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Full Length Research Paper

Salt weathering, bio-deterioration and rate of weathering of dimensional sandstone in ancient buildings of Aachen City, Germany

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Over the last decade, the concept of stone decay being caused by one or two processes has been revised in favour of a more holistic notion that many operate together, cyclical or seasonal. Certain of these processes may become pre-dominant over others at a particular environment that enhances the impact of these process or processes. The rock's mineralogy is an effective factor in controlling rock's susceptibility to weathering and deformation of its original form. Salt weathering has been reported to act at almost all environments even at low intensity, many researchers from different disciplines agree that biological deterioration is of significance. The rate of weathering varies and controlled by different factors, it is increased at areas with multiple cycles of weathering that repeat on short time with high intensity. In the current study we aim to examine salt weathering, Bio-deterioration on the sandstone blocks constituting ancient buildings "walls" at Aachen City, as well as compute the rate of weathering at such humid region on such sandstone. The aims have been achieved from sampling from weathered parts, measuring depth of greatest back weathering forms, laboratory analyses for collected rock samples. The results indicated high impact of salt weathering that result from chemical interaction of air pollutants with mortars' carbonate content, the growth of lichens affects on such sandstone as noted in SEM presentation. The rate of weathering is noticeably high.

Key words: Weathering, rock mineralogy, rate of weathering, environmental conditions.

INTRODUCTION

Minerals' transformation is a natural event in the continuous evolution of the biosphere on the earth. One of the consequences of such processes is gradual deterioration of stone (Eckhardt, 1985; Griffin et al., 1991; Sand et al., 1989). Deterioration of stone; including building stone and man-made construction materials, exposed to the natural environment occurs through physical, chemical, biological processes in combination or in isolation. It has been estimated that about 30% of stone deterioration is a result of biological activity (Wilimzig, 1996).

The evidence of bio-deterioration by micro-organisms ranges from the microscopic, of cracked or etched materials, to macroscopic scales of flaking, spaling and powdering of the stone surface.

Salt weathering is another wide-spread damaging factor for almost all rocks at all types of environments (Hutchinson et al., 1993; Takahashi et al, 1994; Kamh, 1994, 2000 and McBride and Picard, 2004). The mechanism of salt weathering is highly sensitive to the dominant environmental conditions (air temperature and its relative humidity) on both macro and/or microscale. The exact mechanism of salt crystal growth and progress of rock's texture deterioration is not well known till now. It may be partially noted if environmental scanning electron microscope (ESEM) is used for a given rock sample containing single or combination of salts examined at certain limits of environmental conditions, but even though, the process is still almost unclear as we can never simulate the whole natural process within the ESEM.

The rock's deterioration is not mainly based on the conditions surrounding the rock, but also and mainly on the rock's texture and composition (mineralogy). The rock may have two types of minerals known as stable minerals and labile (unstable) minerals. The later can easily and at certain conditions be altered to other minerals that are more stable and resistant to weathering processes. Clay minerals increase rock's susceptibility to weathering processes "particularly smectite if exposed to moisture from any source" (Winkler, 1966; Dobereiner and DeFreitas, 1986; Gonzalez and George, 2004).

The current study throws light on how and why two weathering processes can act on the same rock, and what is the mechanism of each process at such humid region on such sandstone. Also, as the construction age of such walls is well recorded, then, it enables us to compute the rate of weathering to raise an alarm on the necessity of preservation of such valuable structure at Aachen City.

Study area

The study area “Aachen City" is a humid city with high relative humidity in most of the year reaching up to 96%, rainfall of about two-hundred and thirty days with an average rainfall 800 mm/year, average maximum air temperature at hottest months "July and August" is 21°C and average minimum air temperature at coldest months "January and February" is 3°C (Local Meteorological Center at Aachen City, and personal communication with Dr. Kurt Heinrichs at RWTH in 2006 and 2007). The ancient buildings under investigation (presenting distinctive weathering forms) have been built from sandstone blocks of fine sand and dated back to fifty years before present. Aachen City has also two ancient walls that have been reconstructed many times at several parts of it, it is namely: Inner wall (First wall "Barbarossamauer") with length 2.5 km, 8 to 10 m in height, and about 1.7 m in thickness. It has ten gates and ten towers. In front of this wall was a trench up to 25 m wide partially filled with water, but in the 17th Century the wall was used as quarry for buildings. The outer wall "Second wall" with length 5.5 km, has eleven gates and twenty two towers. It was destroyed by order of the French Emperor "Napoleon" at the very beginning of the 19th Century.

This study aims to define the role or charity of rock's mineralogy, salt weathering and micro-organisms in deforming rock's morphology and creating the present day weathering forms at the study area. Also, it aims to compute rate of weathering of this sandstone at such humid cold region.

METHODOLOGY

To achieve the aims of the current study, field and laboratory investigations have been conducted. The field study includes sampling from the parts presenting weathering forms, measuring dimensions of each weathering form (for example, depth from stone surface, width ……etc.) to quantitatively define its damage category using Kamh (2009) pre-model. This pre-model presents six damage categories starting from low damage category up to very severe damage category. Table 1 presents the weathering forms (matching those noted at the study area) and its dimensions through the six damage categories listed by Kamh (2009). This is to enable the definition of the damage category of the ancient buildings at Aachen City using the field measurements and dimensions of the weathering forms on this construction sandstone.

The rate of weathering of this dimensional rock at such environment can be computed following Matsukura and Matsuoka (1996) equation that is presented as follows:

Rate of weathering "R_w" = D_w / T_e

Where D_w is depth "mm" of largest ten back-weathered parts of the examined surface; and T_e is time in years of rock's exposure to sub-aerial conditions.

Laboratory investigations include the following:

1- Petrographic study using transmitting polarizing microscope to investigate rock's texture and composition; scanning electron microscopic study to investigate rock's weathering and bio-deterioration on grain scale; X-ray diffraction has been used on rock powder to find out rock's mineralogy that clarify rock's susceptibility to weathering.
2- Hydrochemical analysis had been conducted for the extracted solutions prepared from the collected rock samples following the method of Rhoades (1982). This is to find out type and percentage of rock's salt content and its total dissolved salts. Also, to clarify the role and mechanism of salt weathering at the study area. Titration and flame-photometer have been used for this analysis.
3- Rock drilling resistance has been conducted to find out the weathering profile from stone surface to deep inside at the weathered parts of these buildings. The drilling resistance has been conducted using the drilling resistance system that works at constant drilling rate for 60 s of working time at each test point. This system provides the data on time-depth profile chart.

RESULTS

The following is list of field investigation and laboratory analyses for the rock samples collected at the weathered parts of the walls under investigation.

Field study

Full documentation and recording of weathering forms and its dimensions have been conducted and listed in Table 2. These forms have been photo-documented as shown in Figures 1 to 6. Based on these weathering forms, its dimensions and Kamh (2009) pre-model compo-scale (Table 1), it can be indicated that these buildings with such weathering forms and these dimensions have damage categories 4 and 5 that is, severe to very severe damage category with main tendency to severe damage category.

Rate of weathering

It has been computed from the field measurements of maximum ten back weathering and using Matsukura and Matsuoka (1996) equation:

Table 1. Listing of weathering forms and its dimensions in the six damage categories from Kamh (2009) pre-model, considering the forms at the study area.

Damage category	Weathering form	Item of measuring	Item range
Low damage category (L. D. C.)	Salt efflorescence	Salt crust thickness (mm)	<0.1 - 2.0
	Exfoliation or scaling	Sheet thickness (mm)	1.0 - 3.0
		Wall side area (%)	< 5 – 20
	Black crust	Thickness (mm)	0.1 - 1.0
		Wall side area (%)	< 5.0 – 25
	Biological cover	Wall side area (%)	1.0 - 7.0
Low to moderate damage category (L. ~ M. D.C.)	Salt efflorescence	Salt crust thickness (mm)	< 0.1 - 0.5
	Exfoliation or scaling	Sheet thickness (mm)	1.0 - 1.5
		Wall side area (%)	40 - >40
	Black crust	Thickness (mm)	0.1 - 0.5
		Wall side area (%)	50 - > 50
Moderate damage category (M.D.C.)	Salt efflorescence	Salt crust thickness (mm)	2.0 - 5.0 / 0.5 - 2.0 / < 0.1 - 0.5 <10 – 15 / 15 – 20 / 20 - 65
	Exfoliation or scaling	Sheet thickness (mm)	3.0 - 6.0/ 3.0 - 1.5/ 1.5 - 1.0
		Wall side area (%)	< 5 - 10/ 10 – 20 / 20 - 40
	Black crust	Thickness (mm)	1.0 - 3.0/ 0.5 - 1.0 / 0.5 - 0.1
		Wall side area (%)	< 5 - 10/ 10 - 25/ 25 - 50
	Biological cover	Wall side area (%)	7.0 - 20.0
Moderate to severe damage category (M. ~ S. D.C.)	Salt efflorescence	Salt crust thickness (mm)	5.0 - > 5.0 / < 0.1 - 0.5 < 10 - 15/ 65 - 90
	Exfoliation or scaling	Sheet thickness (mm)	6.0 - > 6.0/ 1.0 - 1.5
		Wall side area (%)	< 5 – 10 / 40 - > 40
	Black crust	Thickness (mm)	3.0 - >3.0 / 0.1 - 0.5
		Wall side area (%)	<5.0 - 10 / 50 - > 50
Severe damage category (S.D.C.)	Salt efflorescence	Salt crust thickness (mm)	2.0 - 5.0/ 0.5 - 2.0
	Exfoliation or scaling	Sheet thickness (mm)	3.0 - 6.0 / 1.5 - 3.0
		Wall side area (%)	10 - 20/ 20 - 40
	Black crust	Thickness (mm)	1.0 - 3.0 / 0.5 - 1.0
		Wall side area (%)	10 - 25/ 25 - 50
	Biological cover	Wall side area (%)	20 - 45
Very severe damage category (V.S. D.C.)	Salt efflorescence	Salt crust thickness (mm)	5.0 - > 5.0/ 2.0 - 5.0 / 0.5 - 2.0 10 - 90/ 20 - 90/ 65 - 90
	Exfoliation or scaling	Sheet thickness (mm)	6.0 - > 6.0/ 3.0 - 6.0 / 1.5 - 3.0
		Wall side area (%)	10 - > 40/ 20 - > 40 / 40 - > 40
	Black crust	Thickness (mm)	3.0 - > 3.0/ 1.0 - 3.0/ 0.5 - 1.0
		Wall side area (%)	10- >50/ 25 - >50/ 50 - > 50
	Biological cover	Wall side area (%)	45 - >45

"R_w" = D_w / T_e , then,

"R_w" = 21 / 50 = 0.42 mm/year

This rate is considered as high rate of weathering on such sandstone if compared with similar studies conducted at such climatic conditions that had been reported to be 0.1 mm/year for the unsheltered sandstone cliff side (Allan, 1991; Halsey et al., 1996).

Table 2. Listing of weathering forms and its dimensions as noted at the study area to quantitatively define rock's damage category based on Kamh (2009) pre-model.

	Item of measuring	Item range	Damage category based on Kamh (2009) pre-model
Salt efflorescence	Salt crust thickness (mm)	<0.1 - 2.0	Severe
Exfoliation or scalling	Sheet thickness (mm)	1.0- 0.3	Severe to very severe
	Wall side area (%)	<5 - 20	Severe to very severe
Black crust	Thickness (mm)	0.1- 1.0	Severe to very severe
	Wall side area (%)	<5.0 - 25	Severe to very severe
Biological cover	Wall side area (%)	1.0 - 7.0	Severe

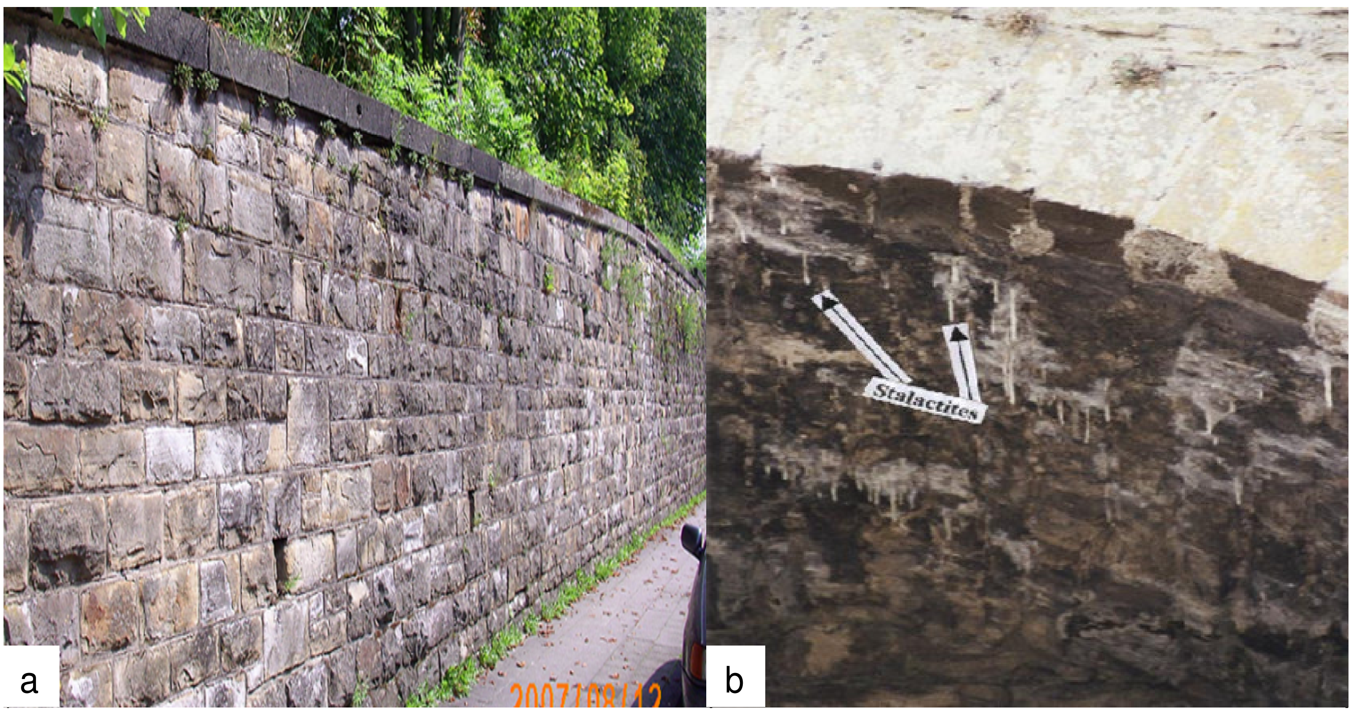

Figure 1. (a) General view for the ancient wall with variable weathering forms and plant vegetation behind and across building stones. (b) Close view on one of gates ceiling presenting stalactites growing down ward originating from the ancient mortar of this ceiling.

Laboratory investigations

Petrography and weathering susceptibility

This investigation throws light on rock's texture and mineralogy that clearly prove rock's susceptibility to weathering. Also, it indicates rock's weathering features on micro-scale or even down to nano-scale. This investigation has been conducted using scanning electron microscope supported with electron dispersive X-ray (EDX), and X-ray diffractometer at Engineering Geology and Hydrogeology Department and Crystallography Department Aachen University (RWTH) Germany.

Scanning electron microscopic investigation aims to examine on micro or nano scale the rock's cement type, weathering on micro-scale and any micro-biological features. It indicated that the cement is mainly kaolinite, K-feldspar is severely fragmented and partially altered to kaolinite, elongated micro-pitting and etching can be noted on quartz grains, microbial community comprised of fungal hyphae, algae and bacterial cells penetrating through rock's texture and components of this weathered sandstone (Figures 7 to 10). Two points have been checked by EDX for two sandstone samples, one at

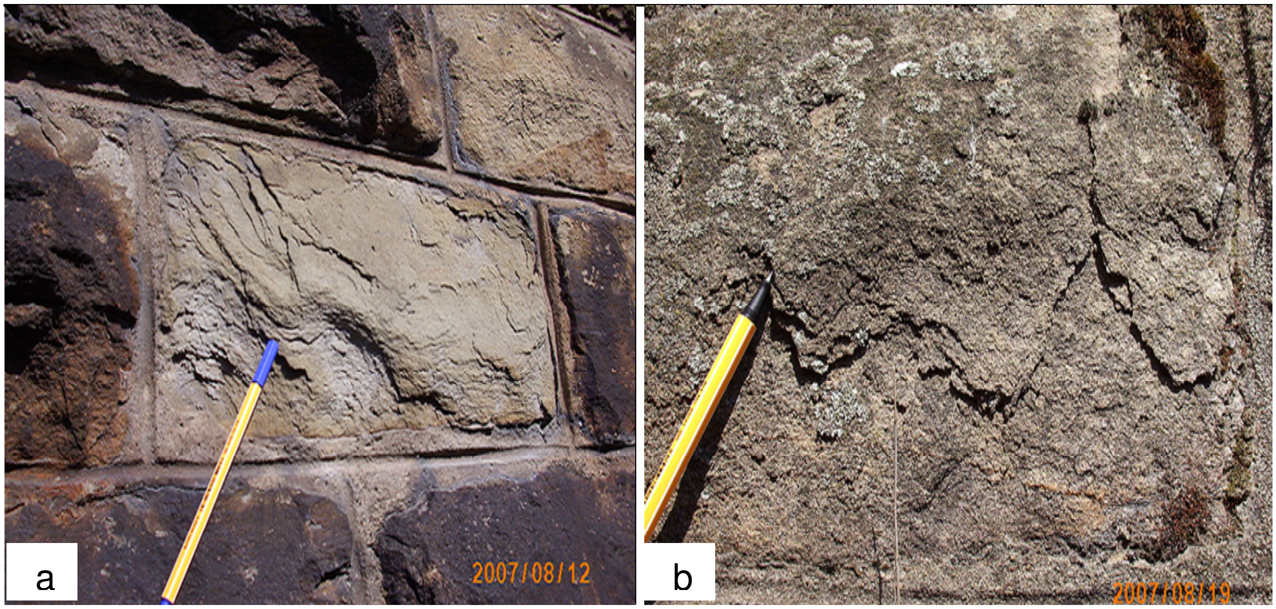

Figure 2. (a) Close view on thin multiple scaling and blackening of stone surface for building stone of the ancient walls in Aachen City. (b) Close view on thick multiple scaling and microbial accumulation on stone's surface for building stone of the ancient walls in Aachen City.

Figure 3. Back weathering and multiple scaling, white salt efflorescence and blackening of stone surface of construction rock of the walls under investigation.

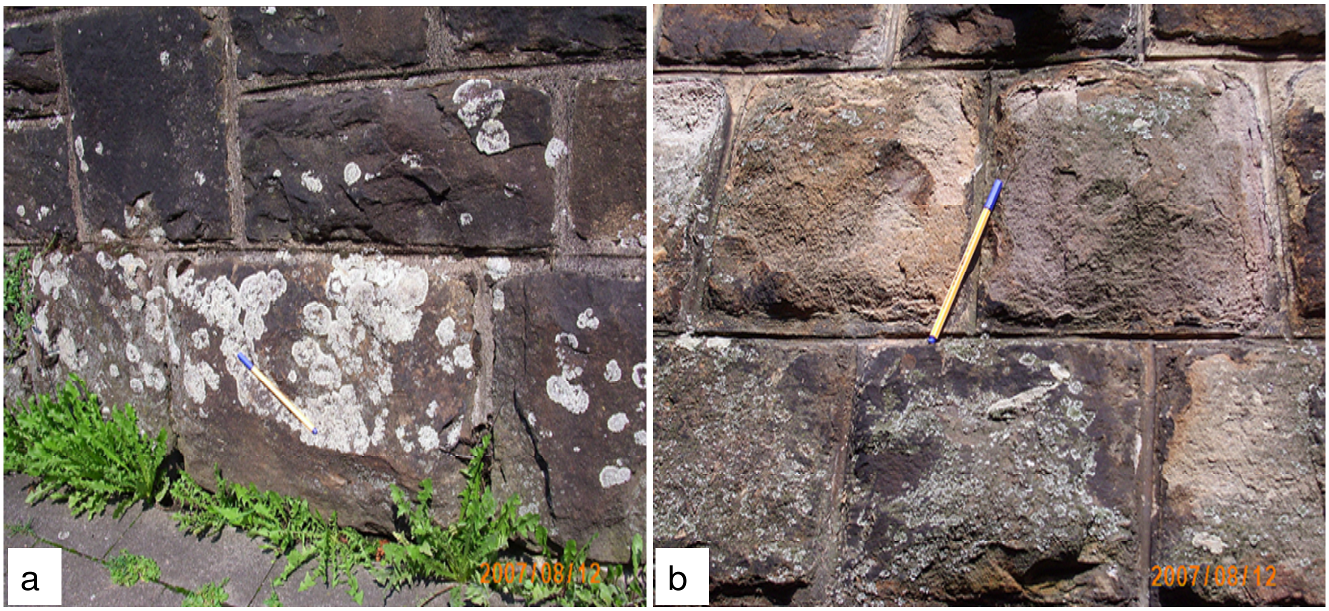

Figure 4. (a) Intense accumulation of Lichens and white salt efflorescence, blackening and detachment of stone surface material at different parts of stone surface. (b) Intense accumulation of Lichens patches, blackening, and detachment of stone surface material and growth of natural mega plants at basal parts of this wall.

Figure 5. Stone surface damage by single scales and fragmentation into sand-grain size, intense linear salt efflorescence at stone's margins, blackening stone surface.

Figure 6. Structuring stone's surface into domal form, single scales, intense Lichens growth, microbial growth as fibrous soft tissue between stone's margins and the surrounding re-pointing mortar.

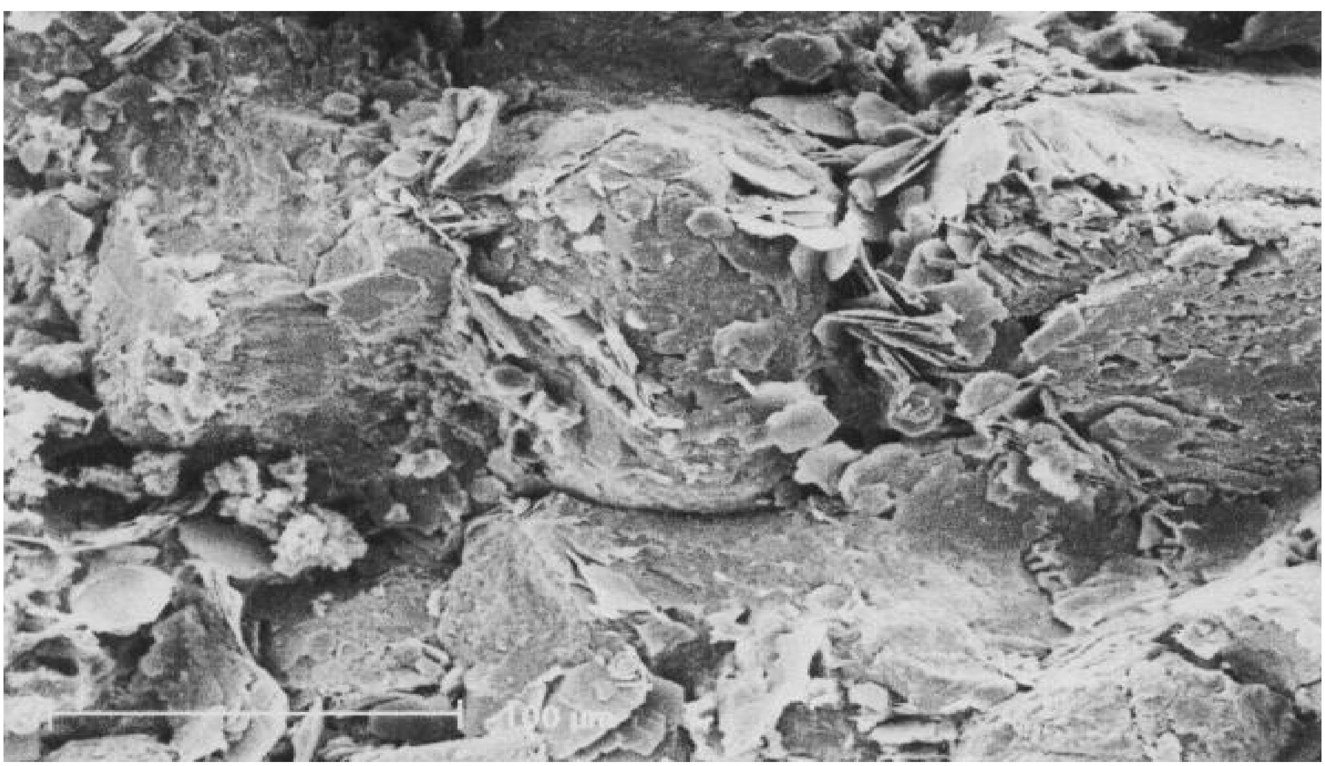

Figure 7. Scanning electron photomicrograph presenting micro-pitting and elongated etches on quartz grains, micro-exfoliation of K-feldspar and alteration to clay minerals for weathered sandstone at the study area, Aachen City.

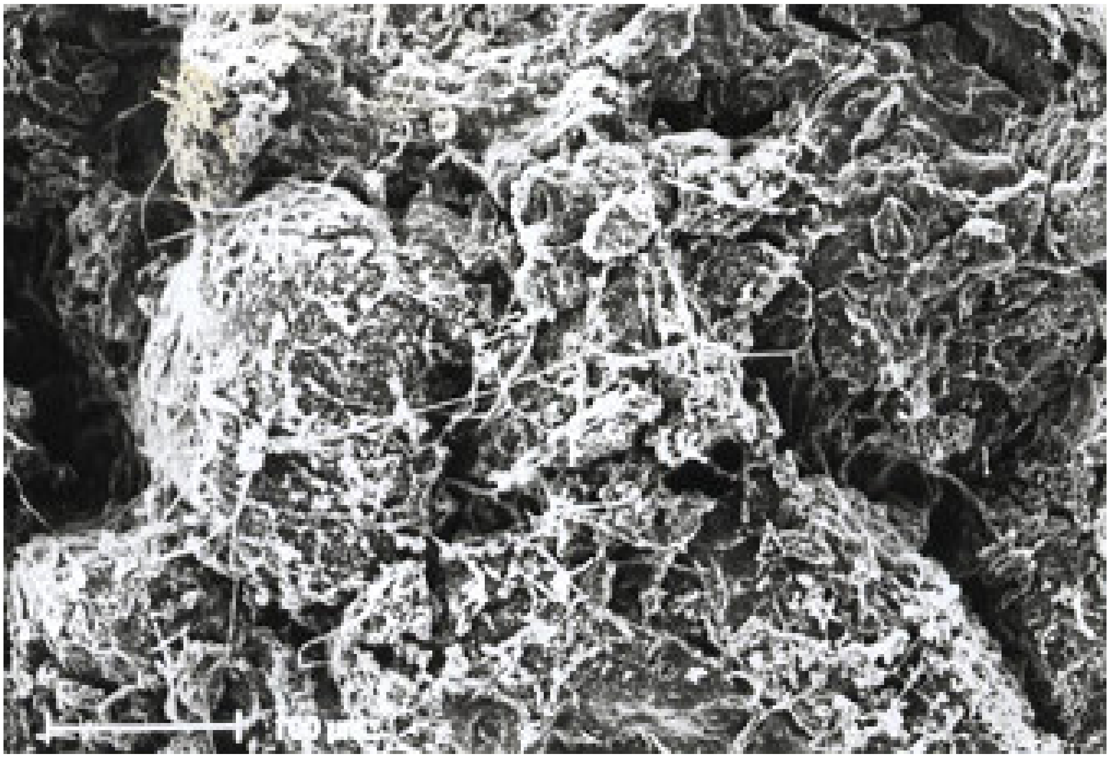

Figure 8. Scanning electron micrograph of presenting some filaments of microbial "algae" within sandstone texture at the study area.

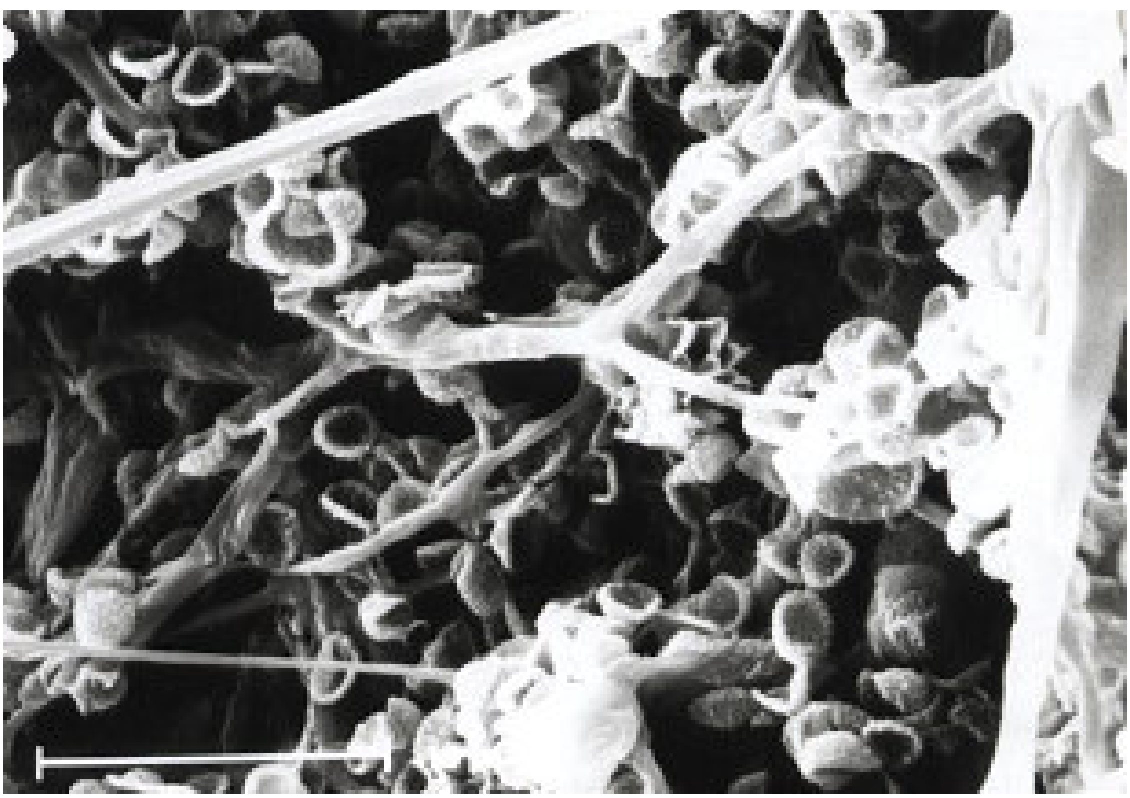

Figure 9. Scanning electron micrograph of microbial community comprised of fungal hyphae and algal and bacterial cells within sandstone rock texture at the study area.

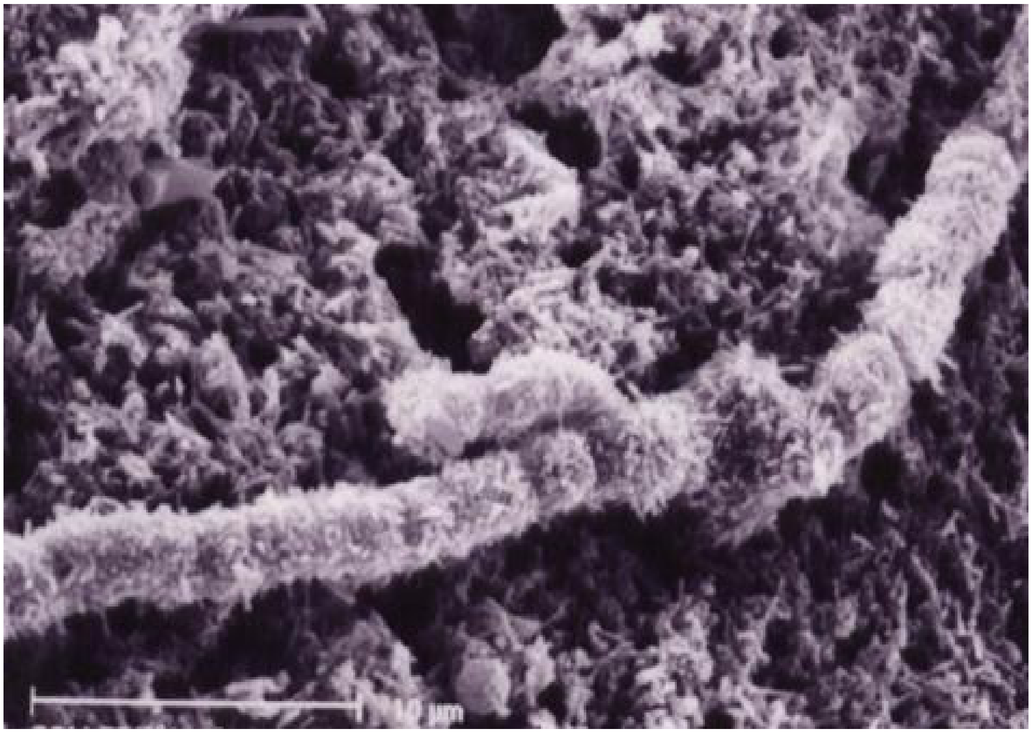

Figure 10. Scanning electron micrograph showing thick filament and tiny microbial accumulation coating sandstone components at the study area.

stone's surface with black crust (Figure 11) indicating very high carbon compared with silica content and other rock's elemental content at the stone's surface; and the other EDX had been conducted for subsurface test point (about 0.9 mm depth from stone surface, Figure 12) indicating considerably high silicon, then aluminum and high reduction of carbon content. This indicates relative variation in air pollutants and black crust from stone surface to deep inside.

The detection of rock's mineralogical composition as well as salts at stone's surface resulting in such weathering forms have been conducted through X-ray diffraction (Figure 13), while X-ray conducted for the deep seated mortar indicated quartz, calcite and gypsum as main components (Figure 14).

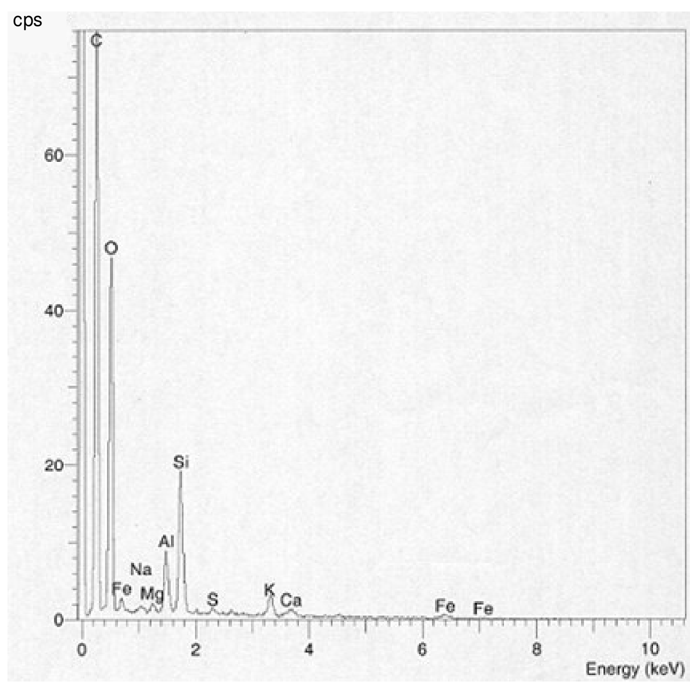

Figure 11. Electron dispersive X-ray for check point at sandstone surface with black crust presenting high cps for carbon compared with silicon and other elemental components of this sandstone, ancient buildings, Aachen City, Germany.

Hydrochemical analysis

It has been conducted for the extracted solutions prepared from the rock samples collected at the weathered parts of these buildings following the method of Rhoades (1982). The results involving samples' height and its orientation to solar heating in addition to the electrical conductivity, total dissolved salts and the hypothetical dissolved salts in these samples, the results are listed in Table 3. It can be noted that the weathered sandstone at parts with intense weathering forms has high total dissolved salts up to 96250 ppm with chlorides and sulphates as dominant salts; while, the weathered sandstone at parts with microbial communities has also

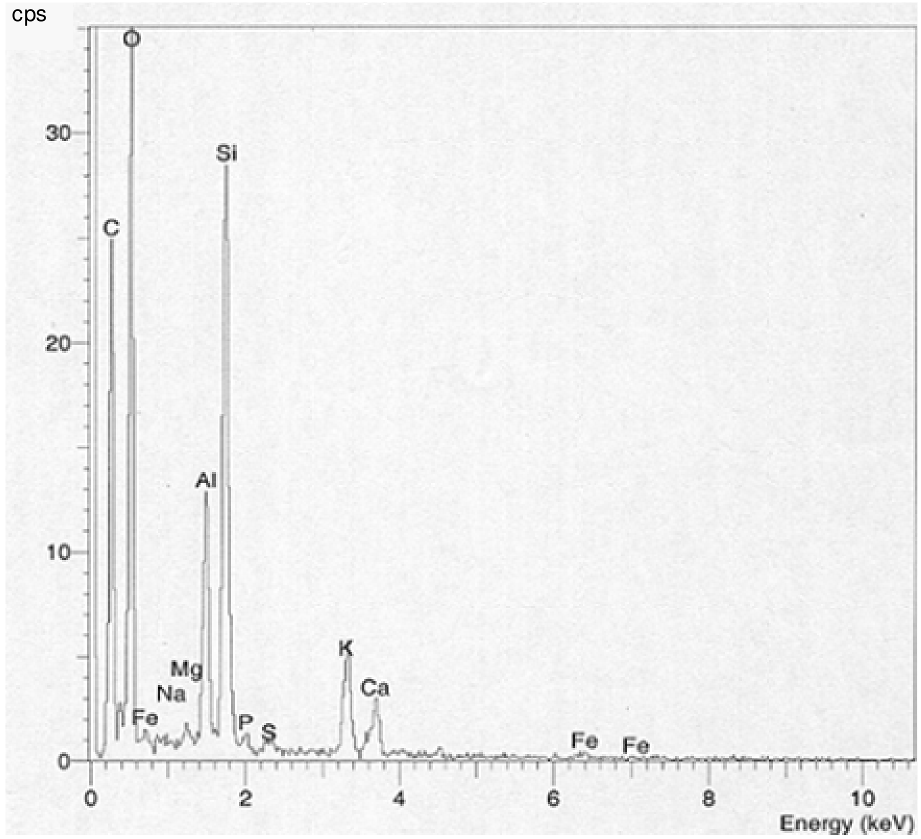

Figure 12. Electron dispersive X-ray for check point 0.9 mm depth from stone surface at the weathered parts presenting relatively higher cps for silicon compared with carbon and other elemental components of this sandstone, ancient buildings, Aachen City, Germany.

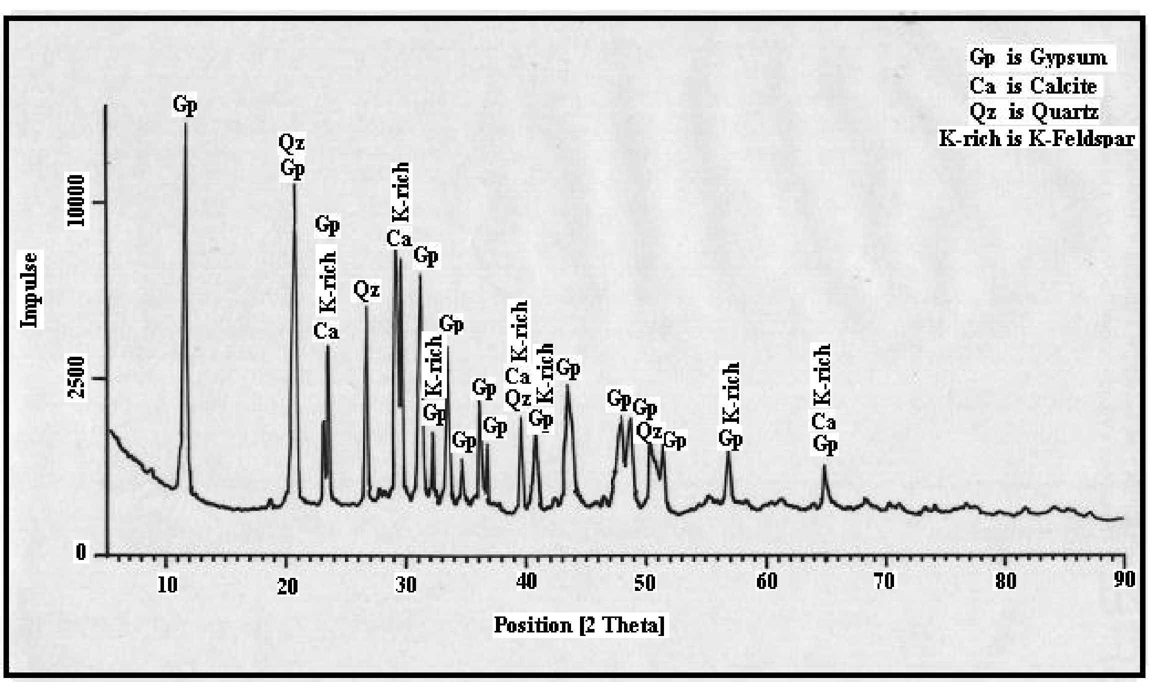

Figure 13. X-ray diffractograph presenting mineralogical composition for sandstone coated with black crust collected from the weathered sandstone constituting the ancient buildings at the study area.

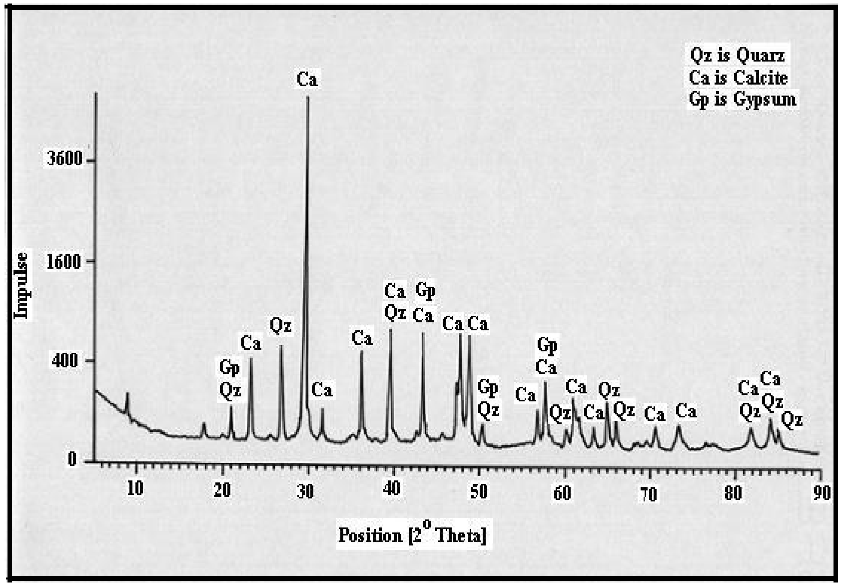

Figure 14. X-ray diffractograph presenting mineralogical composition for mortar sample collected from the weathered ancient buildings at the study area.

Table 3. Hydrochemical analysis for weathered and deep seated sandstone and weathered mortar for ancient weathered buildings at Aachen City, Germany.

Sample number	Sample type and state	Wall side orientation	Sample height above ground surface (m)	E.C. mmhos/ cm	T.D.S. (ppm)	Cations (ppm)			
						Ca++	Mg++	Na+	K+
1.	Weathered sandstone at exfoliated parts (margins)	South to South-West facing unshltered wall side	2.5	17.4	96250	11153	2000	43910	1230
2.	Weathered sandstone at exfoliated parts (near surface center)		1.1	15.5	91114	10096	2765	36026	2016
3.	Weathered sandstone at microbial parts		0.9	11.2	83027	9206	1556	19400	1164
4.	Weathered mortar		2.4	19.3	105380	13222	2011	31201	2988
5.	Weathered mortar		1.0	18.7	99750	12709	1866	29270	1766
6.	Deep seated sandstone		2.3	4.1	5720	521	136	2117	682
7.	Deep seated sandstone		1.2	3.7	4244	411	209	2808	320

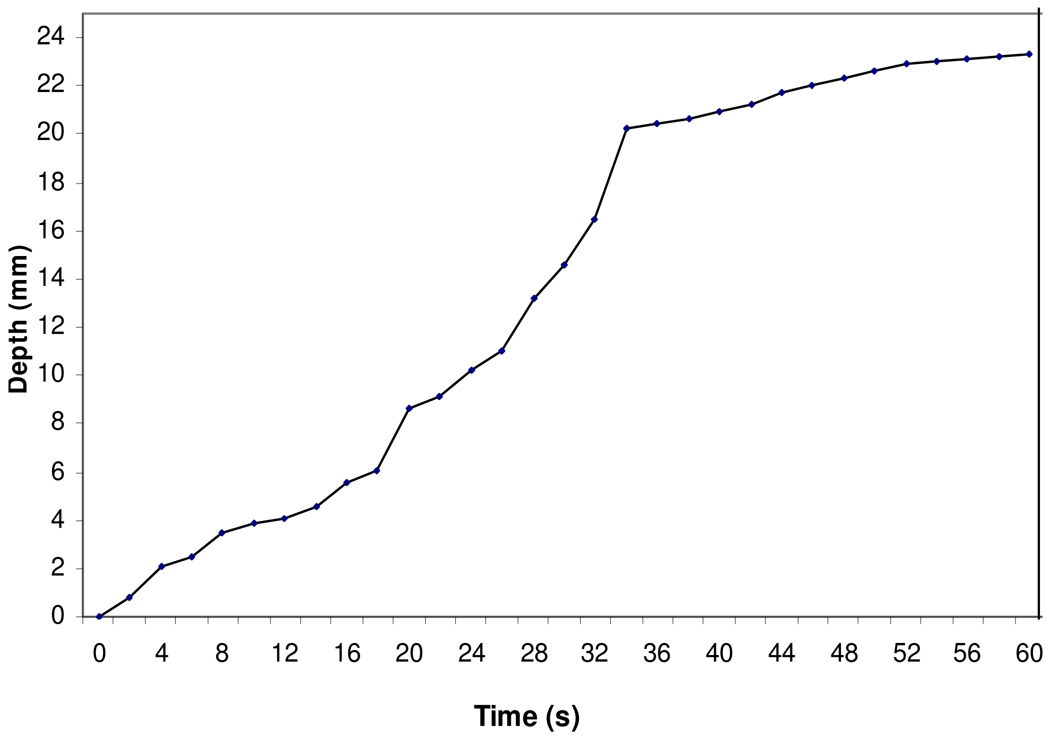

Figure 15. Drilling resistance profile at highly exfoliated parts of the weathered sandstone walls.

high total dissolved salts but with sulphate salts dominating than chlorides. The weathered mortar in these walls has high total dissolved salts reaching up to 105380 ppm with sulphate salts dominating over chlorides (Table 3). All these limits of total dissolved salts are compared with that of the deep seated "nearly fresh" samples collected from the construction sandstone of these buildings (Table 3).

Geotechnical properties' limits

It includes examining rock's hardness examined by drilling resistance system that expresses the rock's resistance to horizontal penetration by drilling system (that is, from stone's surface to deep inside) at these weathering forms as well as at areas nearly free from weathering and the deep seated mortar "original mortar".
The drilling resistance's results are graphically presented in a binary relation (depth of penetration against total drilling time) for the highly weathered exfoliated parts of the construction sandstone of these walls, central less weathered parts of these sandstone blocks as well as the original hydraulic lime mortar of these walls (Figures 15 to 17 respectively). It is clear that the rock at the weathered parts has low resistance to drilling where high penetration rate is noted for the surfacial 2.1 cm thick of stone's surface, then, this rate is highly reduced and becomes steady (Figure 15); similarly, the original hydraulic lime mortar cementing the sandstone blocks of these walls (Figure 17); while the less weathered parts of this rock has higher resistance to drilling that is, has better limits of mechanical properties (Figure 16).

DISCUSSION

Not only the climatic conditions at a given area controls type, mechanism, rate and intensity of physical, chemical and/or biological weathering; but also the rock's physical (mineralogy and texture) and mechanical (resistance to weathering) properties' limits controls its vulnerability to such weathering processes. Consequently, the climatic conditions (air temperature and its relative humidity) and wall side orientation to solar heating; rock's texture, mineralogy, geotechnical limits and microbial content have been considered to discuss the main reasons for creating the weathering forms on nano-scale as well as macro-scale for such ancient building at Aachen City. The rate of weathering has also been computed using Matsukura and Matsuoka (1996) equation especially that such building has a well defined building age and exposure to sub-aerial conditions.

The high limit of air relative humidity and acid rainfall (a result of high pollution limit noted as blackening of sheltered and semi-sheltered stone surfaces) at Aachen City result in flourish of micro-organisms for example, Lichens' patches at stone surface free from salts and pollution, green algae at wet surfaces as noted in Figures (4b and 6), fungal hyphae and bacterial cells within rock

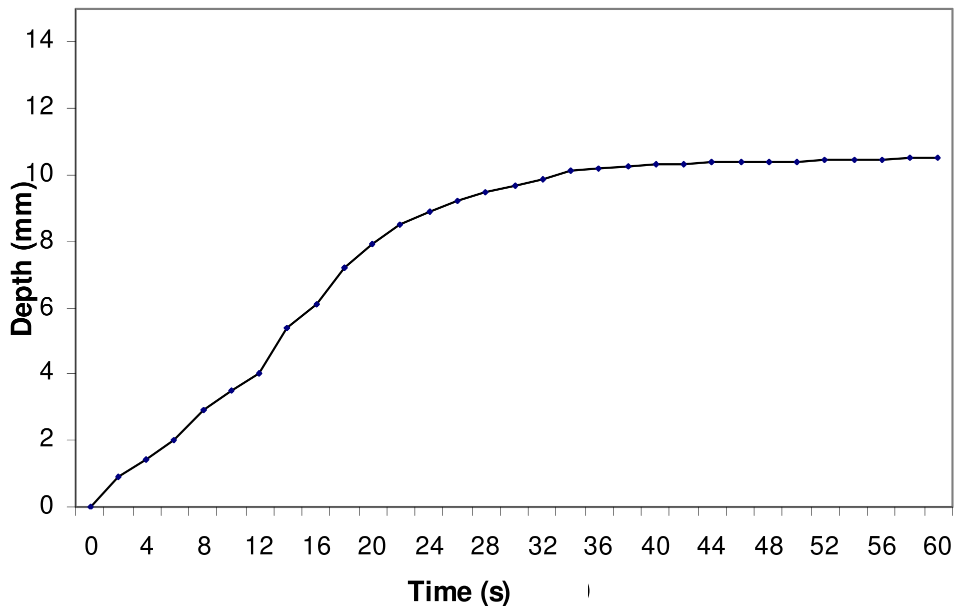

Figure 16. Drilling resistance profile at highly exfoliated parts of weathered sandstone walls.

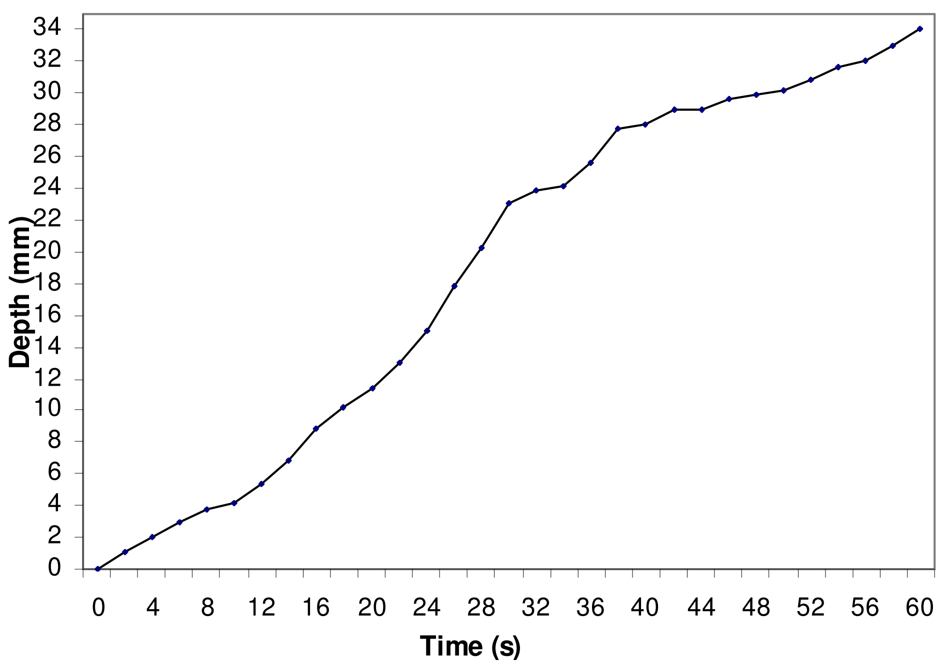

Figure 17. Drilling resistance profile for the original mortar of the walls under investigation.

pores as noted in scanning electron photomicrograph (Figures 8 to 10). The acid rain has also resulted in creation of stalactites growing downward from the gates' ceiling among the construction rocks that is, from the hydraulic lime mortar cementing these dimensional stones (Figure 1b). In the meantime, sulphate salts resulted from chemical interaction of sulphur oxides (either coming from microbial excretion "acid laden" and/or air pollution to stone and/or mortar's surface by wet deposition at such humid or rainy weather, that is altered to sulphuric acid at such humid climate) with rock or mortars' carbonate content producing sulphate salts that are non-hygroscopic salts that is, less mobile salts that stand at certain depth from stone's surface of such construction rock. Then, these sulphate salts, detected hydrochemically at the parts with microbial flourish as well as those at the hydraulic lime mortar (Table 3), started their intensive damaging impact of such sandstone creating the forms noted in the study area. These sulphates experience hydration-dehydration cycles particularly at such suitable environmental conditions (low air temperature down to zero and very high relative humidity up to 95%) (Sperling and Cooke, 1985). These salts on its hydration exerts high pressure on rock's pores or even on nano-scale (crystal lattice of K-feldspar constituting this sandstone) deforming rock cohesion force and/or result in mineral alteration of rock's labile minerals as K-feldspar to clays. The end product of such sheet accumulation of salts at limited zones beneath stone's surface is creation of such weathering forms (for example, scaling, exfoliation, rock meal and micro-cracking).

The variation in rock's strength from stone's surface to deep inside due to weathering and deformation of rock on both nano-scale (Figure 7) and mega-scale (Figures 1 to 6) is graphically and numerically presented in the non-linear direct proportional relation at the weathered parts (Figure 15), and the semi-linear relation at the massive less weathered parts of this construction rock (Figure 16) of the drilling resistance curves for the testing points examined on these walls. The very high rate of drilling at the lime mortar of these walls (Figure 17) indicated that it is a good and an easy stuff for weathering (especially chemical weathering) by acid rain resulting in high limit of total dissolved salts (Table 3), with dominance of sulphates that easily attack the rest of this mortar creating the protruded stalactites (Figure 1b) and probably migrates to the margins of the surrounding construction rock (Figure 5) resulting in creation of the reported weathering forms (Figures 2a, 3 and 4a).

The microbial noted either on stone's surface (Figures 1, 4b and 6) or within rock's texture (Figures 8 to 10) contributes to stone deterioration by both chemical and/or mechanical mechanisms. The mechanical action of hyphae causing stresses, movement and cracking of the stone's matrix (Del Monte and Sabbioni, 1986). The forces exerted by hyphal growth can lead to considerable disruption of the stone matrix and also result in lifting and flaking of stone's surface (Figures 2b, 6 and 10). Mechanical and biochemical deterioration can act together to bring about mineralogical alteration within the stone matrix (Koestler et al., 1985). The deterioration and powdering of dimensional stones (as noted for the buildings under investigation) can be a result of physical decay by fungal hyphae (Caneva et al., 1993; Danin and Caneva, 1990; Krumbein and Dyer, 1985). It can also share in creation of such weathering forms (scaling and exfoliation) noted at the study area (Figures 2a, 2b, 3, 5 and 6).

Lichens are composite organisms, comprising fungal component and alga. It certainly plays a considerable part in rock weathering either physically or chemically. The wet and dry cycling lead to expansion- contraction of the Lichens thalli resulting in detachment of the surfacial micro-thickness sheet adhering to this lichens (Figures 2b and 6).

The noticeably high rate of rock weathering computed for the construction sandstone of the buildings under investigation that reaches up to 4 times as the previous studies revealed for the same rock type at similar climatic conditions (Allan, 1991; Halsey et al., 1996). This may be a result of combination of many weathering processes that is, salt weathering and biological weathering in addition to rock's clay content that reduces the mechanical limits of a given rock.

CONCLUSION AND RECOMMENDATIONS

Salt weathering as well as biological weathering can act in combination or solitary on a given rock resulting in rock's deformation on nano or mega scale. Testing and quantification of rock's damage category can be conducted through detailed field measurements of weathering forms' dimensions and using Kamh (2009) pre-model. The detection of weathering forms on nano-scale as well as main factors of rock's weathering can be conducted on small rock samples through petrographic and geotechnical investigations as well as hydrochemical analysis for the rock's extracted solutions. In the current study rock's weathering on nano-scale has been detected as well as on mega-scale, the construction rock and its surrounding mortar have considerably high limit of total dissolved salts indicating the role of salt weathering on such buildings. The microbial role is an important role in rock weathering. The rate of weathering at the study area is considerably high as many parameters of weathering are acting in combination.

Continuous restoration of such ancient buildings with sandstone blocks of high durability "with very low carbonate and clay content", also the mortar must be with very low or no carbonate content to control creation of salts from chemical reaction with air pollutants if the later cannot be limited.

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