources of particles, different from continental Europe, especially Sahara dust and sea salt, which would be measured as part of the particulate matter (PM_{10})

(ERA 2018). The weathering impact on limestone from this natural PM_{10} fraction could be different from that of anthropogenic PM_{10} . The influences of sea salt or chlorides are not specified in Eqs. (1, 2). In sea-exposed locations increased weathering due to deposition of sea salt aerosol is expected. This deposition would be higher close to the coasts (Grøntoft and Svenningsen 2009), especially where there is no sheltering from wind, by the terrain, vegetation or buildings (Gustafsson 1997). Sea salt contains chlorides and other ions, which can weather limestone by salt crystallization-dissolution processes, and this is common in the sea-exposed island environment and would be evident especially on the west coasts of Malta (Cassar 2010). Sea salt consists, mostly, of particles with significantly smaller diameter than $10\ \mu\text{m}$, but most of the mass concentration is in the super-micron range (coarse mode) (Yu et al. 2019). In coastal locations in Malta the mass contribution of sea salt to PM_{10} -particulate matter could be significant. Equations (1, 2) represents weathering by air pollution species including PM_{10} -particulate matter, with an unknown uncertainty in Malta different from the typical European continental situation, due to the combined effect with sea salt and exposure to different natural background particles such as Sahara dust. At the rural Għarb monitoring station, about 20% of the PM_{10} -particulate matter was measured to be Sahara dust from 2012 to 2013, and at the urban Msida station from 15 to 28% was measured to be Sahara dust between 2009 and 2015 (ERA 2018). In a recent study by Scerri et al (2018), $PM_{2.5}$ collected at the Msida station in 2016 was found by factor analysis to be composed of (by weight): Saharan dust (15%), traffic dust (27.3%), ammonium sulfate (23.6%), aged sea salt (12.7%), shipping emissions (5%), fresh sea salt (4.6%), fireworks (2.9%) and an unassigned portion (8.8%). Six of the sources (except the fireworks) were considered to be typical for the atmospheric composition of Mediterranean coastal traffic sites. The work also reports the major elemental composition in each of the factors. The Sahara dust could possibly contain less corrosive compounds than the typical European PM_{10} -particulate matter. Considerable amounts of aluminium, Al, (7%) and iron, Fe, (4.5%) and noteworthy amounts of phosphorus, P, (0.08%) have been reported in Sahara dust, with the deposited Fe and P representing 96% and 30–40% respectively of the total deposition of these elements at locations in Southern France and Corsica (Guieu et al. 2002). The Saharan dust may also, on average, form a relatively larger part of the PM_{10} -particulate matter in rural than in urban locations in Malta, as in the latter, local anthropogenic emissions are somewhat greater. The presence of Sahara dust would imply a possible risk for overestimation of the PM_{10} -particulate matter effect on the weathering [by Eqs. (1, 2)], as in the case of determining transgression of health limits (ERA 2018), and more so

in rural locations. Volcanic ash from Etna has also been known to reach Malta, most recently in 2013 (Azzopardi et al. 2013).

Comparison of the experimental recession of Portland and Globigerina limestone with the calculated recession of Portland limestone

The recession of Portland limestone calculated by Eqs. (1, 2) can be compared with direct measurements of the weathering of Portland and Globigerina limestone. Cabello-Briones (2015) and Cabello-Briones and Viles (2017) report results from exposures of similar quadruple sets of limestone samples inside and outside open shelters. Portland limestone was exposed at the Bishop's Palace in Witney, South-East England from August 2012 to July 2013, and Globigerina limestone was exposed at the Bishop's Palace and the Ħaġar Qim temple in Malta from August 2013 to July 2014. The uniform average surface recession, calculated from the reported annual weight change, sizes and densities of the Portland and Globigerina limestone samples, exposed at the Bishop's Palace from 2012 to 2014, is compared in Fig. 6.

Figure 6 shows a relatively small difference in the annual surface recession of Portland and Globigerina limestones, which is not significant within the limits of the calculated normal standard deviation shown in the Figure. The density of the Globigerina and Portland limestone samples exposed by Cabello-Briones (2017), were 1790 and $2100\ \text{kg/m}^3$, as compared to $2265\ \text{kg/m}^3$ for the Portland limestone exposed in the ICP-materials project as a basis for Eqs. (1, 2) (Yates 2007).

Cabello-Briones (2015) reported that their Globigerina limestone samples had a significantly larger open porosity (BS EN 1936: 2006) (31% as compared to 14%) and water absorption capacity at atmospheric pressure (15% as compared to 7%), than the Portland limestone samples. And, that the low water absorption capacity, low open porosity and high apparent density indicate that the (Portland) stone is less porous with a coarser pore structure and hence is more durable. She concluded that the test results for Globigerina limestone samples were very similar to chalk (“a particularly vulnerable chalk form the south of England”), which “indicates that they are more fine grained (than Portland limestone) and that a greater degree of weathering (under similar conditions) is expected in chalk, and Globigerina, which are less dense and with more and thinner pores”.

The comparable measured recession for Globigerina limestone in 2013–2014 with Portland limestone in 2012–2013 (Fig. 6) thus indicates some difference in the environmental conditions from 2012–13 to 2013–14. Cabello-Briones (2015) considered the freezing/thawing cycles and sodium chloride (NaCl) crystallization cycles events to be the most

Fig. 6 Surface recession (μm) calculated from measured weight change (%) of Portland limestone from 8/2012 to 7/2013 and of Globigerina limestone from 8/2013 to 7/2014, inside (centre) and outside the shelter at Bishop's Palace, Witney, England (Cabello-Briones, 2015). *SD* standard deviation

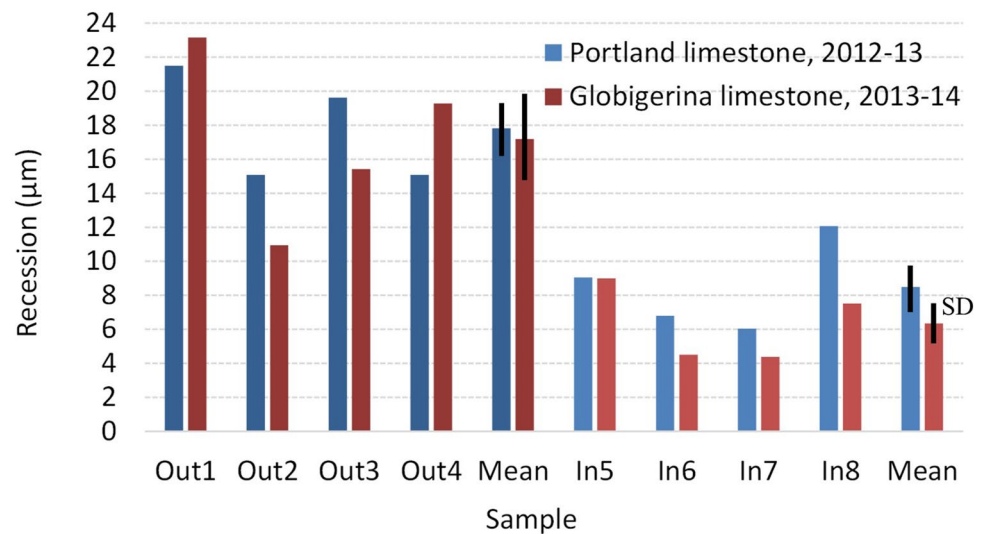


Table 2 Freezing and NaCl crystallization events at the Bishop's Palace in Witney, England, from August 2012 to July 2013, and from August 2013 to July 2014 (Cabello-Briones 2015)

	Outside	Periphery	Centre
Freezing events in a year			
2012–2013	81	43	39
2013–2014	17	5	9
NaCl crystallization events (mean number per day)			
2012–2013	0.84	0.76	0.79
2013–2014	0.9	1.1	1

important single climate influences on the weathering of the limestones at the Bishop's Palace. The numbers of these events calculated for the 2 years of exposures at the Bishop's Palace, are shown in Table 2.

The smaller number of freezing events and greater number of salt (NaCl) crystallization events in 2013–2014 than 2012–2013 correlates with higher reported mean daily winter temperatures and mean diurnal relative humidity (RH) in the winter of 2013–2014 than in the winter of 2012–2013 for that site. Due to the opposite effects on the overall weathering, it is difficult to know if the change in climate from 2012–13 to 2013–14 was increasing or decreasing the contribution to the sum total of the decay of the stone samples, and if this cumulative effect was different for Portland and Globigerina limestones. It could be hypothesized that the decrease in the freezing events was more important than the increase in salt crystallization events and reduced the measured weathering of the more vulnerable Globigerina limestone in 2013–2014 when compared to the Portland limestone in 2012–2013, but this would need verification. Figure 6 shows considerable variation between the recession of the quadruplicate of the

Globigerina and Portland limestone samples, respectively. The figure also shows that the outdoor decay of Globigerina limestone was measured to be greater than for Portland limestone for two of the quadruplicate samples (“Out1” and “Out4”), but oppositely, greater for Portland than for Globigerina limestone for the two remaining quadruplicates (“Out2” and “Out3”). This variation in the recession of the quadruplicate samples exposed outdoors, indicate that other differences between the samples may be relatively more important for their weathering than any systematic difference in the climate at the Bishops Palace from 2012–13 to 2013–14. In an ideal situation, it could be assumed that the micro-climate to which similar (here quadruplicate) samples are exposed is the same. If then, the climatic conditions fully explain the measured decay, the recession value for the four similarly exposed Globigerina and Portland limestone samples (respectively), and thus the difference between the measured decay (recession) of the Globigerina, and of the Portland, samples, would be the same. Assuming insignificant instrumental uncertainty in the weight measurements, the absence of such observed similarity in Fig. 6, indicates differences between the samples or their micro-climates. The purpose of exposing parallel samples is to obtain a measure of such variances, and to obtain more reliable values represented by the means.

Ideally, comparison of the recession of Portland with Globigerina limestone in Malta should be based on data from simultaneous parallel exposures of the different stone samples. Due to the difference in the physical properties of the two stone types, as explained above, their weathering in similar environments is not expected to be the same. However, the results from Bishop's Palace (Fig. 6) show a relatively similar initial weathering of these stones. Thus, when taking into account the differences between the general European and Maltese exposure environments, it should be

realistic to evaluate the variation in the weathering of Globigerina limestone in Malta by Eqs. (1, 2).

A further evaluation can be made by comparing measured values for the weathering of Globigerina limestone in Malta with the values calculated by Eqs. (1, 2), as given in Fig. 5. Figure 7 shows the uniform average surface recession of Globigerina limestone exposed at the Ħaġar Qim temple, calculated directly from the physical sample properties and the measured experimental weight change reported by Cabello-Briones and Viles (2017). This evaluation does not consider the possible influence of the deposition of particles or aerosol on the reported weight loss and recession values of the samples.

Figure 7 shows average recession ranging from $2.9 \pm 0.7 \mu\text{m}$ outside the shelter to $6.2 \pm 1.1 \mu\text{m}$ inside the shelter, with a difference of about three μm between the outside and inside location. Cabello-Briones and Viles (2017) state that the difference in the measured weight loss for the Globigerina stone samples exposed outside and inside of the shelter was not significant. A *t* test performed here showed a less than one per cent (0.5%) chance that the reported values for the two samples sets (of four replicas) would have the same mean. It should be noted that, contrary to the situation at Bishop’s Palace (Fig. 6), the recession was observed to be greater inside than outside the shelter of Ħaġar Qim temple. This will be discussed below. The recession values in Fig. 7 are in a similar range, and inside the temples within nearly exactly the same range, as was predicted by Eqs. (1, 2) for the outdoor weathering of Portland limestone at the three air quality measurement locations (from 6 to $6.8 \mu\text{m}$, Fig. 5).

The expected underestimation of the surface recession by Eqs. (1, 2) (Fig. 5) as compared to the measured values for Globigerina limestone (Fig. 7) does not appear, probably due to differences in the average continental European

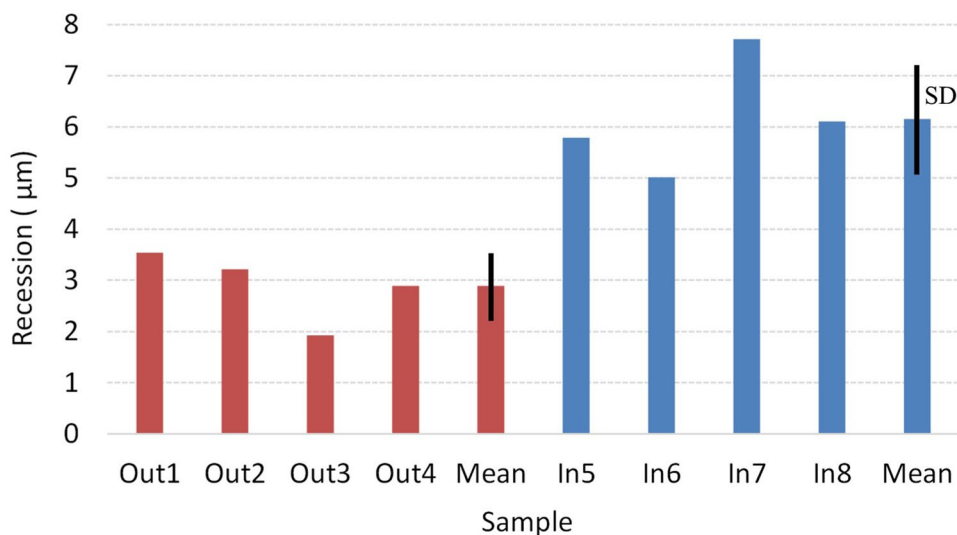
and Maltese exposure situations, with more natural and less corrosive airborne particles in Malta, as was already explained above.

Comparison of limestone weathering between sites in Malta and with European continental sites

Due to the westerly winds and coastal location a greater presence of wind-carried sea salt aerosol is expected at the Għarb than Msida and Żejtun stations, even if the total PM_{10} -particulate matter is less. The sea salt can accumulate on surfaces and because of its small size, and when it dissolves in surface water, it can be transported deeper into the rock than other dust particles, and have the effect (contrary to that of Sahara dust) of accelerating the weathering compared to values calculated from Eqs. (1, 2). The possible impact of volcanic ash from eruptions of Etna is unknown. Differences in wind speed and direction between locations would be important for sea salt transport and erosion effects.

The measured weight loss for the Globigerina stone samples exposed outside the shelter of Ħaġar Qim (of $2.9 \mu\text{m}$, Fig. 7) is slightly lower than the average first year background recession for Portland limestone in continental Europe, evaluated to be $3.2 \mu\text{m}$ in the ICP-materials project (CLRTAP 2014). Ħaġar Qim is a typical rural background site. The first year limestone recession calculated by Eqs. (1, 2) for Malta in 2017, of $6\text{--}7 \mu\text{m}$ (Fig. 5), is in the low range of values recently measured for Portland limestone (for the year 2011–2012), for 13 continental European sites in the ICP-materials project (Tidblad et al. 2014). These are typical values for rural sites, but also common in urban sites since about the mid-1990s, when the sulfur dioxide (SO_2) concentration in air had in most locations dropped to below $10 \mu\text{g}/\text{m}^3$ (Tidblad et al. 2017). The calculated difference between the urban (with traffic) (Msida) and rural (Għarb) locations

Fig. 7 Annual recession of Globigerina limestone exposed inside and outside the shelter of Ħaġar Qim temple in Malta, recalculated from weight change values reported by Cabello-Briones and Viles (2017). *Out* outside, *In* inside shelter *SD* standard deviation



in Malta, of 1 μm , is small and well within the typical measured yearly variation for single European sites (Tidblad et al. 2017). Although air pollution would generally contribute to slightly higher weathering rates of limestone buildings in the urban areas of Malta, the annual variation between sites due to natural climatic factors could well be larger in the present air pollution situation. Annual climate statistics for Malta between the year 1956 and 2000 show, for example, a variation in the annual average mean relative humidity (RH) at 0000 UTC (Universal Time) and noon 1200 UTC, of 78–87% and 57–68% (Environment statistics 2002). Ignoring for the moment possible correlations between the relative humidity and other parameters in Eqs. (1, 2), and keeping the values for the other parameters constant (according to Table 1), a 10% variation in relative humidity (RH) for the Msida location, from 72 to 82%, would according to Eqs. (1, 2) give a variation of 1.6 μm in the annual recession of the Portland limestone (from 6 to 7.6 μm). This could probably represent the approximate variation also for Globigerina Limestone. The construction of the shelter and its openings would influence wind parameters such as turbulence, and the wind transport of sea spray into the shelter, and this is currently being measured, monitored and modelled.

Environmental influences on the surface recession inside and outside the shelters, including air pollution

Cabello-Briones and Viles (2017) discuss the possible reasons for the observed difference of about three μm (Fig. 7) in the weathering between the stones exposed outside and inside the shelter of *Ħaġar Qim*. This difference is larger than the expected difference of about 1 μm (Fig. 5) for the first year weathering between Maltese urban and rural locations due to “continental air pollution”. The greater weight loss inside than outside the shelter could be explained by a higher number of annual relative humidity events crossing the 75% threshold for crystallization of sodium chloride (NaCl), inside the shelter. At present, however it seems uncertain what the general effect of the sheltering is on the number of crystallization events. Becherini et al. (2016) report a reduction in the risk of NaCl and epsomite ($\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$) crystallization events inside the shelters of the *Ħaġar Qim* and *Mnajdra* temples, whereas Cabello-Briones and Viles (2017) report an observed 13% higher number of annual NaCl crystallization events inside (812 events) than outside (707 events) the *Ħaġar Qim* shelter.

Cabello-Briones and Viles (2017) observed less variation in relative humidity inside than outside the shelter at *Ħaġar Qim*, with values mostly in the range of 70–80%. Also, the surfaces of samples exposed outside were observed to become yellower than inside, probably due to chemical reactions of the chromophores in mineral phases with oxidized

ferric compounds, which occur in significant amounts in the *Globigerina* limestone (Cassar 2004). No significant difference in the change in surface hardness was observed. It seems however not known if, and possibly how, such surface alterations affect the surface weathering of the limestone. The rates of surface oxidation processes are expected to be faster in the warmer and more sun-exposed situation outside, where ozone concentrations would probably also be higher. Tropospheric ozone is produced by photo-catalytic processes in the atmosphere and its concentration is generally higher in Southern than Northern Europe. Even if air exchange in the open shelter would be quite high, ozone concentrations are expected to be somewhat lower inside due to the absence of sunlight and the larger available shelter material surfaces for its deposition (relative to the air volume).

It is uncertain if the pollution influence on the weathering, as calculated by Eqs. (1, 2), would be different outside and inside the shelter. It seems a reasonable first hypothesis that the air pollution weathering inside the shelters would be somewhat lower than outside of the shelters, depending on their location in urban or rural areas (Fig. 5). The possible changes in the concentrations of the influencing air pollutants, from outside to inside the shelters, and also seasonal variations, should be evaluated. As rain and sulfur dioxide (SO_2) would have little influence on the weathering values calculated inside the shelters from Eqs. (1, 2) (see Fig. 4), these would depend on the concentrations of nitrogen dioxide (NO_2), ozone (O_3), the relative humidity (RH) and temperature (T), and on the particulate matter (PM_{10}). Nitrogen dioxide is less reactive than ozone, and its concentrations would change less. Nitrogen dioxide concentrations are affected both by homogeneous reactions with sunlight and ozone, which in warm and sunny climates can increase indoor concentrations compared to those outdoor (Brimblecombe et al. 1999; Weschler and Shields 1997), and by surface deposition (Grøntoft and Raychaudhuri 2004). Thus, the concentration of nitrogen dioxide (NO_2) will probably decrease more inside the shelters in winter than summer, when it could even increase. Cabello-Briones (2015) report that the daily mean temperatures are somewhat higher and diurnal temperature ranges somewhat less inside than outside the shelter at *Ħaġar Qim* both in the summer and winter. It is further reported that the diurnal relative humidity range is much higher outside than inside the shelter at *Ħaġar Qim* and that the daily mean relative humidity is higher inside (72%) than outside (62%) in the summer, but lower inside (82%) as compared to outside (91%) in the winter. The colder winter and also relatively colder summer nights than days give more surface wetness at these times. In the summer when the temperature is the highest, the relative humidity is thus usually above 60%, which will result in pollution weathering (according to Eqs. 1, 2), whereas the relative humidity is often below 60% in the warmer daytime in the other seasons. The rates of weathering by air pollution

increases with air humidity and temperature (Fig. 4). The effect on the air pollution weathering, of wetting by condensation at drops in the temperature, is uncertain. Higher concentrations of ozone (O_3) in the summer, relatively higher concentrations of nitrogen dioxide (NO_2) inside than outside the shelter in the summer than winter, and higher inside than outside values of relative humidity in the warmer summer, would probably give relatively more nitric acid (HNO_3) in the air inside than outside the shelter (I/O ratio) in the summer than winter, and, generally, more pollution weathering.

As regards direct NO_x (nitrogen dioxide, NO_2 , + nitrogen monoxide, NO) emissions, these are usually higher in urban locations (such as Msida) than rural locations (such as Għarb) (Table 1). The higher NO_x emissions in urban locations would generally contribute to lower ozone (O_3) values, as the ozone reacts with the emitted nitrogen monoxide (NO) to create nitrogen dioxide (NO_2). The concentration of nitrogen dioxide (NO_2) can be higher in winter than summer due to larger NO_x emissions, from combustion sources and less splitting in sunlight, and reaction to ozone (O_3). The higher outdoor winter concentrations of nitrogen dioxide (NO_2) would then be transported into the shelters, depending on the direction of the wind. Downwind from emission sources, such as for the situation in Tarxien with black dust from the, now demolished, Marsa power station (Vella 2011), episodes with elevated concentrations of both nitrogen dioxide (NO_2) and ozone (O_3) could also occur. This would be so when the nitrogen monoxide (NO) has already been consumed in its quick reaction with ozone (O_3) to nitrogen dioxide (NO_2), and the nitrogen dioxide which is transported to the area then contributes to increasing ozone concentrations by reaction with the sunlight and oxygen (O_2). In such situations, the winter load of pollutants inside the shelters could increase. The air pollution weathering inside the shelters would further depend on variations in the outside to inside ventilation of particles into, and their resulting concentration and composition in, the shelter.

Overall, however it seems likely that the average levels of air pollution and then the influence on the weathering would be less inside than outside the shelter. The shelters would, nonetheless, probably give less protection against air pollution weathering on warm, humid and windy summer days, when air pollution ventilated into the shelter readily reacts on the stone surfaces. The air pollution weathering would probably be at its maximum on such humid summer days and on humid winter days at locations with elevated outdoor concentrations of nitrogen dioxide (NO_2). The Tarxien temple site, which is located close to Valletta and itself is an urban area, would probably be more affected by anthropogenic air pollution than temple sites in rural locations. For a better assessment, by Eqs. (1, 2), it would be necessary to do simultaneous annual measurements of air pollution, temperature and relative humidity, both inside and outside a shelter. These measurements are being planned.

The relative humidity is clearly a critical factor for initial limestone weathering, as it is, in addition to other influences like condensation, wetting and drying, a determining factor for salt crystallization cycles and for the general influence of air pollution (Fig. 4). The weathering effects of the salt and pollutants would depend on their presence on the stone surfaces and in the stones. Galea et al. (2015) measured the concentration of water-soluble ions in dust deposited on the exposed and sheltered temple of Ħaġar Qim. The results showed less chloride deposition on sheltered than exposed areas, probably due to deposition on the shelter fabric. Cabello-Briones and Viles (2016) also stated that a greater measured first year weight loss inside than outside the shelter may be due to the added weight of deposited salts on the stones outside of the shelter. Dry deposited pollutants and salt accumulating on surfaces, inside and out, would vary with varying environmental conditions. This includes the wind conditions and is currently under evaluation in a separate research project. It should also be noted that salt crystallization and transport processes proceed in very complex ways (Cassar et al. 2018). This too forms part of a current research project probing the mechanisms of damage on the limestone megaliths.

Based on the literature, it is difficult to determine whether the greater first year weight loss observed inside compared to outside the shelter at Ħaġar Qim by Cabello Briones and Viles (2017), is due to changes in humidity cycling inside the shelter, changes in salt crystallization-dissolution cycles, and/or other impacts on the stones, such as possibly surface oxidation and more salt aerosol deposition outside. Cabello-Briones and Viles (2017) state that a longer set of exposure trials would be needed to provide information on salt content and a definitive assessment of the impacts of the shelters on limestone deterioration. This assessment is currently underway, and the gathering of more pollution data inside and outside the shelters is also planned. The evaluation of the data for air pollution and its influence on the weathering does not change the hypothesis that differences in weathering inside as compared to outside the shelters are today probably mainly explained by the influences of climate (precipitation, wind, RH and T), salt aerosol, and possibly light, rather than anthropogenic air pollution. The levels and fluctuations of relative humidity (RH) inside the shelter are critical for the weathering of the limestones as these determine both the salt crystallization events and pollution influences.

Conclusion

The data and evaluations presented in this work support a hypothesis suggesting that differences in levels and fluctuations of relative humidity, and presence of salts on and in the limestone megaliths, inside and outside open-sided shelters over

the Prehistoric temples in Malta account for more of the variations in the first year weathering of Globigerina limestone than variations in air pollution. Observed first year weight changes inside as compared to outside the shelters cannot be explained by air pollution effects alone, but more likely due to differences in salt aerosol deposition and changes in salt-crystallization events due to changes in relative humidity fluctuations, related also to temperature (and possible condensation events), wind parameters and rainfall, as well as ground water replenished from areas beyond the shelter. To evaluate the air pollution influences on limestone weathering inside the shelters, measurements of air pollutants inside and outside one or more of the shelters are needed and indeed planned. The atmospheric chemical weathering of limestone in rural locations in Malta is similar to the calculated European continental background and rural situations. The present variation in the first year weathering recession of limestone between urban and rural locations in Malta due to air pollution was calculated to be about one micrometer loss of stone surface, which is probably less than annual variations due to the influence of natural climatic factors, and small compared to present European geographical and annual variations for rural and urban sites, and hence can be probably be considered as near negligible.

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