

DEGRADATION OF GNEISS AND GRANITE IN HISTORIC BUILDING FACADES IN CENTRAL RIO DE JANEIRO

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ABSTRACT

The majority of the cultural heritage built in Rio de Janeiro city, Brazil, is made of gneiss or granite, and exposition of the façades to the polluted environment leads to strong degradation. In order to understand these weathering processes, five historic buildings in the city were studied. The selected historic buildings studied exhibited iron staining, granular disintegration, blistering, incipient fractures and contour scaling, and black crust development. Samples collected from these buildings were examined in an attempt to understand the mechanisms of surface weathering. Rock samples were collected from areas that exhibited serious symptoms of stone decay. The anion and cation content of the building materials was evaluated by AA spectrometry and Ion Chromatographic analysis. The samples were also studied by Field-Emission SEM, petrographic analysis and for their biological content by SEM and DNA analysis using Illumina Mi-Seq Next Generation Sequencing. All the chemical analyses showed high concentrations of soluble salts, such as halite and gypsum, which play a very important role in stonework weathering. FE-SEM with EDS analysis allowed the detection within the rock of sparse filamentous fungi, groups of bacterial cells, rare diatoms and, especially interesting, filamentous photosynthetic bacteria encrusted with re-precipitated gypsum, showing the participation of microorganisms in stone degradation.

Keywords: weathering, black crust, urban pollution, rock decay.

1. INTRODUCTION

A substantial proportion of the world's tangible heritage is constructed from rocks (Scheerer et al., 2009). Rocks are materials whose characteristics are stable in the conditions of high pressure or temperature under which they were formed within the Earth's crust. When they become exposed at the surface, however, they encounter different temperature and pressure regimes, and are newly exposed to a range of substances such as water, atmospheric gases, and living organisms. Under these conditions they become unstable and undergo consequential changes in material properties and mineralogical composition (Mottershead et al., 2003). Many buildings of cultural interest are located in urban environments, where pollution caused by road traffic and industry has harmful consequences for outdoor and indoor constructional and decorative materials (Fassina, 1991; Sabbioni and Zappia, 1992; Sabbioni et al., 1996; Goudie and Viles, 1997; Van Grieken et al., 2000; Esbert et al., 2001; Moropoulou et al., 2001; Smith et al., 2002; Jordan et al., 2009). On the exterior of buildings, the impact of environmental gases, particles released from combustion of fossil fuels and aerial biological organisms leads to the formation of black crusts and other undesirable aesthetic effects, and may also compromise the integrity of the materials (Esbert et al., 2001; Moropoulou et al., 2001; Bonazza et al., 2005; Sanjurjo Sánchez et al., 2009; Xu et al., 2010). The formation of weathering crusts on natural stones in urban areas is regarded as an important factor in stone decay (Amoroso & Fassina, 1983; Török, 2003; Baptista Neto et al., 2006). During the decay of a stone building, apart from the intrinsic characteristics of the construction material itself (chemical and mineralogical composition, petrophysical properties, etc.), the environmental characteristics of the site of the building are also involved (Iñigo & Vicente-Tavera, 2001).

The aim of this study is to examine the main agents that affect the degradation of some of the important historic buildings in central Rio de Janeiro (Figure 1).

2. ENVIRONMENTAL CONDITIONS

The metropolitan area of Rio de Janeiro (6,500 km²) is the second most populated and one of the most prosperous areas in Brazil. According to the Brazilian Institute of Geography and Statistics – IBGE, in 2014 the metropolitan area housed 12.229,867 residents. This area experiences a humid sub-tropical climate, an average annual temperature of 22°C with summer temperatures ranging from 30-32°C and average rainfall of 1200-1800 mm. Temporal variation in precipitation results in dry (from June to August) and wet (from November to March) periods, with marked differences in the mean rainfall inputs. Although rainfall is concentrated between November and March, there may be rain and

relative humidity during any month, especially in littoral locations. Therefore, rainfall remains high throughout the year, together with high concentrations of marine aerosols. A significant decline in air quality over recent decades has occurred, caused by increased vehicle emissions. It is now common under still-air conditions for much of the area to be blanketed by a photochemical smog (Smith et al., 2004). This smog contains high concentrations of carbonaceous and sulfate rich particles that originate from industrial emissions from construction sites, soil disturbance and stone masonry activities (Daisey et al., 1987; Azevedo et al., 1999). Marine aerosols can also be added to this list, since sodium and chlorine deposition rates of 2.2 and 4.2 t/km²/per annum, respectively, have been estimated (Moreira-Nordemann et al., 1988). Extractable organic matter in urban aerosols has been used as a marker for fossil fuels amongst the aliphatic hydrocarbons monitored in low level sites in Rio de Janeiro (Azevedo et al., 1999). Elsewhere in Rio de Janeiro State, in industrial and urbanized environments, Quiterio et al. (2004) have identified, in the absence of strictly enforced emission controls, heavy metal concentrations and traces in airborne particulates that reach levels significantly in excess of those generally recorded for similar areas around the world. The presence of these metals in any deposited particulates could catalyze the formation of gypsum on stone surfaces (Camuffo et al., 1983). In addition, microorganisms deposited on the surfaces of the buildings can lead to neoformation of gypsum, calcite, or other deposits (Gadd, 2017).

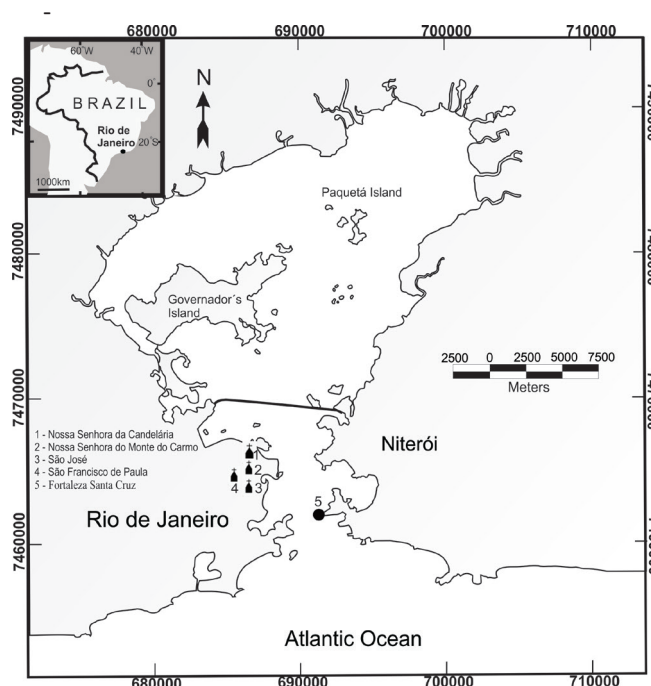


Figure 1. Location map of the studied sites.

According to Smith & Magee (1990), high levels of vehicle emissions are reflected in the physical appearance and

soiling of many buildings within the city center, where high densities of private and commercial traffic are frequently channeled along streets flanked by high-rise buildings. Pollution tends to concentrate and persist within these corridors and its most obvious long-term effect is the black staining of many buildings near street level.

3. MATERIALS AND METHODS

3.1 Sampling and analytical methods

In order to identify the main agents that affect rapid block retreat on the surface of gneiss in historic building façades in central Rio de Janeiro, samples were taken from the front façades of four churches in the center of the city of Rio de Janeiro and from the Santa Cruz Fortress in Niterói, Brazil: Nossa Senhora do Monte do Carmo (CA), situated on Praça XV de Novembro; São Francisco de Paula (SF), in the Largo de São Francisco de Paula; São José (SJ), in Av. Pres. Antônio Carlos; and Nossa Senhora da Candelária (C), located in Praça Pio X. The Santa Cruz Fortress is located in the entrance of Guanabara Bay. Rock samples were collected from areas that exhibited well developed black crusts and showed serious symptoms of stone decay. In the laboratory, the samples were analyzed by various techniques in order to identify the main agents that could influence the weathering processes (McAlister, 1996). Because of the sensitivity of sampling historic buildings, it was only possible to obtain small chips (usually <2X2cm) from surface crusts. These small chips of weathering stone and black crust were mounted separately onto aluminium stubs and coated with gold, and their surface analyzed using a Scanning Electron Microscope (SEM) (Joel Winsen JSM 6400). Other samples were coated with gold-palladium or iridium, examined with a Field Emission (FE)-SEM and characterized by energy dispersive X-ray spectroscopy (EDS). Water-soluble salts were extracted from the <63 µm fractions by shaking 2 mg of sample in 10 ml of deionized water for two hours and allowing them to stand overnight at room temperature prior to centrifugation and filtration through 0.2 µm membrane filters for analyses. The cations Na, K, Mg, Ca, Cr, Mn, Fe, Ni, Cu, Pb and Zn were determined using a Perkin Elmer Model 3100 atomic absorption spectrometer. An air/acetylene flame was employed to atomize the sample solutions. Water soluble ions were also extracted from 0.5mg of the sample in 2.5cm³ of 0.2µm-membrane-filtered deionized water and, after membrane filtration (0.2µm), subjected to Ion Chromatographic analysis. Water-soluble anions, F, Cl, NO₃, PO₄ and SO₄, were analyzed using a Dionex Model DX 500 ion chromatograph. An Ion-Pac AG4A-SC (4mm) guard column and an AS4A-Sc (4m) anion exchange column were used and the injection loop was 25 µL volume. A 1.8m M Na₂CO₃/1.7mM NaHCO₃ mi-

xed solution at a flow rate of 2ml per minute was used as elutant, and the conductivity detector limit for all anions was 1 ppm (Baptista Neto et al., 2006).

3.2 Microbial population analysis

The microbial populations in the samples were determined by the Next Generation Sequencing (NGS). DNA was extracted from samples and subjected to 16S rDNA (bacteria) and ITS (fungi) gene amplification, followed by NGS, as described in Gaylarde et al. (2017a; 2017b) and Ogawa et al. (2017a).

4. RESULTS AND DISCUSSION

Rio de Janeiro contains some of the most important colonial architectural buildings in Brazil. Augen gneiss and granitic rocks were used in the construction of most of the historic buildings of the city, including museums and churches (Smith and Magee, 1990; Smith et al., 2004 and 2007; Mansur et al., 2008; Ricardo et al., 2017). They were also used in the production of sculptures, ornamentations, façades, and door and window frames, because of its weathering resistance. Nowadays, however, these buildings are experiencing a rapid block retreat, showing symptoms of severe stone decay, which has increased in recent decades. The majority of the buildings and monuments in the center of Rio de Janeiro are typically blackened; this is considered to be due to the accumulation of atmospheric pollutants on their surfaces.

The degree of stone weathering of the façades of the churches was observed in the field. Areas most affected by black crusts and weathering were selected; iron staining, granular disintegration, blistering, incipient fractures, and contour scaling of the stone façades were observed in all buildings. Figure 2 shows the main pathologies observed in the buildings.

The analyses carried out by SEM on the collected samples extracted from the black crusts from the building façades showed high concentrations of various salts, such as gypsum and halite (Figure 3). Black and often irregular surface coloring can be observed on many historic stone buildings and statues worldwide, decreasing their aesthetic value. The processes behind the production of these thin black coatings are, however, much more serious than merely the aesthetic aspects. They are connected with weathering through increasing air pollution, microbial activity, and climatic conditions (Saiz-Jimenez, 1991; Fitzner et al., 1995; Galletti et al., 1997). In order to observe the possible occurrence of salts within the rocks, petrographic thin sections were produced (Figure 4) and showed the formation

of gypsum in the rock microfractures, indicating the importance of this salt in the weathering rock. According to Sabbioni (1995), the formation of the gypsum crust is due to the wet and dry deposition of atmospheric sulfur and the subsequent transformation of calcium carbonate into calcium sulfate dihydrate. However, the black crusts develop in rain-sheltered areas of stone buildings, following acid deposition (dry deposition) of atmospheric pollutants (Rodríguez-Navarro and Sebastian, 1996). According to Baptista Neto et al. (2006), the obvious source of calcium in the study area is the mortar in the joints between the granite blocks, which is also used to “protect” the areas already affected by weathering. Calcium can also originate from the fly ash that was found in all the analyzed samples. Fly ash particles are spherical in shape, have irregular porous surfaces, containing high concentrations of carbon, silicon, sulphur, aluminium, and calcium and originate mostly from vehicular emissions (Del Monte et al., 1981). Currently, the majority of the studies on black crust formation have been focused mainly on sedimentary carbonate stones and marbles (Brimblecombe and Grossi, 2007). The formation of black crusts on siliceous rocks, such as granite and gneiss, has not been as extensively studied as in calcareous rocks (Silva et al., 2009, Pozo-Antonio et al., 2016).

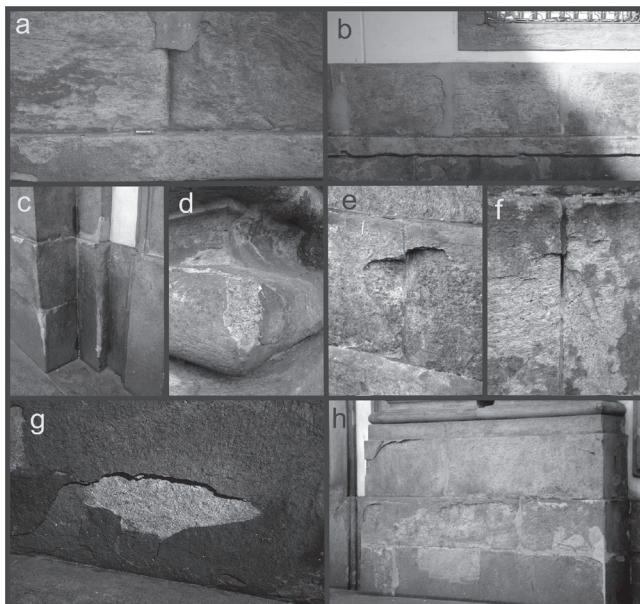


Figure 2. Photograph showing the main pathologies observed in the buildings on the street facing façade of near to street level.

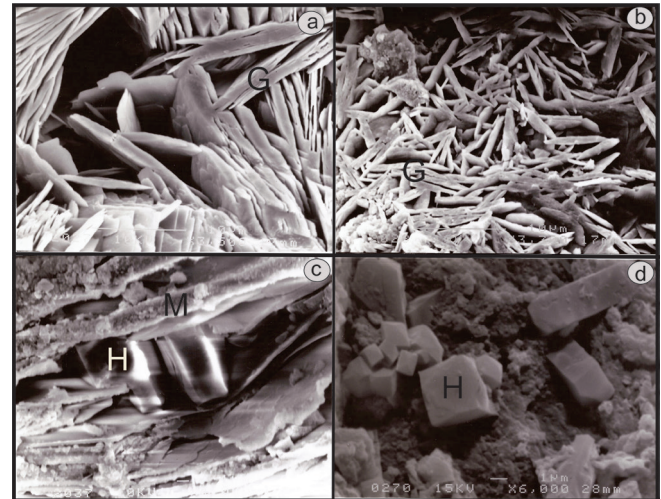


Figure 3. SEM micrographs of black crusts showing (A and B) needle-like crystals of gypsum accumulated in rock façade and halite crystals gaps between open cleavage planes in micas causing deformation and breaking (C), and in the surface of the rock (D)

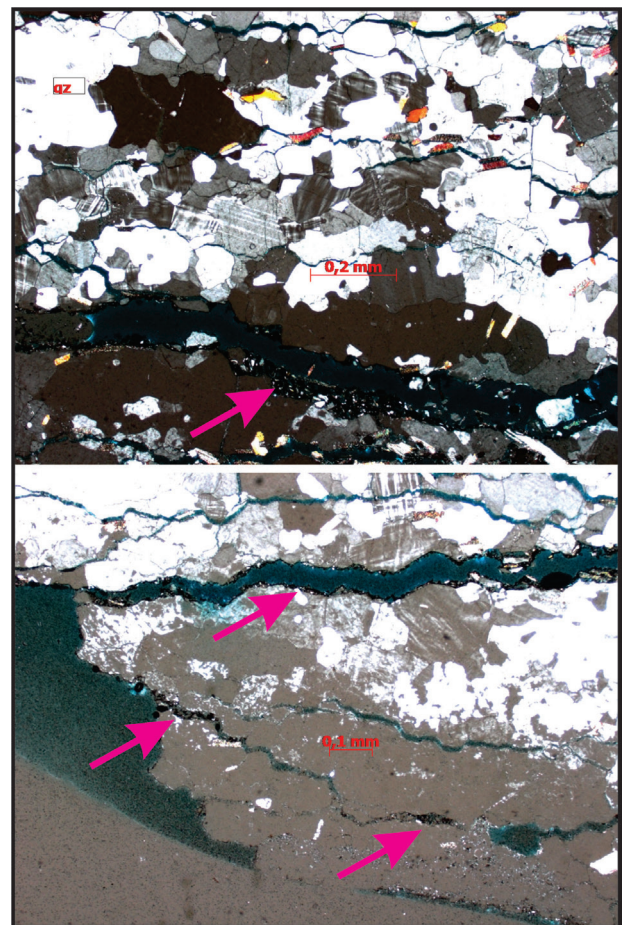


Figure 4. Petrographic thin section showed the formation of gypsum crystals in the rock microfractures

The geochemical analyses performed by ion chromatography and AAS (Table 1), showed the high concentrations of Ca, Na, Cl and SO_4 in all the samples. The strong correlation between elements such as Ca and SO_4 , shown in Figure 5, confirms the presence of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) in all the building façades; this salt has already been observed by other studies in Rio de Janeiro (Smith and Magee, 1990; Smith et al., 2004; Baptista Neto et al., 2007; Baptista Neto et al., 2011; Gaylarde et al., 2017). Strong correlation was also observed between the elements Na and Cl, which confirmed the presence of halite (NaCl), related to interaction with marine aerosols from the sea in Guanabara Bay. These two salts play a very important role in stonework weathering and are very effective agents of salt weathering once they

find their way into the stonework (Smith et al., 2002). Several authors have shown that black crusts may also have an important biological component; numerous organisms and microorganisms have been detected in association with the crusts (Ortega-Calvo et al., 1991; Crispim et al., 2006; Gaylarde et al., 2007). Biological colonization and the growth of organisms, including algae, fungi, lichens, bacteria, and cyanobacteria, on the stone surface reduces the value of the monument (Pozo-Antonio et al., 2016; ICOMOS, 2008). This biological growth can contribute to both physical and chemical decay (Pierivittori et al., 2004; Gaylarde et al., 2012) and microorganisms were, indeed, identified in the current analyses.

FE-SEM with EDS analysis allowed the detection within the

Table 1. Water-soluble anions and cations (ppm)

Location	Cl ⁻	NO_3^-	SO_4^{2-}	Na^+	Mg^{2+}	K^+	Ca^{2+}
São José	176.13	123.5	357.4	276	43.8	187	2837
São José	65.51	73.9	150.4	65	21.4	190	2954
São José	164.6	358.4	2156.8	286	16.7	288	2376
São José	237.9	388.3	3040.87	286	48.5	240	2439
Nossa Sra do Carmo	33.26	70.00	131.50	20	4	50	56
Nossa Sra do Carmo	24.30	51.48	143.08	20	3.5	50	57.5
Nossa Sra do Carmo	55.50	160.92	303.20	35	6	65	109.5
Nossa Sra do Carmo	405.40	142.31	6730.78	150	20.5	180	2750
Nossa Sra do Carmo	227.32	373.19	7079.05	150	43	130	2850
Nossa Sra do Carmo	223.12	97.28	1239.47	185	24.5	160	450
São Francisco Xavier	431.79	414.8	278.975	135	19	65	305
São Francisco Xavier	24.48	49.785	176.3	25	7	75	75
São Francisco Xavier	344.12	1147.34	2750.375	300	33.5	220	1160
São Francisco Xavier	396.33	1160.065	7693.735	200	42.5	220	3600
São Francisco Xavier	415.01	1162.24	6944.55	200	26	205	2850
São Francisco Xavier	1646.09	3096.34	7835.09	750	125	460	3950
São Francisco Xavier	89.28	194.93	3713.515	95	17	125	1345
Candelária	170.32	75.19	99.12	45	15.5	15	85.5
Candelária	389.79	376.26	347.82	215	33.5	55	305
Candelária	172.32	113.11	224.64	55	5	95	255
Candelária	597.63	984.25	3167.13	475	28.5	175	1754
Candelária	414.98	843.76	3765.32	320	38.5	190	2973
Candelária	353.75	917.38	4892.51	275	19	185	1894
Candelária	487.84	1098.48	4623.95	490	35	375	2739
Candelária	344.24	238.92	1083.91	215	25	95	1495
Candelária	567.37	2156.13	6073.92	575	47.5	475	3775
Candelária	647.32	3050.9	7683.19	850	55	550	3965
Santa Cruz Fortress	11	0	29	27	6	18	19
Santa Cruz Fortress	267	30	176	169	119	130	41
Santa Cruz Fortress	2070	2923	7998	6559	0	53	761
Santa Cruz Fortress	123	114	5753	178	9	1681	2740
Blank	87.6	0	0	0	0	0	0

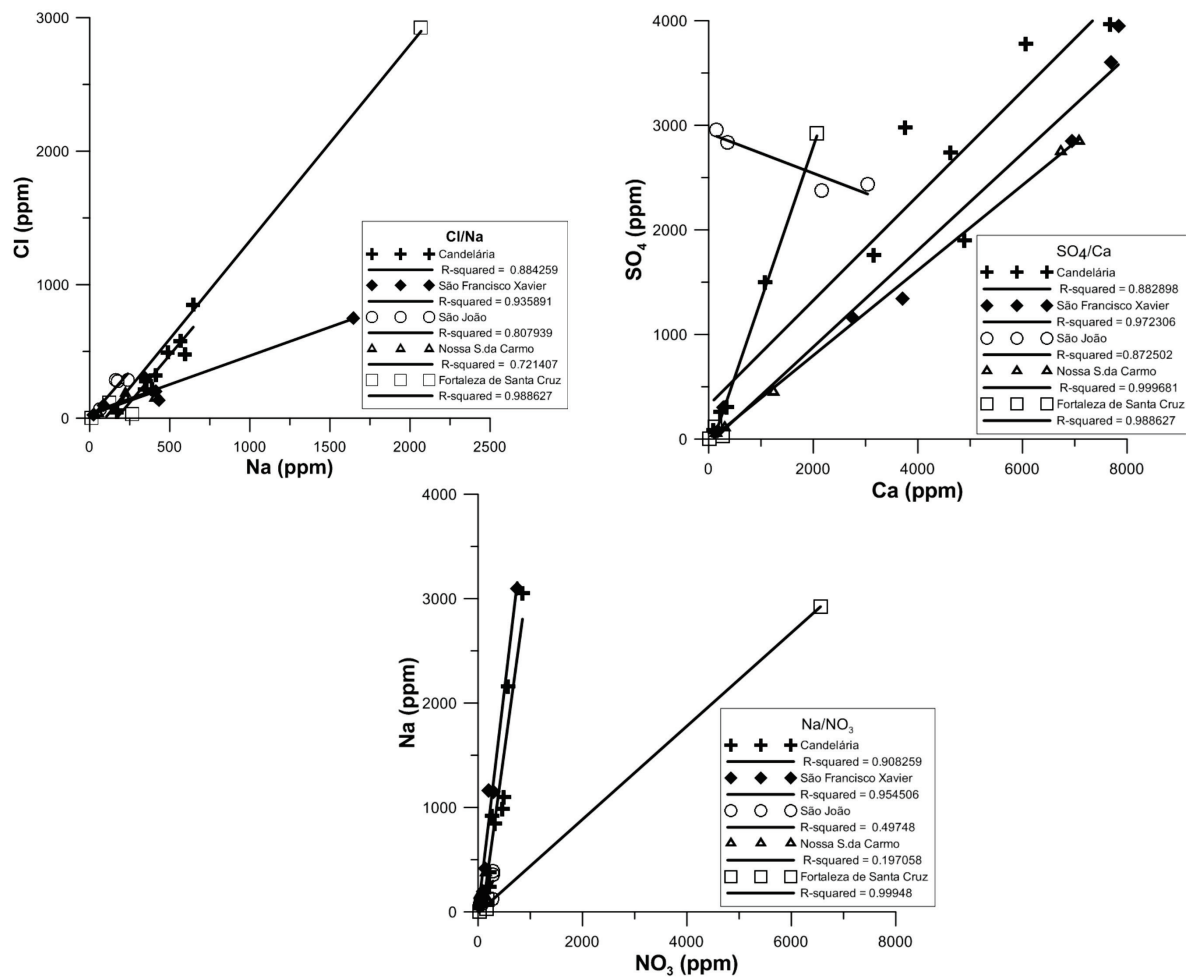


Figure 5. Correlation between some chemical elements.

rock of few filamentous fungi, groups of bacterial cells, rare diatoms and, especially interesting, filamentous photosynthetic bacteria (cyanobacteria) encrusted with re-precipitated gypsum (Figure 6). A wide range of biological organisms are known to solubilize minerals within rocks (Meldrum and Cölfen, 2008). Sterflinger and Pinar (2013) suggested that fungi could be the most important microorganisms involved in stone degradation, but it is also known that cyanobacteria can penetrate and grow within rocks (Gaylarde et al., 2012), obviously causing physical damage merely by their presence. However, the ability of cyanobacteria to metabolically solubilize and then either allow or actively cause redeposition and relocation of minerals is well documented in salt crusts (Canfora et al., 2016), as well as sandstone buildings and itacuru, an iron-rich type of rock used in the Jesuit missions in South America (Tazaki et al., 2009; Barrionuevo et al., 2016). The gelatinous sheaths of both filamentous and spherical cyanobacteria are considered to be nucleation sites for calcium deposits (Gerbersdorf and Wieprecht, 2015),

and the cyanobacteria detected in the samples of this study could also be, at least in part, responsible for the abundant calcite crystals found, as could filamentous fungi (Bindschadler et al., 2016).

The surface discoloration of the façades was certainly increased by microbial colonization and the microorganisms present were identified using the Next Generation DNA Sequencing technique. The populations detected were typical of those occupying stressful environments, in this case, high temperature stone surfaces with little or no available nutrients and hard-smooth surfaces which encourage rapid water run-off (Gaylarde et al., 2017a; 2017b). Many of the groups identified contained microorganisms that produce colored pigments (e.g., the bacterial and fungal genera *Kocuria*, *Rubrobacter*, *Rhodotorula*, *Sporobolomyces*, as well as all phototrophs). In addition, a number of groups were thermophilic or halophilic, indicating the high temperatures and salt concentrations of the stone.

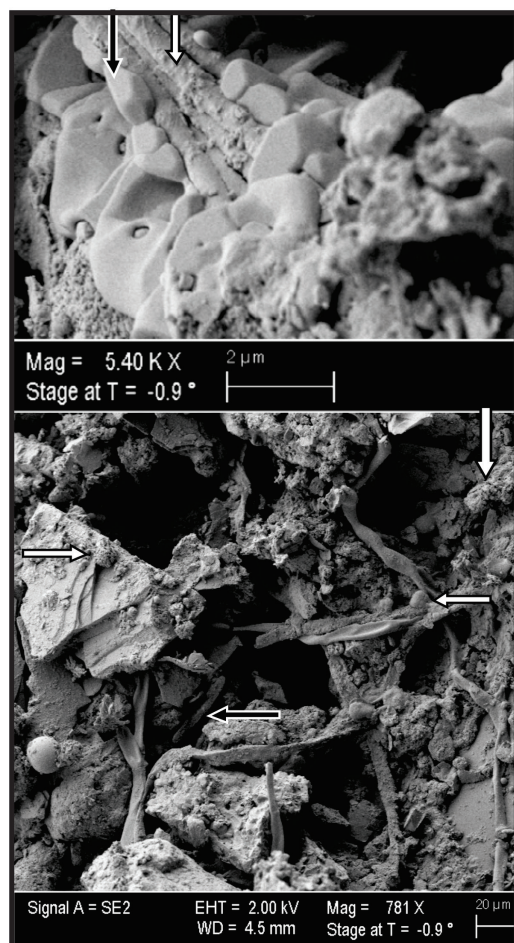


Figure 6. FE-SEM with EDS analysis allowed the detection of filamentous photosynthetic bacteria (cyanobacteria) encrusted with re-precipitated gypsum.

Lichens were seen only occasionally on some of the surfaces (Figure 7); it is likely that their scarcity is due to the polluted atmosphere of central Rio de Janeiro. Lichens are symbiotic associations of fungi with phototrophic microorganisms, algae or cyanobacteria, which are sometimes used as indicators of air quality, since they are very sensitive to atmospheric pollution (Conti and Cecchetti, 2001). The crustose types are especially damaging; they produce rhizoids (small roots), which penetrate into the stone substrate, degrading it (De Los Rios et al., 2009). The foliose lichen *Parmelia saxatilis* was found on the surface of stone parapets at the Fortress of Santa Cruz, Niterói, Brazil (Ogawa et al., 2017b), an area with little urban pollution, next to the sea. When removed, with a hydrogen peroxide (H_2O_2), the damage caused by its penetration into the stone, probably accompanied by production of oxalic acid, could be clearly seen (Figure 7).

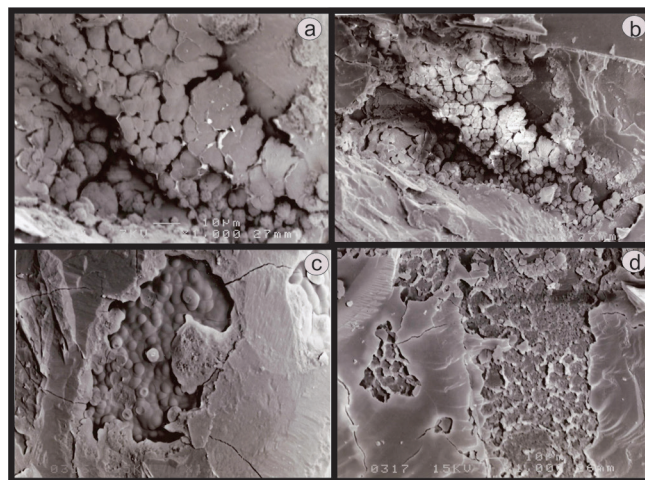


Figure 7. SEM micrographs of the foliose lichen *Parmelia saxatilis* (A and B) was found on the surface of stone parapets at the Fortress of Santa Cruz, when removed, it is possible to observe damage caused by its penetration into the stone (C and D).

Microorganisms can cause stone deterioration in various ways: acid production, physical penetration, mobilization of cations, etc. (Allsopp et al., 2004). Surface discoloration may be caused by pigments within or released from microorganisms. Fungi detected on buildings are frequently dark-colored because of the melanin within their cells and genera with this characteristic were detected in all the buildings sampled in this research. Photosynthetic organisms, algae and cyanobacteria may be green, brown, pink or orange, depending on their production of carotenes and photosynthetic and protective pigments. However, the NGS method used on these samples indicated the presence of very few algae and the cyanobacterial genera identified were not those renowned for dark pigment production. One algal genus frequently found on stone and painted surfaces, the filamentous *Trentepohlia* typically confers a red/orange coloration on the surface, caused by carotene-containing oil that can stain the stone crystals (Scheerer et al., 2009). This alga has been seen colonizing the stone parapets of the Fortress of Santa Cruz, in Niterói (Ogawa et al., 2017b). In the current study, a number of pink-colored bacterial and yeast genera were detected on the stone churches by NGS, including *Rubrobacter* and *Rhodotorula*. The latter yeast genus has been shown to be responsible for a pink stain on the internal walls of Évora cathedral, Portugal (Rosado et al., 2014). However, even in the case of non-pigmented microorganisms, they often produce polymeric materials outside the cells that can trap dirt and organic molecules, thus promoting the dirty appearance of the building. The formation of such biofilms is certainly a contributing factor to stone deterioration and degradation.

5. CONCLUSIONS

The damage of historic stone buildings, monuments and architectural places in Rio de Janeiro is apparently accelerating. The structures are experiencing a rapid block retreat, showing symptoms of severe stone decay, which has increased in recent decades. The stone used in the buildings examined in this report, augen gneiss and granitic rock, is considered very resistant. Nevertheless, most of the historic church façades were found to experience severe degradation, mainly iron staining, granular disintegration, blistering, incipient fractures, and contour scaling. The field work, and the petrographic, SEM, chemical and biological analyses have identified potential components of importance for the understanding of the processes affecting the degradation of the stone façades in historic churches in Rio de Janeiro city. The results of the geochemical study (ion chromatography and atomic absorption) demonstrated high concentrations of several chemical elements that prove the importance of salts in the weathering processes; the elements presenting the highest concentrations were: SO_4 , NO_3 , Cl, Ca, K and Na. The correlations between these elements demonstrated the occurrence of several salts; however, the main salts present in all buildings were halite (NaCl) and gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). The occurrence of these two salts was confirmed by SEM and petrographic analyses. These two salts have already been demonstrated as very effective combined agents of salt weathering in stonework. This type of “anthropogenic” weathering process acts concomitantly with natural processes, speeding up the degradation that would otherwise occur over a longer period of time. Not only salts, but the biological colonization on the façades contributed to the physical and chemical decay. The analyses proved the occurrence of the following organisms: filamentous fungi, groups of bacterial cells, rare diatoms and, especially interesting filamentous photosynthetic bacteria (cyanobacteria) impregnated with newly formed gypsum.

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This paper is dedicated to the memory of Professor Bernard J. Smith (Queen's University of Belfast).

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