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ENVIRONMENTAL IMPACT ON STONE DECAY: CRUST FORMATION AT THE COLOGNE CATHEDRAL

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Abstract

Different building stones of the Cologne cathedral show a large variation of weathering phenomena. "Drachenfels" trachyte, which was the original building material of the medieval construction period, shows significant surface deterioration as well as massive formation of gypsum crusts. Crust formation on limestone, sandstone, and volcanic rock from the Cologne cathedral as well as from the Xanten and Altenberg cathedrals are investigated. These three buildings located in different areas and exposed to varying industrial, urban, and rural pollution, show varying degrees of deterioration.

The material investigated ranges from thin laminar crusts to black framboidal crusts, which incorporate particles from the pollution fluxes. Major and trace element distribution analyses with LA-ICP-MS on thin cuts show an enrichment of sulfur, indicating the presence of gypsum, lead and other pollutants which generally can be linked to traffic and industry. This indicates that even though the SO_2 emission decreased, the pollutants are still present in the crusts on the building stones. SEM observations confirm that the total amount of pollution is less pronounced in the Altenberg cathedral compared to the Cologne and Xanten cathedrals. The obtained data show lower amounts of gypsum formation in the Altenberg samples. This correlates well with the site specific SO_2 concentration and the intensity of the stone decay at the different locations. The black weathering crusts on Drachenfels trachyte contribute to the degradation of the historic building material. The stone degradation due to mechanical moisture related deterioration processes and chemical corrosion of rock forming minerals is enhanced by salt deterioration processes.

The combination of different analytical techniques made it possible to distinguish samples from different environmental situations. If data are compared to actual pollutant emissions, the analyzed samples imply present but also past pollution fluxes. Thus, the soiled zones of the built environment can function as environmental proxies.

Keywords: Cologne, Xanten, Altenberg cathedrals, Drachenfels trachyte, black weathering crust, pollution impact, stone decay processes

1. Introduction

The cathedral of Cologne is one of the most important cultural monuments of northern Europe and faces severe stone deterioration. The effect of air pollution onto stone decay has been a subject in the field of stone deterioration for long (Kaiser 1910;

Grün 1931; Winkler 1970; Luckat 1973-1977; and listed in Charola and Ware 2002). Mostly debated is the crust weathering of limestone as a matter of transformation of calcium carbonate into calcium sulfate due to the impact of air pollutant concentration in the atmosphere and the deposition of anthropogenic sulfur. Although the SO_2 concentration decreased over the last years, degradation still advances in the context with weathering crusts. Pollution has changed to a multi pollutant situation with increasing particulate matter, enhancing the acidic impact in terms of dust deposition. Already Grün (1931) addressed the environmental influences as deterioration factors for the building stones of the Cologne cathedral. Knetsch (1952) pointed out the geological and climatic context of the deterioration of the Cologne cathedral. Efes and Lühr (1976) accentuated the influence of environmental pollutants as a significant factor in stone decay. Further studies dealt with different deterioration processes in several natural building stones of the Cologne cathedral (Wolff 1972; Kraus 1980, 1985, 1985a; Mirwald et al. 1988; Knacke-Loy 1989). The investigation of stone deterioration due to weathering crusts, not only has to consider contemporary or recent emission but also the pollutant concentrations of the past.

In this paper the formation of black weathering crusts as a function of pollution conditions on different building stones in three different environmental settings is discussed. Crust formation on Drachenfels trachyte, a building stone used in the Cologne cathedral as well as in the Xanten and Altenberg cathedrals is investigated and compared to crust formation on limestone of the same cathedrals. The three cathedrals are major gothic buildings of the Rhineland region, built during the 13th and 14th century implementing the same construction material. Construction started in Cologne at 1248, in Xanten at 1263, and in Altenberg at 1259. The main construction material of the medieval building period is Drachenfels trachyte from the quarries of the Siebengebirge. Later restoration phases mainly in the 19th century employed similar building materials at all three monuments: “Stenzelberg” latite, “Obernkirchen” and “Schlaitdorf” sandstone, and “Krensheim Muschelkalk” as well as “Londorf” basalt lava. Mortars employed were lime mortars in the mediaeval period and customary made cement mortars in the 19th century.

The building stones show severe deterioration phenomena. Black laminar and framboidal crusts cover the building stones. Especially on Drachenfels trachyte the crusts tend to detach, and further structural deterioration follows. Contour scaling and flaking are characteristic decay features on Drachenfels trachyte, leading to granular disintegration, crumbling and total fabric collapse.

2. Environmental settings

The three buildings are located in different environmental settings. The Cologne cathedral (53 m above NN) is located in a metropolitan center with one million inhabitants next to the river Rhine. The Xanten cathedral (22 m above NN*)* is the catholic church of a small city at the Lower Rhine with 21,500 inhabitants and with a certain industrial impact. In contrast to the afore mentioned buildings, the Altenberg cathedral (149 m above NN*)* lays in a rural area surrounded by forested hills in the “Bergisches Land”.

The western German climate is maritime influenced with mild winters and moderate summers. The buildings are exposed to air pollutants of anthropogenic impact.

These are mainly gaseous pollutants like SO_2 and NOx, which are currently both decreasing in its concentration. This corresponds to an increase in the precipitation pH (Fig. 1). The effect of particulate matter (PM10) in form of settling dust shows no clear trend yet. It is monitored since 2003/2004; the highest values are reported for Xanten and the lowest for Altenberg (Fig. 1). The relatively high values for Xanten in comparison to Cologne may be explained by a certain pollution impact from the bigger city of Arnhem, NL from where pollution fluxes are transported by west winds. The comparable low values for Altenberg reflect a low impact region and thus a rural area. The Cologne cathedral is located in the city center of a metropolitan industrial region next to the main railway station, which is an active traffic interchange since the onset of the Industrial Revolution.

3. Methods

To address the problem of stone deterioration due to weathering crusts related to atmospheric pollution, different mineralogical and geochemical methods are combined. A sample field at the Cologne cathedral was mapped, analogous to Fitzner et al. (1995) illustrating the context of building stone variation and deterioration features. Samples from host rock, crusts and decay material were collected at the Cologne, Xanten and Altenberg cathedrals from the different building stones. Analyses were performed on the different sample types by using mineralogical and geochemical methods. The chemical composition was obtained by X-ray fluorescence spectroscopy using a Phillips PW1480. Sulfur and carbon were additionally analyzed by METALYT CS 1000RF (Eltra GmbH). LA-ICP-MS (laser ablation inductively coupled plasma mass spectrometry) analyses were performed on thin slices perpendicular to the deteriorated stone surface. The laser used was a Compex 110 Excimer (ArF 193 nm) by Lambda Physik, with an ablation pit diameter of 120 µm, laser pulses of 10 Hz repetition rate, and 3 J/cm^2 available energy. The mass spectrometer is a Perkin Elmer DRC II (Siex, Canada). To visualize micro-fabric of crust, host rock and to detect elemental-mineralogical composition of samples SEM-EDX techniques were applied (LEO GEMINI SEM 1530 and 1455 as well as AMRAY 1630). EDX-analyses were performed on a Quantra 200F (Fei) with a field emission cathode with initial stress of 20 kV.

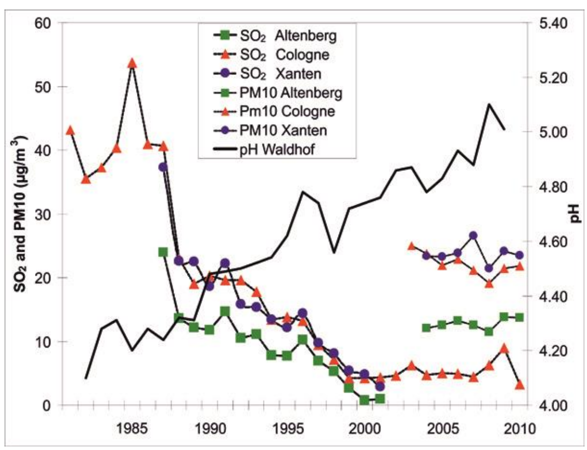

Figure 1. SO_2 and PM10 concentrations of Altenberg, Cologne and Xanten versus pH precipitation (Waldhof), 1981 – 2010 (mean annual fluxes) data compiled after LANUV 2010 and UBA 2011.

4. The decay of Drachenfels trachyte

Drachenfels trachyte was used for construction in the Rhineland since Roman time (Wolff 2004) and was the main construction material for the three investigated buildings. The porous trachyte has a light grey color. The main components are phenocrysts of sanidine, which are included in a micro to cryptocrystalline matrix composed of feldspar and quartz. The sanidine phenocrysts, of a size up to 7 cm, are a characteristic feature of the stone and generally show a preferred orientation, tracing the flow fabric. The rock consists mainly of sanidine (50%), plagioclase (24%) and quartz (13%). Other components are augite (5%) and biotite (5%) and common accessory minerals are ore (2%) and apatite, zircon and sphene (1%) (Grimm 1990). The porosity shows values of 12%. (Graue et al. 2011)

The different building stones of the Cologne cathedral show a large variation of weathering phenomena. Namely Drachenfels trachyte shows severe decay in the form of cracks, erosion and surface recession coexisting with flaking and structural disintegration to crumbling as well as the massive formation of gypsum crusts. Areas of surface recession often display a stronger further decay in terms of microcracks and crumbling to total fabric collapse. Scaling is observed and scales often show a granular disintegrated zone on the reverse side, whereas the original stone surface generally still exists (Fig. 2). Formation of fissures may also propagate many centimeters into the stone. On Drachenfels trachyte the formation of thin laminar crusts as well as thick framboidal crusts is observed. Black framboidal crusts tend to bulge out and detach from the stone surface. The stone structure in the background of these crusts is strongly weakened and disintegrates in form of multiple flaking, exfoliations and further crumbling. Thin laminar crusts often build on structural intact stone surfaces but are often accompanied by contour scaling. These surface parallel scales show a thickness of a few millimeters to 1–2 centimeters with the formation of a brittle zone on its backside.

The three cathedrals show clear differences when compared with one another in terms of deterioration gradients. While the Altenberg cathedral displays only very little stone deterioration, the cathedrals in Xanten and especially in Cologne show significant decay. At the Cologne cathedral severe damage can be observed presuming static disturbances.

5. Crust classification

The formation of crusts is widespread over the complete building of the Cologne cathedral. This surface deterioration feature appears in different manifestations, from patina like, grayish black surface depositions and soiling, to thin black laminar crusts and thick framboidal or cauliflower like black crusts. Especially in the context with framboidal crusts, surface detachment and loss as well as further disintegration are observed. These cauliflower-like forms of weathering crusts have an extremely varying thickness from 2 to 15 and even 30 millimeters (Török et al. 2011). Their specific morphology also named them as dendritic, globular, ropey or framboidal (Török et al. 2011 and references therein). They contain a large amount of gypsum, as well as calcite and quartz particles which cover the stone surface and incorporate organic as well as inorganic particles from the pollution fluxes. They generally build in sheltered to moderately exposed areas as well as in cavities on vertical stone surfaces. Thin laminar black crusts trace the stone surface and may cover complete sections of the building's structure, not necessarily preferring protected sites. This kind of crust does not change the morphology of the stone surface (Fitzner et al. 1995) and seems to have very strong bonds between the crust and the stone surface (Török et al. 2011; Siegesmund et al. 2007).

The other building stones of the Cologne cathedral also show crust formation. At Schlaitdorf sandstone, massive gypsum crusts build due to the carbonate cement (app.

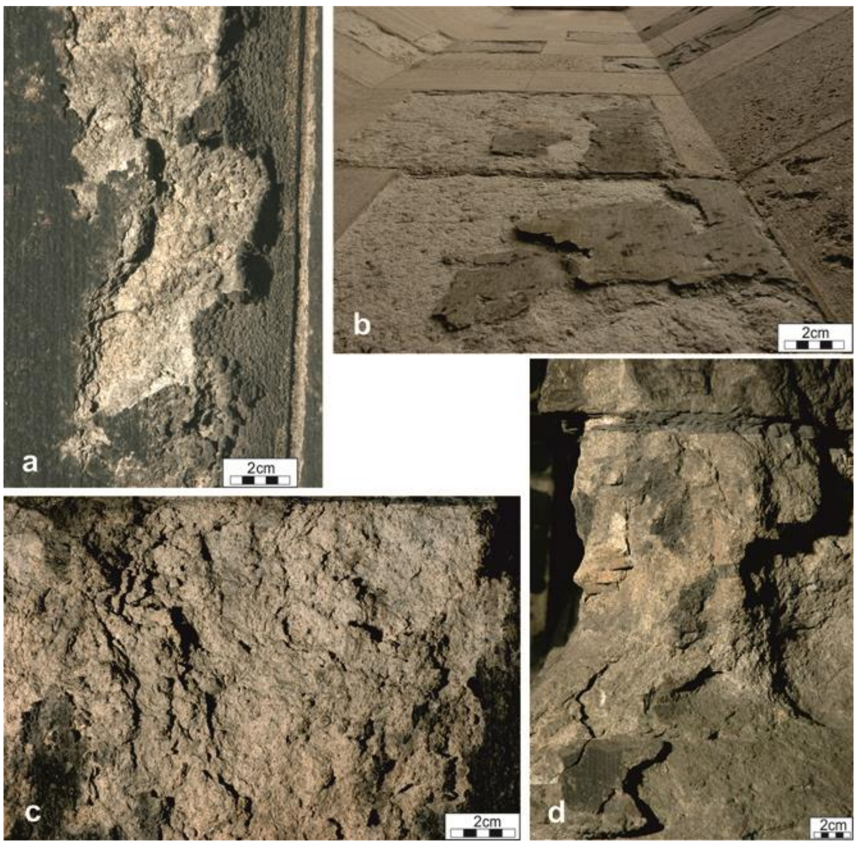

Figure 2. Weathering forms of Drachenfels trachyte. **a:** framboidal weathering crust showing bulging and flaking of the rock underneath; **b:** deformed and detached scale; **c:** surface deterioration and flaking; **d:** structural disintegration and crumbling to total fabric collapse.

14%) leading to characteristic deterioration in form of contour scaling of 2–4 cm thickness, flaking and granular disintegration (Grimm 1990). The main deterioration phenomenon of Stenzelberg latite is a typical formation of scales with a thickness of 2–3 mm, which tend to detach from the stone. Obernkirchen sandstone has a high weatherability, but also shows thin black crusts as well as framboidal crusts in sheltered areas. Krensheim Muschelkalk, as a carbonate building stone, shows massive gypsum crust formation in rain protected areas. These crusts seem to temporarily stabilize the stone surface (see Siegesmund et al. 2007). On surfaces exposed to rain, solution phenomena can be observed, e.g. microkarst (Graue et al. 2011).

6. Mineralogy and fabric

Microscopic observations reveal high porosity for the framboidal crusts (Fig. 3a) and very small and evenly spread acicular gypsum crystals (10–50 µm). They form cavities causing the high porosity of the crust. The crust contains a significant amount of widely spread organic matter (black opaque particles). The crust formation on Drachenfels trachyte is characterized by a thin dense black layer on the surface of the host rock (10–20 µm) mainly consisting of organic material, which may function as a catalyst for the formation of gypsum (Rodriguez-Navarro and Sebastian 1996; Ausset et al. 1999). On this surface layer a porous framboidal crust builds with finely distributed gypsum crystals and crystal aggregates together with quartz and feldspar particles (<0.1 mm). The brownish tanning derives from iron-oxides/hydroxides (Do 2000). Framboidal black crusts on Drachenfels trachyte can be described as a porous composition of gypsum, organic compounds, iron-oxides, quartz, and feldspar particles but very little calcite. A large number of siliceous as well as carbonaceous fly ash particles are commonly embedded in such crusts (Török et al. 2011). SEM-EDX analyses of the crust surface reveal a high gypsum concentration for framboidal crusts, whereas on laminar crusts a composition mainly of silicate and organic components is detected.

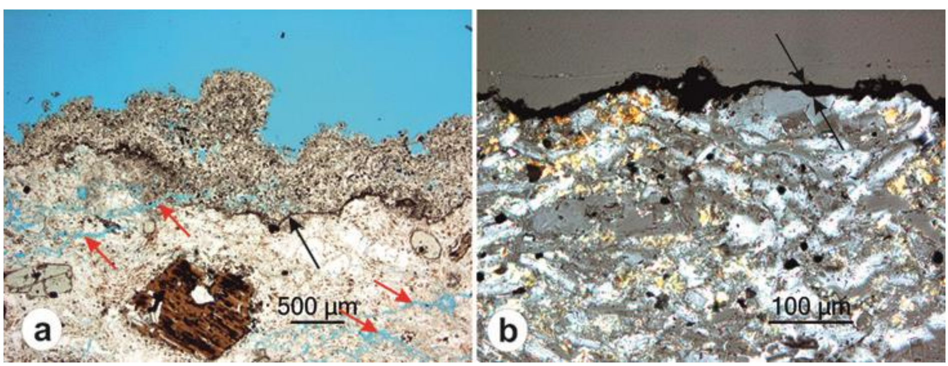

Figure 3. Crusts on Drachenfels trachyte **a:** framboidal crust (crossed polars) with a black interface layer (black arrows) and surface parallel cracks (red arrows) in the host rock and crust; **b:** laminar crust (crossed polars) with a very thin opaque surface layer.

The thin opaque black layer on the surface of the host rock marks the boundary of the stone, onto which the crust forms. In some places, this defined line is distorted, which may be attributed to the structural disintegration of the stone material underneath. Surface parallel cracks are often observed in the host rock beneath the crust. These cracks not only run along grain boundaries, but also characteristically cut through larger grains and minerals (Fig. 3a). This structural degradation of the host rock finally leads to the detachment of the crust including the upper surface region of the host rock. The width of this detached zone is not limited to the crust but reaches into the host rock with a thickness of 3–10 mm. The host rock shows further structural disintegration in the form of multiple flaking and exfoliation. Laminar crusts are very thin (5–15 µm, where bulging occurs up to 50 µm) and have a dense composition of mainly organic compounds with parts of gypsum, iron-oxides, quartz and feldspar particles (Fig. 3b). As observed in the context with framboidal crusts, the host rock beneath laminar crusts also shows surface parallel cracking but with a lower quantity and latitude of the cracks.

7. Geochemical characterization

The results of the XRF analyses show a similar chemical composition of the black weathering crust compared to the silicate host rock with an enrichment of calcium and sulfur (Tab. 1). An enrichment of CaO within the crust is clearly noticeable for the samples from Cologne (177%) and from Xanten (75%). The average CaO-enrichment in the Altenberg samples is within the measurement accuracy. The increase in SO_3, respectively the sulfur concentration, shows a similar tendency: Cologne samples have an enrichment of sulfur with factor 139, Xanten with factor 65 and the Altenberg samples with factor 7 in respect to the host rock. The depletion of the oxides associated with silicate phases (SiO_2, Al_2O_3, Na_2O, K_2O) correlates with the increase in SO_3. The mean enrichment of Fe_2O_3 is for the samples from Altenberg 12%, in Xanten samples 16% and in Cologne samples 11%. Noticeable is the increase of P_2O_5 concentration (5–188%).

Table 1. Main element composition of samples (data set of XRF analyses in wt. %)

sample	No. sample	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	H_2O	CO_2	SO_3	∑
host rock	n = 5	64.31	0.72	16.97	3.49	0.11	0.85	2.43	4.56	5.22	0.18	0.92	0.22	0.04	99.9
Altenberg	n = 3	63.17	0.77	16.68	3.92	0.09	0.63	2.41	4.58	4.69	0.19	1.56	1.21	0.30	100.2
Cologne	n = 10	54.45	0.65	13.79	3.86	0.09	0.68	6.72	3.71	4.05	0.51	0.92	0.92	5.90	100.3
Xanten	n = 5	59.80	0.71	15.12	4.06	0.14	0.74	4.24	4.24	4.39	0.22	0.99	0.54	2.77	99.8

In crusts on limestone, where the substrate consists almost entirely of $CaCO_3$, a depletion of CaO and an enrichment of SiO_2 as well as aluminum and iron in the crust are detected (Török et al. 2011). The contrary is found for crusts on silicate stones: a depletion of oxides associated with silicate phases and an enrichment of CaO in the crust along with an enormous increase in sulfur, indicating high gypsum enrichment in the crust. The average sulfur concentrations normalized to the host rock correlate to gypsum contents for the samples of Altenberg of 0.6 wt. %, for Xanten of 5.9 wt. % and for Cologne of 12.6 wt. %. The sulfur concentration found in crust samples on limestone – investigated by Török et al. 2011 – correlates to an average gypsum amount of 22 wt. %.

8. Micro chemical characterization

LA-ICP-MS analyses show clear trends in terms of major and trace element distribution in the black crusts and the host rock. The micro chemical investigation reveals little change in oxides associated with silicate phases. An enrichment of sulfur, lead, antimony, bismuth and arsenic in black weathering crusts developed on Drachenfels trachyte is detected (Fig. 4). The

Table 2. Mean values for S, Ca, Pb, Sb, Bi, and As in the host rock and in the crust from the samples of Altenberg, Cologne, and Xanten.

	sample No. n	lead (Pb)		antimony (Sb)		bismuth (Bi)		arsenic (As)	
		value ppm	increase factor	value ppm	increase factor	value ppm	increase factor	value ppm	increase factor
host rock	n = 1	18		0.3		0.02		1.8	
Altenberg	n = 1	890	50	12.2	37	1.08	48	28.0	16
Cologne	n = 2	1849	105	37.2	114	3.26	145	367.4	205
Xanten	n = 4	1944	110	15.5	48	2.64	117	50.4	28

crusts of the Cologne cathedral show an average concentration of 1,849 ppm Pb, those of Xanten 1,944 ppm Pb, while the crusts of Altenberg only show 890 ppm Pb – which corresponds to enrichment factors of 105, 110 and 50 in respect to the host rock (Tab. 2; Fig. 4). The samples from the three locations are clearly distinguishable. Comparing industrial, urban and rural samples, the data show high concentrations of heavy metals (e.g. Pb, and Bi), As and Si in black crust samples collected from the industrial and urban sites (Cologne and Xanten). The sample from the rural area (Altenberg) contains significantly lower concentrations of heavy metals. This clearly indicates a very strong pollution impact for the Cologne and Xanten samples, since the content of these elements is due to the impact of combustion emission.

In view of decreasing SO_2 fluxes, the ban of leaded petrol, and emission regulations, the high concentrations of heavy metals as “anthropogenic combustion tracers” in the samples from Cologne result not only from recent air pollution but show the long history of industrial development at this place. The high lead concentrations in the Xanten samples can be traced back to a strong industrial impact of Arnhem (NL) over a longer period of time.

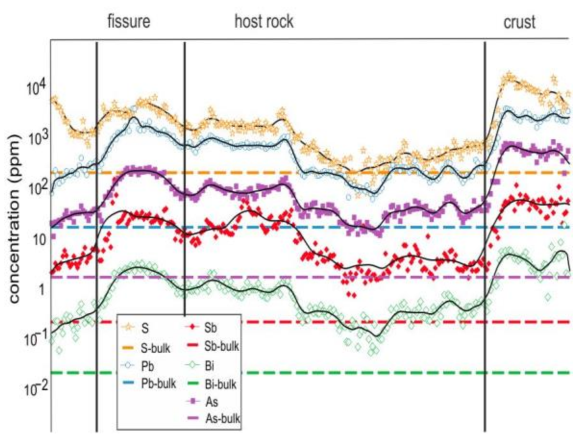

Figure 4. LA-ICP-MS analyses along a profile perpendicular to the surface on a thin cut of a black weathering crust (Drachenfels trachyte; Cologne cathedral). Concentration profiles of S, Pb, Sb, Bi and As normalized to the unweathered host rock (dotted lines).

9. Discussion

The investigation shows, that gypsum crusts not only build on calcareous stones but also on silicate stones. In comparison to black weathering crusts on limestone, the crusts on silicate Drachenfels trachyte are much lower in their gypsum content. Limestone is a vast source for calcium ions to the formation of gypsum crusts, whereas Drachenfels trachyte, with an original content of 1.73 wt. % Ca does not primarily tend to form gypsum. For crusts on limestone a significant difference in chemical composition between host rock and crust is reported (Török et al. 2011) due to the transformation of $CaCO_3$ to $CaSO_4$. This clearly is not the case for black weathering crusts on Drachenfels trachyte, where the crusts show a similar composition for several elements like the host rock, but enrichment in non-silicate phases. The crusts have higher sulfur and calcium concentrations relative to the fresh stone (enrichment factor for sulfur: 8–148 and for calcium: 1–3). The original low calcium (1.73 wt. %) and sulfur (< 0.02 wt. %) in Drachenfels trachyte indicate an external source for sulfur and calcium required for the crust formation.

Sulfur derives from anthropogenic pollution impact and is imported via wet and dry deposition (Charola and Ware 2002). Primarily dry deposition of SO_2 in the context with corresponding humidity (e.g. fog, condensation) functions as sulfur source for the gypsum formation (Torfs et al. 1997). The calcium import comes from aerosol deposition and leaching products from mortars, e.g. joint mortar (Snethlage and Wendler 1997; Kraus 1980) as well as from neighboring calcareous stones. The role of particulate matter in the form of dry and wet deposition and their contribution to the formation of black weathering crusts has already been discussed (Charola and Ware 2002 and cited within). Airborne particles also include calcium rich aerosols, which therefore have their part in calcium import. Already Grün (1931) and Wolff (1972) considered mortars; e.g. joint mortars, as a potential source for the formation of gypsum crusts and related deterioration at the cathedral of Cologne. A commencement of decay features in Drachenfels trachyte from the joints is observed, which is indicated by gypsum crusts, flaking and scaling (Graue et al. 2011). Besides alkaline mortars, also neighboring calcareous stones, especially limestone are in question of contributing as a source for calcium. Already Kraus (1985), Mirwald et al. (1988) and von Plehwe-Leisen et al. (2007) noted that there are strong indications that the decay of Drachenfels trachyte is especially critical when it is placed adjacent to carbonate stones. As surface roughness evaluations on several calcareous stones show, a continuous increase of roughness is detectable indicating the dissolution of calcite from the stone (Grimm and Völkl 1983). Tests on the acid binding capacity show, that at an acidic impact the lime stone readily dissolves and provides calcium ions – with a rate 320 times higher than volcanic stones – which therefore may be transported to other stones and contribute to the formation of gypsum crusts. Investigations on site reveal that crust formation on Drachenfels trachyte can be enhanced by neighboring limestone depending on the exposition of the affected section of the wall (Fig. 5). Krensheim Muschelkalk exposed to an acidic environment solutes into its ion compounds. Rainwater leaches the rock components of the limestone. This calcium ion loaded water runs off and is directly transported to the building stones in flow direction. It is absorbed by Drachenfels trachyte and accumulates in the cavities. Due to the environmental impact of sulfuric

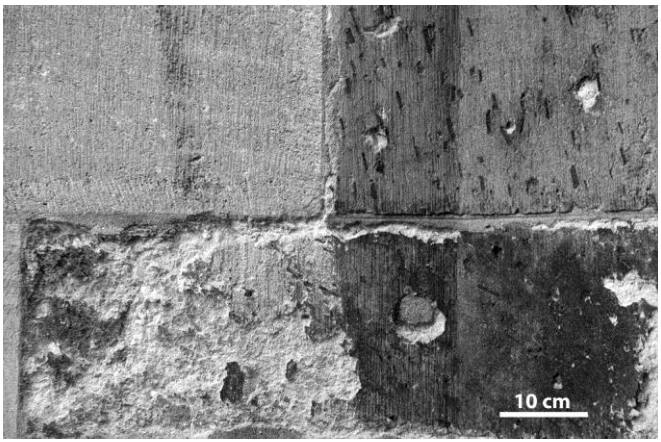

Figure 5. Wall section at the Cologne cathedral. Upper left stone: Krensheim Muschelkalk, upper right and bottom stones: Drachenfels trachyte. The left side of the bottom Drachenfels trachyte is in the flow direction of ion loaded water from the limestone, which enhances the crust formation and decay. As well, a stronger deterioration of Drachenfels trachyte is observed close to the joints, where alkaline leaching products of the mortar might pose a certain impact.

pollutants, salt formation; e.g. gypsum, takes place. As illustrated in figure 5 the affected wall section has to be in the relevant flow direction. Water transport and input have to be in relation allowing enough ions to be dissolved and transported. At the same time, the appropriate amount of water has to guarantee the formation and precipitation of salts. This means that interferences of adjacent ashlars are strongly dependent on the exposition of the wall section in question and the placement of the corresponding stones to each other. However, neighboring calcareous stones may not play an initial role in the formation of crusts, but contribute to it, if extrinsic factors such as environmental impact, exposition, and localization as well as surface characteristics are met accordingly.

Through the investigation it is shown, that crust formation is strongly correlated to the structural deterioration of Drachenfels trachyte. Analyses of depth-specific samples show, that gypsum distribution is coherent with the detected decay phenomena. Microscopic, SEM, and EDX analyses clearly indicate that gypsum enrichment is not only found within the crust but also in deeper zones of the disintegrated stone material. Gypsum accumulates in cracks (Fig. 6a) and on the backside of the detached scales, where significant gypsum formation is observed (Fig. 6b). The detection of structurally disturbed zones, where no gypsum is found (Fig. 6c), suggests that already preexisting fabric and especially mineral inhomogeneities are disrupted due to a chemical and mechanical deterioration impact. Microscopic analyses show not only the displacement of separate grains but also detect the opening of cleavage planes in minerals and their disruption (Fig. 6d). Salt solutions are given new pathways, the formation of gypsum and salt deterioration processes play an essential part in the structural disintegration. This indicates that even though the gypsum content in the crusts themselves is not as high as in the crusts built on limestone, gypsum clearly contributes to the damage process of flaking, scaling and crumbling to total fabric collapse of Drachenfels trachyte.

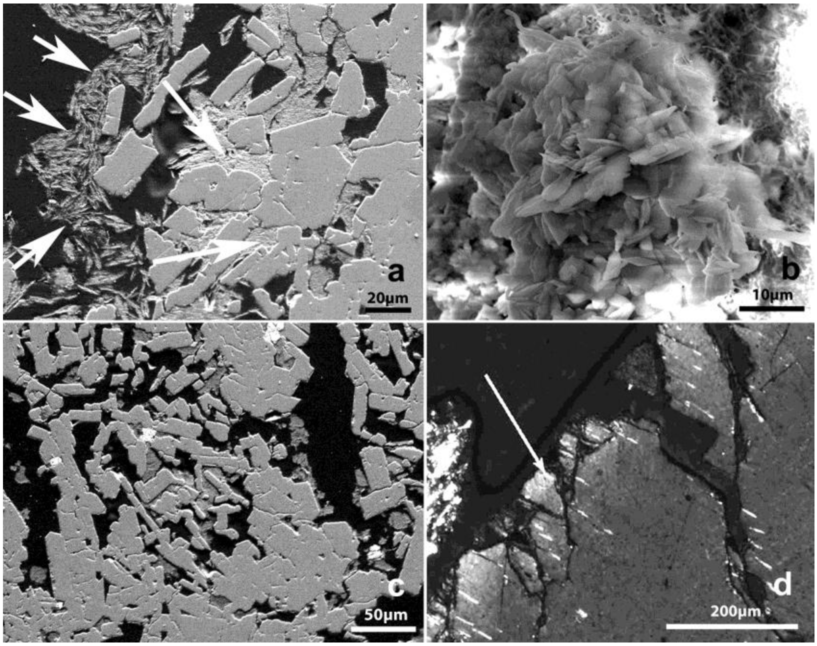

Figure 6. a: Gypsum (marked by arrows) is enriched in the crust as well as in the cracks (SEM); **b:** rosette shaped gypsum crystals on the backside of a scale (SEM); **c:** structural disturbed zones, where no gypsum is detected indicating the corrosion of feldspar grains (SEM); **d:** fine grained birefringent secondary minerals (marked by arrow) formed along the cleavage planes of sanidine phenocryst.

If gypsum formation is compared within obtained data of the three different cathedral sites, data from XRF analyses are consistent with SEM observations. Gypsum formation on samples from Cologne shows a high accumulation of large, occasionally rosette like, gypsum crystals. In comparison to that, gypsum crystals in Xanten are smaller and less abundant. In the samples from Altenberg the gypsum crystals are of even finer grain size and mostly present in the form of salt efflorescence.

10. Conclusions

Investigations on the crust formation on Drachenfels trachyte clearly identify the impact of pollution. The elevated contents of S, Ca, As, Pb, Sb, Bi and As in the crusts mainly result from the impact of combustion emission. The lack of an important intrinsic source for calcium and sulfur for the formation of gypsum crusts demonstrates the major environmental impact. Although SO_2 shows a strong decrease in its concentration over the past 30 years, degradation in the context with weathering crusts is still observed. This indicates that not only recent emission but also the pollutant concentrations in the past have to be considered and that the impact of particulate matter in form of settling dust plays an important role today. In addition, the formation of gypsum crusts can be enhanced by interferences between different building materials, e.g. mortars and neighboring stones (e.g. limestone). Although crust formation is not as significant as on limestone, black crust formation on Drachenfels trachyte correlates with greater structural degradation of the stone. Besides mechanical deterioration processes, chemical corrosion and degradation of rock forming minerals result in structural disintegration.

11. Acknowledgement

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