Effect of Khoms power station stack emission impact on the ancient city of Leptis Magna, Libya

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Abstract

It is well established that emissions from combustion of heavy fuels, light fuels and natural gas cause negative impacts on surrounding environment, including the erosion of antique buildings. This study investigates the effect of emissions from the Khoms Steam and Gas power station on the neighboring ancient city of Leptis Magna, using an air dispersion software (DISPER) to calculate the concentrations of sulfur oxides and nitrogen oxides in archaeological city under the influence of climate variables. The objective of this study is to calculate marble erosion rates caused by the chemical reactions on the surface of selected Marble structures. The study reveals high concentrations of the targeted pollutions on the surface's columns & status of the city.

Keywords: Air Pollution; Power Station; Leptis Magna, Ancient City.

Introduction

The ancient city of Liptis Magna was established by the phoenicians as a commercial station during the first millennium BC. The oldest monuments date back to the sixth century BC. This city became the capital of the Roman Empire, after the enthronement of Emperor Severus at 9 April 193. This city is considered one of the most important archaeological sites in the world, due to its invaluable treasures and diversity in the walls and columns engravings. Therefore, the UNISCO has recogonized this city as a global human heritage since 1982 (Abu Arabiya, *et al*. 2004).

The limestone sand rocks are the main material used the city which were fetched from the formation of Gargaresh - Karrout. These formation consist of fragmentation and accumulation of marine shells mixed with sand in various proportions that make the nature of rocks soft and weak-fleshed (Garrison, 2003). These rocks spread along the coastal strip surrounding the city of Lebda, where it differs in the thickness of its detectors, the degree of their cohesion, and their chemical components from one location to another (Al-Khalisi, 2012). The studies notes that as the topography decreases, the use of schist columns increases. The increase in the use of granite columns is accompanied by an increase in the use of limestone in the city walls, and the older buildings, the greater uses of calcarenite rocks and the less use of dolomite limestone and the schist in the archaeological city (Garrison, 2003). There were three main factors responsible for the nature of the distribution of the different rock blocks of the rock structure, these were represented by theology, time, and topography (Minas, 2003).

Coastal archeological cities are constantly exposed to erosion factors that are controlled by the laws of nature, causing different degrees of damage severity. One of the biggest causes of damage to these buildings is the interaction between the archaeological material of rocks and natural erosion (Shushan, *et al*. 2019). The damage of marble pillars and statues increase in presence of polluted gases, due to its composed of calcium carbonate ($CaCO_3$). The carbonate material is strongly affected by sulfur and nitrogen oxides, especially in the presence of high levels of air humidity or rain (Boden, 1989). For example, sulfur dioxide and water in the air react with limestone to form calcium sulfate $CaSO_4$ and gypsum ($CaSO_4.2H_2O$). This process cause erosion of building blocks and mortar used in the cohesion of building blocks. Moreover, calcium sulfate can permeate the pores of limestone and crystallize again and expands to cause more corrosion and weaken rocks (Tian, *et al*. 1999). It is known that Bricks are not affected by sulfur dioxide, however, the binder materials in the bricks are not. They are made of sand, calcium carbonate, and calcium hydroxide. These materials can be broken down by acid pollutants, in general, carbonate bonding materials are more likely to be penetrated than sandstone (UKBERG, 1990; Lipfert, 1987).

Several studies conclude that corrosion effect caused by sulfur dioxide on calcite-containing marble occurs at higher rates than corrosion of dolomite-containing marble and the corrosion increases significantly at higher humidity levels (above 70%) (Lan, *et al*. 2005). Sulfur dioxide (SO_2) is highly reactive and corrosive causes acid rain formation that damages building materials and structures (Gandhi, *et al*. 2017; Zhang, *et al*. 2017 & Duan, *et al*. 2016). The pH of rain is reduced as low as 5.6 when exposed to acidic pollutants such as sulfur and nitrogen oxides (Dondapati, *et al*. 2013). Acid rain is harmful to cultural heritage, especially the outdoor marble and bronze sculptures (Livingston, 2016).

The term acid deposition is more accurate than acid rain, and this deposition consists of two main types (dry and wet). Dry deposition refers to the deposition of polluted gases through dissolving in moisture and particles in the absence of rain, and wet precipitation relates to the bonding and dissolving of pollutants in clouds or raindrops. Dry deposition is more important than wet deposition in highly polluted areas (Furlan & Girardet, 1983). The main effect of sulfur dioxide on limestone is the formation of crusts and the loss of materials due to dissolving, which can represent 30-50% of building material damage (Pérez Bernal & Bello, 2003), damage can also occur when formed crusts reach a certain thickness and then fall from the surface of the stone (Camuffo, *et al*. 1983). The stone surface in which the veneer is separated usually turns fractional, porosity higher area than the original stone, and becomes weaker in additional weathering operations (McGee & Mossotti, 1992).

Many studies reported that the effects of nitrogen oxides on building materials have increased dramatically in recent years (Hilmi, *et al*. 2013). Despite, the relatively low concentrations of sulfur dioxide, exposure to 10 ppm of SO_2 for a year can cause an effect of erosion from 1.9 µm to 4.5 µm as a result of the dissolving limestone from the original marble (Yerrapragada, *et al*. 1996). In different places of the archaeological

European sites, it was found that the formation of gypsum is the primary cause for demolition. The second step in the demolition process is a reaction between gypsum and calcium aluminum hydride (Sabbioni, *et al.* 2001; Sabbioni, *et al*. 2002; Viana, *et al.* 2014).

Natural resources contribute about 66% of sulfur amount entering the atmosphere, while human activities contribution were 33%. However, contribution of human resources increases annually due to increases in energy demand and mineral ores (Al-Sattouf, 1995). The combustion of waste and fossil fuels are the main causes of acid rain (Gandhi, *et al*. 2017). For example, electric power plants contributed by 70% of nitrogen oxides that emitted from industry and the contribution of industrial furnaces were 30% (Raczynski & Watson, 1999). Despite, the electric power station emissions have clear negative effects on walls and columns engravings of Liptis Magna city, this problem is still a matter of public opinion only. This research aims to identify the gaseous and PM emissions (solid pollutants) and investigate their effect on the integrity of the archeological structures.

Materials and Methods

Site of study area

The ancient city of Leptis Magna is located on the northwestern coast of Libya at the mouth of Wadi Lebda, 3 km east of Khoms city, between the latitudes 35° 55' 84" -35° 35' 25" N and longitude 18° 15' 23.13" - 18° 15' 01" E

Figure (1): Study area locations in the ancient city of Leptis Magna, (A = Power station, 1 = Theater, 2 = Market, 3 = Septimius Severus Arch, 4 = Temple of Jupiter, 5 = Church, 6 = West Gate (Oia Gate), 7 = Hadrian Baths, 8 = Nymphaeum, 9 = Severan Forum, 10 = Amphitheatre).

Dispersion of pollutants

Assessment of SO_2 and NO_x emissions over the study site was carried out using utilizing a dispersion modeling software "DISPER V: 4.0, which was produced by Canarina Environmental Software Company in the year 2007, this software is based on Gaussian model to represent the spread of pollutants, it is assuming that the emissions from the source are continuous and constant. The program has the ability to project data on geographical or satellite maps at different scales with the use of Cartesian coordinates GPS, the software program also deals with a constant speed and direction of the wind in each estimation process, and it is characterized by being able to deal with several emission sources at the same time, and the program can be used to calculate average concentrations resulting from the conditions Different climatic conditions and therefore can be used in the analysis of the spread using different repetitions of wind directions and speeds.

DISPAR software was used in this study to estimate the spread of Nitrogen and sulfur oxides emitted from Khoms power station under the influence of local climatic conditions in Khoms meteorological station, by used of maps and satellite images (Google earth) of the site, after determining the location of the power station, and the important sites in the archaeological city on the map as shown in Fig. 1.

Source of pollutants

The diffusion simulation process is based on emission rates of sulfur and nitrogen oxides obtained from the simulations of the steam and gas power stations mentioned in (Ibrahim et al 2012), in addition to real data on the dimensions of their chimneys as shown in Table (1).

Table (1): Emission Rates used in the propagation simulation (Ibrahim et al 2012).

The source	Location		Chimney Dimensions*		Emission** (g / s)	
	east	North	Height (m)	Diameter (m)	NO_x	SO_2
Gas power station	14.328025	32.623128	30	6	3179.2	-
Steam power station	14.331181	32.621387	100	5	2130.7	806.2

* Four stacks for each station.
** Average emission rates of a stacks.

Meteorological data

The study area is generally characterized by a moderate climate, where the average daily temperatures are 17^o in winter and 26^o in summer, with an average rainfall of about 17 mm/day in the months from September to March, with annual rainfall of about 285 mm. The winds are northern and northwestern in the months from October to April, where the Gibli winds blow from the east and south during the summer months (from June to August) (Table 2). The climatic data used in this study represent fifteen years records from 1/1/2004 to 12/31/2018 obtained from the meteorological station located in Khoms, No. (62012), with consideration the frequency of winds heading towards the ancient city from the side of the power station.

Table (2): Summary of climatic data used in the study.

month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Temperature (°C)	12.7	13.5	15.7	18.3	21.5	24.5	24.5	27.3	26.2	22.7	19.3	14.3
Wind speed (km/hr.)	13.4	15.2	14.4	14.8	14.3	12.7	11.8	11.7	10.8	11.5	12.0	15.1
Sunny hours (hr./day)	5.8	7.1	8.3	8.0	10.0	11.0	12.2	10.9	9.2	7.7	7.3	5.8

Corrosion rates calculation

There are many empirical equations for calculating marble corrosion rates caused by air pollutants, the chosen equation was the one developed by (Roots, 2008), because it includes the effects of pH of rain, moisture and concentrations of acidic pollutants as sulfur and nitrogen oxides.

$$R = 3.1+ [0.85+0.0059 [SO_2] RH_{60}+0.054 \text{ rain } [H^+]+0.078 [HNO_3] RH_{60}+0.0258PM_{10}]t$$

Where: R = annual corrosion rate (µm/year); RH = relative humidity, Rain = amount of rain (mm); $[H^+]$ = hydrogen ion concentration in rain water; $[SO_2]$ = sulfur dioxide concentrations (micrograms/m^3); and $[HNO_3]$ = Nitrogen Oxide Concentration (micrograms/m^3)

X-ray fluorescence testes

Thirty collected samples from different areas in the ancient city represent different types of rocks constitute the archaeological columns and statues in the ancient city. The rocks are marble, granite, schist, limestone, sandstone by three replicates for each type from the original sample before affecting it, and the crusts which expected to be caused by the change in Chemical composition, samples were examined by X-ray fluorescence device.

Results and discussion

Concentrations of pollutant in the ancient city

The results fig. (2) shows that the concentrations of nitrogen oxides under the effect of winds blew from the side of the power station over the ancient city are maximal in the vicinity of the Amphitheatre of the ancient city reaching 6509 µg/m^3, whereas the concentrations west of the Amphitheatre are lesser ranging from 4340 - 3255 µg/m^3 .

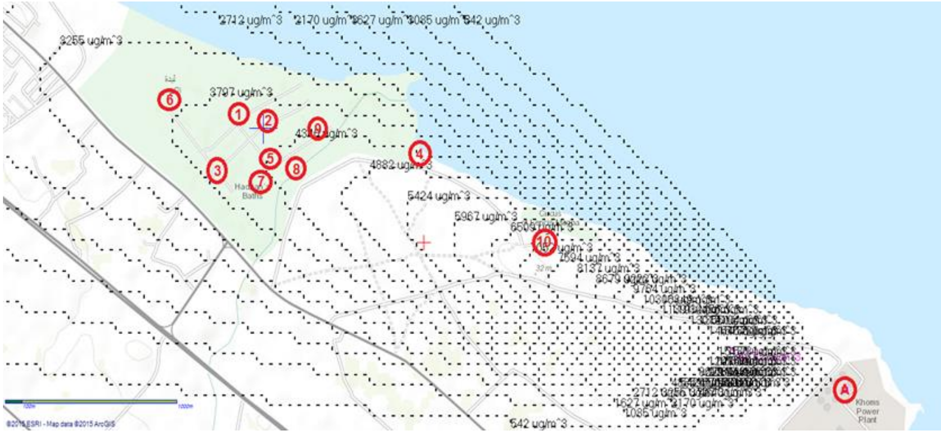


Figure (2): Concentrations of nitrogen oxides in the vicinity of the power station and the ancient city of Lebda.

Similarly, calculated sulphur dioxide concentrations in the air are also found to be excessive reaching 976 micrograms/m^3 in vicinity of the amphitheatre and ranging between 651-488 micrograms/m^3 in the central area of the ancient city (fig. 3). The excessive concentrations of sulfur and nitrogen oxides in Leptis Magna can be attributed mainly to proximity of the city to the power plant. It is worth noting that this rise in concentrations of polluting gases in the study area occurs only during the times that wind blows from the side of the power station towards the city, that constitutes 16% of year times. Raczynski and Watson (1999) indicate that high concentrations of sulfur dioxide and nitrogen oxides gases can be felt in the atmosphere by smell if their concentrations reach of 1,000–3,000 micrograms/m3 respectively. Based on our field observation, the locals live near the power station confirm the presence of sharp and unpleasant odors in the air coming from the power station direction which enhances the validity of the results.

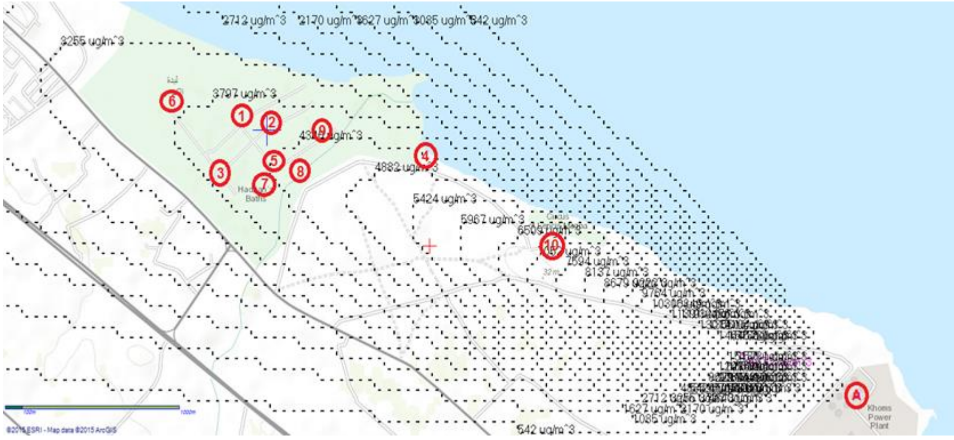


Figure (3): Concentrations of SO_2 in the vicinity of the power station and the ancient city of Lebda.

The expected corrosion rates

Marble rocks are selected as a model for investigation as the prominent features of the archeological structures of marble. The monthly rates of marble erosion are determined using the concentrations of sulfur dioxide and the nitrogen oxides found on its surface through the process of treating the spread using the DISPER program, and the marble erosion equation (Roots, 2008). The results show a clear effect of pollution resulting from the two power plants on the marble pillars of the ancient city (Fig. 4)

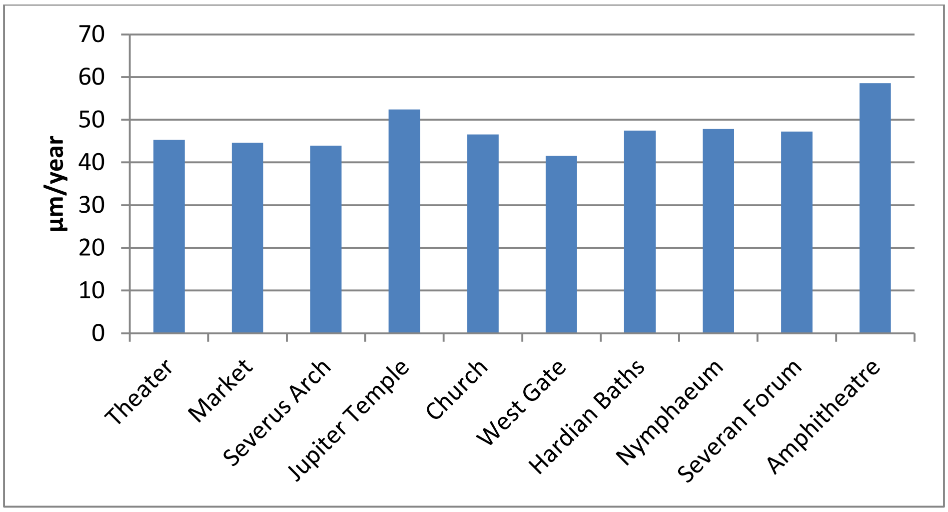


Figure (4): Annual rates of corrosion in marble in the ancient city of Lebda under the influence of stack emissions.

Due to its alkaline nature, composed of $CaCO_3$, marble can strongly be affected by acidic air pollutants such as Sulphur and nitrogen oxides especially in presence of high levels of humidity. Exposed marble surfaces turn into sulphate salts and nitrates resulting in corrosion as follows (Boden, 1989):

$CaCO_{3(s)} + SO_{2(g)}$-------- $CaSO_{4(s)} + CO_{2(G)}$

$CaCO_{3(s)} + 2NO_{2(g)}$------- $Ca(NO_3)_{2(k)} + CO_{2(g)}$

According to the first equation, sulfur dioxide reacts with marble to form gypsum (calcium sulfate) which has a larger size than the original marble, causing cracks and scaling of marble columns and statues. Similar reaction occur on the surface of limestone and granite as is showed in figures (5-9). Reaction products gypsum and calcium nitrate are more soluble in water than Calcium carbonate which facilities the erosion by rain water. It is also noted that the presence of nitrogen oxides is causing corrosion by dissolving marble as calcium nitrate, that leads to an increase in the effect of sulfur oxides through the process of oxidation.

The obtained results are consistent with previous finding by results of Abu Arabia and others in the year (2004), who found that the marble columns and granite of the ancient city are noticeably exposed to severe erosion.

Figure (5): picture of scales formed on marble under the influence of air pollution.

Figure (6): Erosion of a marble column forming crusts under the influence of air pollution.

Figure (7): image of a statue carved on limestone that has been damaged due to chemical weathering, with the loss of at least 2 mm of its surface.

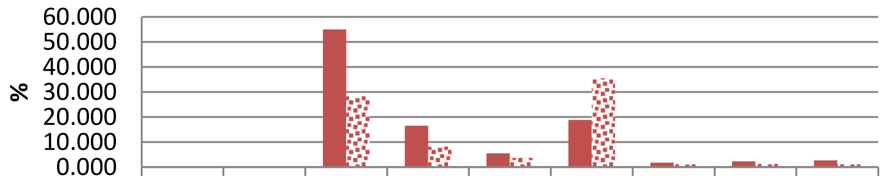
Figure (8): image of statues carved on marble, which have started to be damaged by pollution.

Figure (9): picture of the granite columns exposed to damage due to chemical weathering, direction of damage to the right of this image where the power station is located.

XRF sampling analysis

Granite samples

Fluorine X-ray analysis of granite samples and the corresponding crust samples fig. (10) reveal that, granite samples originally consist of silicone dioxide (55%), calcium oxide (18.85%), and aluminum oxide (16.00%). Different weathering processes on the rock surface have led to a noticeable decrease in the percentage of the main constitutes of the original rock such that SiO_2 (28.5%, Al2O_3 (8.28%), whose due to the fragmentation of the rock and the fall of some of its constituent compounds or its volatilization with the wind and other erosion factors. It can also be noted that calcium oxide in granite crusts was increased (35.575%), which indicates the presence of insoluble calcium chloride salts, the possibility of chemical weathering of other rocks that are insoluble compounds. This hypothesis is supported by an increase in the sulfur oxide content in the crust samples (0.175%) compared to the original granite samples (0.02%). Likewise, with regard to chloride, its presence was 0.03% in the original samples and this percentage reached 0.11% in crust samples.

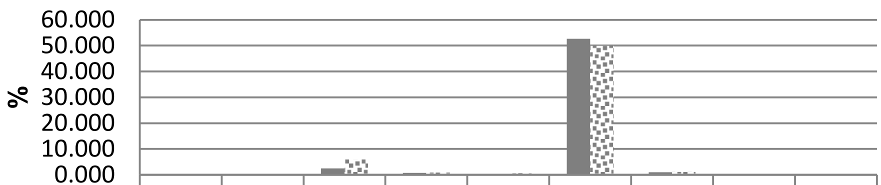


	SO3	Cl	SiO2	Al2O3	Fe2O3	CaO	MgO	Na2O	K2O
Rock	0.020	0.030	55.100	16.530	5.620	18.850	1.890	2.350	2.770
Crusts	0.175	0.110	28.445	8.280	3.770	35.575	1.205	1.300	1.175

Figure (10): Percentage of main elements present in a granite sample and a sample of crusts formed on it

Marble samples

A number of marble samples and the corresponding crust sample examined by XRF, fig (11) reveal that the main component is calcium as CaO. Its presence in the original samples is 52.637% of the total components of the sample. It is also noted that this percentage is reduced to 49.962% in the crust samples taken from the same marble samples, this decrease can be mainly attributed to Chemical weathering processes caused by the formation of calcium sulfate. Despite being poorly soluble in water (Ksp $_{(CaSO4)}$ = 4.93x10^{-5}), it remains more soluble than the compounds of calcium carbonate (Ksp $_{(CaCO3)}$ = 3.36x10^{-9}). The presence of sulfur compounds in the crust samples is 0.088%, which are rare in the original samples.

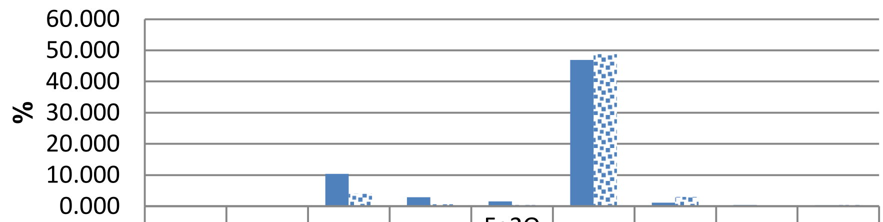


	SO3	Cl	SiO2	Al2O3	Fe2O3	CaO	MgO	Na2O	K2O
Rock	0.002	0.002	2.453	0.753	0.250	52.637	0.952	0.063	0.085
Crusts	0.088	0.088	6.015	0.937	0.672	49.962	1.152	0.073	0.182

Figure (11): Percentage of main elements present in a marble sample and a sample of crusts formed on them.

limestone samples

Calcium compounds are the main component of limestone samples, where the percentage of their presence in these samples is approximately 47% of the total components as shown in Figure (12), which also shows the presence of silicon compounds in the original limestone samples by more than 10% of the total components, the components change slightly in the crust samples compared to the original samples where the calcium compounds increase in the crust samples and reach 48.813% while the presence of silicon compounds is reduced to 4.06%. Changes in chemical composition between the original samples and the crust samples can be attributed to the effects of chemical reactions and weathering processes, which confirms this hypothesis a noticeable decrease in the percentage of the presence of compounds of some other elements in the crust samples compared to the original samples. Sodium compounds, for example, decreased from 0.383% to 0.042%, as is the case with silicon, aluminum and iron compounds, while magnesium compounds increased relatively from 1.149% to 3.043%, as well as sulfur compounds, their presence increased from 0.011% to 0.232%, this could be due to the formation of magnesium sulfate and calcium sulfate.

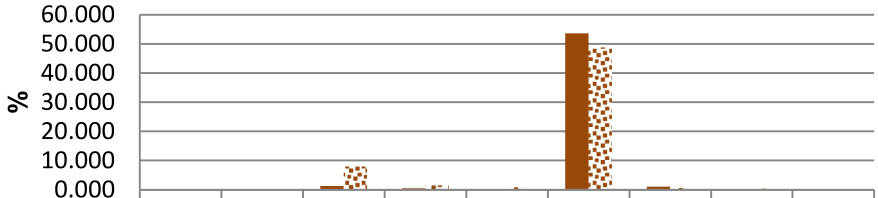


	SO3	Cl	SiO2	Al2O3	Fe2O3	CaO	MgO	Na2O	K2O
Rock	0.011	0.036	10.403	2.861	1.596	46.937	1.149	0.383	0.357
Crusts	0.232	0.063	4.060	0.607	0.412	48.813	3.043	0.042	0.393

Figure (12): Percentage of main components present in limestone samples and crust samples formed on them

Samples of schist

The results of analysis of schist samples shown in Figure (13) appears that more than 53.5% of the components of the original sample consist of calcium compounds, these compounds decreased in crust samples (crumbs) to 48.75%, this decrease can be attributed to weathering, fragmentation and volatilization with winds for schist samples, the samples were taken from areas that are relatively far from the sea and have little exposure to rain water, therefore the relative decrease in the effect of the melting process on the original rock samples, this hypothesis is reinforced by the noticeable increase in the presence of the compounds of the most soluble elements such as sodium, potassium, aluminum and iron, which rose from 0.16% to 0.35%, from 0.02% to 0.04%, from 0.55% to 1.46% and from 0.2% to 1.46% for each of them respectively, also the absence of chloride salts in the samples indicates that rock samples were far from the sea, which means not being exposed to sea spray, chloride salts were observed in the samples of the other types of rocks studied, however, the presence of a relative increase in the presence of sulfur salts in the crust samples compared to the original samples can be considered an indication of their exposure to chemical weathering processes as a result of contamination with sulfur oxides that dissolve in air humidity water, then deposited on the outer surface of the rocks.

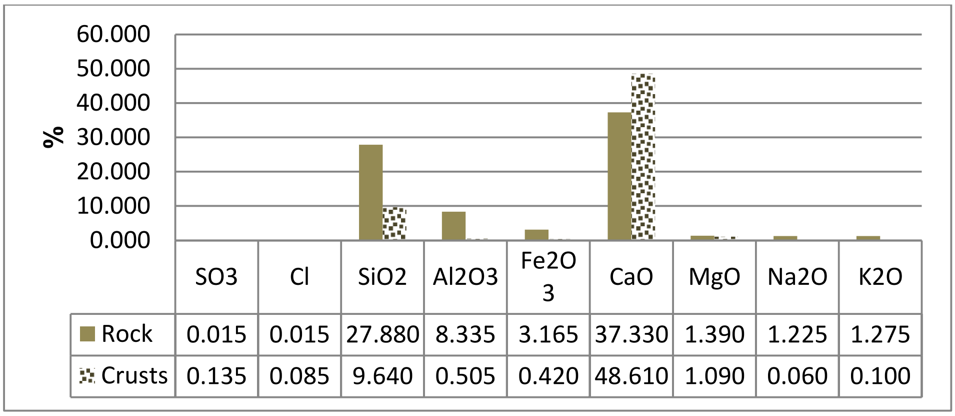


	SO3	Cl	SiO2	Al2O3	Fe2O3	CaO	MgO	Na2O	K2O
Rock	0.001	0.000	1.300	0.550	0.200	53.540	1.100	0.160	0.020
Crusts	0.020	0.000	8.020	1.460	0.870	48.750	0.660	0.350	0.040

Figure (13): The percentage of the presence of the main elements in samples of schist and samples of crusts formed on them

Sandstone samples

Sandstone consists mainly of silicon compounds associated with links of calcium compounds, this can be observed in the composition of the original samples of sandstone that were taken from the region and analyzed using XRF apparatus represented in figure (9), where the percentage of silicon and calcium compounds was present In the studied samples 27.88% and 37.33% respectively, as aluminum and iron were present in these original samples by 8.335% and 3.165% respectively, also noticed from figure (14) that the chemical composition of sandstone has changed significantly in crusts samples compared to the original samples.



	SO3	Cl	SiO2	Al2O3	Fe2O3	CaO	MgO	Na2O	K2O
Rock	0.015	0.015	27.880	8.335	3.165	37.330	1.390	1.225	1.275
Crusts	0.135	0.085	9.640	0.505	0.420	48.610	1.090	0.060	0.100

Figure (14): Percentage of main elements present in sandstone samples and samples of crusts formed on them.

The presence of silicon compounds decreased significantly to 9.64%, and the presence of aluminum and iron compounds decreased by 0.505% and 0.42% respectively, this decrease in the presence rate also included sodium, potassium and magnesium compounds, While there was a clear increase in the presence of calcium compounds, which amounted to 48.61%, as well as a noticeable increase in the presence of sulfur and chloride compounds, these apparent changes in the chemical composition between sandstone samples and formed crust samples (crumbs) indicate the occurrence of chemical weathering.

Conclusions

The study shows that an annual rate of corrosion formation in marble can reach 58.53 micrometers. This figure can be significant on the long term, this means that we may lose this important human heritage, accordingly, we must work from now to protect this ancient city from damage. Based on this rate, the total corrosion over 30 years (since the commissioning of the power station) can reach 1.75 mm leaving a permeant damage on the features of the city. This means protecting the city cannot be postponed.

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