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Salt-induced alveolar weathering of rhyolite tuff on a building: causes and processes

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Abstract

Studies of stone texture and spatial distribution of soluble salts were carried out at the historical façade of the church of St. Kunigunden in Rochlitz in order to explain the occurrence of alveolar weathering on the building stone, a local rhyolithe tuff. The rhyolithe tuff contains lapilli inclusions with porosities and water uptake quite different to those of the stone matrix. No alveolar weathering was observed in the walls near the ground where gypsum was the dominant salt. In the zone above, which, in addition to nitrates and chlorides, is loaded with magnesium sulphate and gypsum, the lapilli inclusions preferentially weather out due to salt cristallization, thus forming single, isolated pits. The formation of these pits leads to a distinct spatial distribution of magnesium sulphate and gypsum. Magnesium sulphate is accumulated in sheltered position at the bottom of the holes whereas gypsum is concentrated near the surface. Accelerated weathering due to hydration/dehydration activities of magnesium sulphate causes further material loss in the holes. The enlargement of the holes becomes a self-perpetuating process at this stage. Finally they coalesce, forming mature alveolar structures with holes and ridges. The heterogeneous structure of the building stone as well as the high content of magnesium sulphate in the affected zone are preconditions for alveolar weathering in the case investigated in this study.

Keywords

Stone masonry, tuff, lapilli inclusions, salt decay, alveolar weathering

1. Introduction

Stone decay caused by salt attack is a very common phenomenon on façades and stone sculptures. Various weathering forms can be observed, dependent on stone properties and salt load as well as on environmental factors. The most peculiar one is the so-called alveolar or honeycomb weathering. These terms are applied to a pattern of various pits and hollows (alveoli) separated by ridges or walls that develop on the external surface of rocks in nature and of building stones as well. Alveolar weathering has been described from natural environments in coastal areas, deserts and arctic landscapes and also from humid inland environments [Mustoe 1982]. Very similar weathering forms were observed on façades made of limestone [Alessandrini et al. 1992, Rothert et al. 2007] or sandstone [Jeannette 1980, Siedel 2008].

The detailed mechanism of alveolar weathering ist still under discussion. The wind might play a role in the initial formation of alveoli in salt-loaded rocks [Rodriguez-Navarro *et al.* 1999, Quayle 1992]. A crucial point in the discussion is the question of wether honeycomb pattern develop randomly from macroscopically homogeneous rock textures (as demonstrated by a weathering experiment of Rodriguez-Navarro *et al.* 1999), or if incipient honeycomb formation is necessarily related to heterogeneities in rock fabric. The latter is suggested by strata-bound development of alveoli described by Jeannette 1980 and by the preferential formation of alveoli in the boundary zone between foralites (i.e. trace fossils originated by endobenthic animals living in a system of burrows) and the stone matrix [Alessandrini *et al.* 1992, Siedel 2008].

On principle, there are two ways to study the development and the mechanism of salt-induced alveolar weathering. These are, firstly, to simulate the overall process by laboratory studies [cf. Rodriguez-Navarro *et al.* 1999] or theorethical models [Huinink et al. 2004, Quayle 1992]. Secondly, direct observations and detailed, small-scale investigations on building surfaces with respect to stone fabric and spatial salt distribution reflect momentary states within a progressive, dynamic process. The following case study of alveolar weathering displays a methodology for such investigations at the historical church of St. Kunigunden in Rochlitz, Germany.

2. Building and building material

2.1 Historical and environmental background

The church of St. Kunigunden was built in Gothic style in the 15th century. Although the building was affected by war damages and several restoration measures, the ashlar investigated here has not been changed and represents a weathering state that has developed over centuries. Salt efflorescence on bases of the walls can be clearly detected on old photographs of the southern façade from 1875 and was again reported in 1933. A photograph taken in 1965 shows a remarkable progress in weathering compared with the situation in 1875 [Siedel 1998]. The small town of Rochlitz is situated in a valley in the area between the industrial centres of Leipzig and Chemnitz. Accelerated weathering in the 20th century was caused by high environmental pollution [Klemm and Siedel 2002].

2.2 Building stone and mortar

The red rhyolite tuff from Rochlitz (Saxony, Germany) has been widely used as a building stone since the 12th century (see Figure 1). The rock of Lower Permian age (Rotliegend) is a fine grained pyroclastic sediment with flat lapilli in the dimension of some mm to cm. Beside pyroclastic fragments, the stone contains quartz grains up to some mm in size, kaolinite, mainly formed by devitrification processes in the matrix as well as by degradation of feldspars, and small amounts of biotite.

The total water uptake under atmospheric pressure lies between 6.9 and 11.1 wt-%. The compressive strength is 13.5 MPa at minimum and 48.9 MPa at maximum, the tensile strength 5.1 and 9.2 MPa, respectively [Siedel 2006]. The stone is frost resistant and normally in a good weathering state on many historic buildings, except for areas stronger affected by moisture and salts. The main weathering forms in such areas are crumbling, flaking and sometimes scaling and alveolar weathering.

Figure 1: Rhyolite tuff from Rochlitz (figure width = 10 cm).

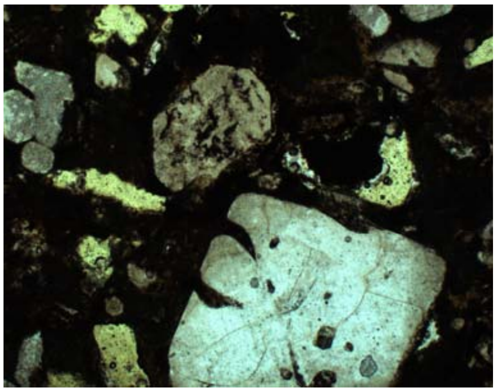

Figure 2: Thin section of Rochlitz tuff (crossed Nicols, figure width = 1.5 mm)

Analyses of the historical joint mortars showed the high magnesium content of their binding agent [Siedel 1998]. The rocks used for historic lime production can be found in dolomite deposits of the Upper Permian (Zechstein) in a distance of about 10 km nordwest of Rochlitz. The rock can contain up to 45 wt-% $MgCO_3$, i.e. burnt lime produced from this material his highly dolomitic.

3. Methods

3.1 In-situ observations and investigations, sampling scheme

A survey of the building including stone deterioration and salt load was made in the framework of planned restoration measures [Siedel 1998]. In the course of these investigations, a mapping of selected representative areas of the southern façade was performed to record the distribution pattern of characteristic weathering forms. Additional laboratory investigations on drill cores taken at different height levels above the ground (see below) provided information about the spatial distribution of salt ions in the walls in decimetre to metre scale. They could be used for a general correlation of weathering forms with salt load. In a selected area with alveolar weathering, addi-

tional drill powder samples from different depths were taken at the bottom of a hole and at the neighbouring ridge for a more detailed study of salt distribution in centimetre scale. Salt efflorescence was scratched off for analysis of salt compounds.
Different stages of the development of alveoli could be visually studied at several places on the façade. Their documentation helped to understand the process of alveolization.

3.2 Laboratory investigations

Analyses of soluble salts in drill core sections were carried out at the University of Hamburg, Institute of Inorganic and Applied Chemistry, with ion chromatography (IC, for anions) and ICP-AES (for cations) after extraction with deionized water.
The drill powder samples from the hole and the ridge were disaggregated in distilled water and filtered. Na was analysed by ion sensitive electrode, other ions in the solution were determined by a spectrophotometer (Hach) using standardized reagents,. The results were referred to dry stone powder (wt-%).
Non-weathered stone samples from drillcores were used for measurements of mercury intrusion porosimetry (MIP), performed with the porosimeters 2000 WS – Carlo Erba for smaller pores (radii < 7 µm) and Pascal 140 – Fisons Instruments for pores with radii > 7 µm. Pyroclastic fragments (lapilli) were isolated from the tuff matrix of quarry samples because it was not possible to obtain material from the historical façade. They were separately measured with MIP. Samples from salt efflorescence were analysed with an X-ray diffractometer Siemens D5000 (XRD analysis).

4. Results and discussion

4.1 Visual observations and mapping

The mapping of a part of the southern façade clearly displays a typical pattern of different weathering forms at different height levels (figure 3). At the base of the wall (up to about 1 m) mainly blackening of the surface and contour scaling was observed. At somewhat higher levels (above 1 m) the dominant weathering forms are weathering out of components (partly with transition to alveolar weathering) and alveolar weathering. The same scheme could be observed on the whole southern façade. In different areas at the higher level, different intensities of weathering could be observed with transitions from local weathering out of lapilli components to the formation of alveoli. Several stages of this process are recorded in figure 4.

4.2 Investigations on lapilli inclusions

Since the loss of lapilli by weathering out seems to be the starting point of the de-velop-ment of pits and later of alveolar structures, the texture of these inclusions was investigated in comparison to the tuff matrix. Results are given in table 1.

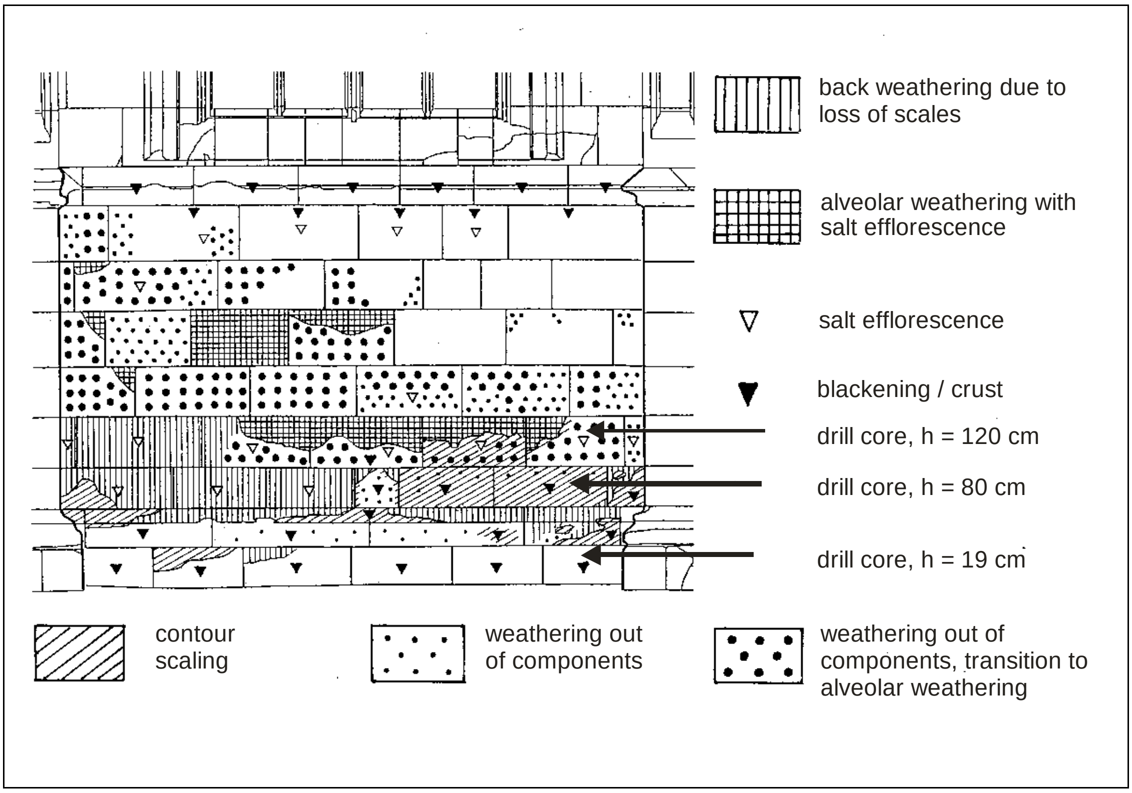

Figure 3: Mapping of weathering forms at the southern façade of the church of St. Kunigunden in Rochlitz with positions of drill cores for salt investigation

Figure 4: Stages of the development of alveoli at the church St. Kunigunden in Rochlitz: Weathering out of single lapilli components (above). Smaller pits can coalesce to bigger alveoli (lower left), salt efflorescence is always present. Further salt weathering forces the formation of a system of holes and ridges (lower right, with the drill holes for detailed salt investigations).

Sample no.	Total water up-take, 24 h [wt%]	Pore radius average from MIP [µm]	Total pore volume from MIP [vol.%]
lap 1	8.4	0.18	23.0
lap 2	6.3	0.50	17.7
lap 3	6.3	n. d	n. d.
lap 4	3.0	0.47	8.3
lap 5	3.2	n. d	n. d.
lap 6	4.9	0.15	7.6
tuff matrix	6.9 to 11.1	0.15 to 0.30	24.7 to 27.4

Table 1: Total water uptake and pore volume of lapilli samples and stone matrix (n.d. = not determined)

Different types of lapilli can be found. Some are similar to the matrix with regard to their porosity and water uptake (samples lap 1 to 3), the others show significantly lower porosities (lap 4 to 6). Examples of pore size distributions for lapilli are given in Figure 5. The heterogeneous texture (lapilli vs. matrix) leads to preferential accumulation and crystallization of salts within the lapilli or at the border between lapilli and matrix as observed at the façade. Thus, the loss of lapilli inclusions is the first result of salt weathering in the zone > 1 m above the ground.

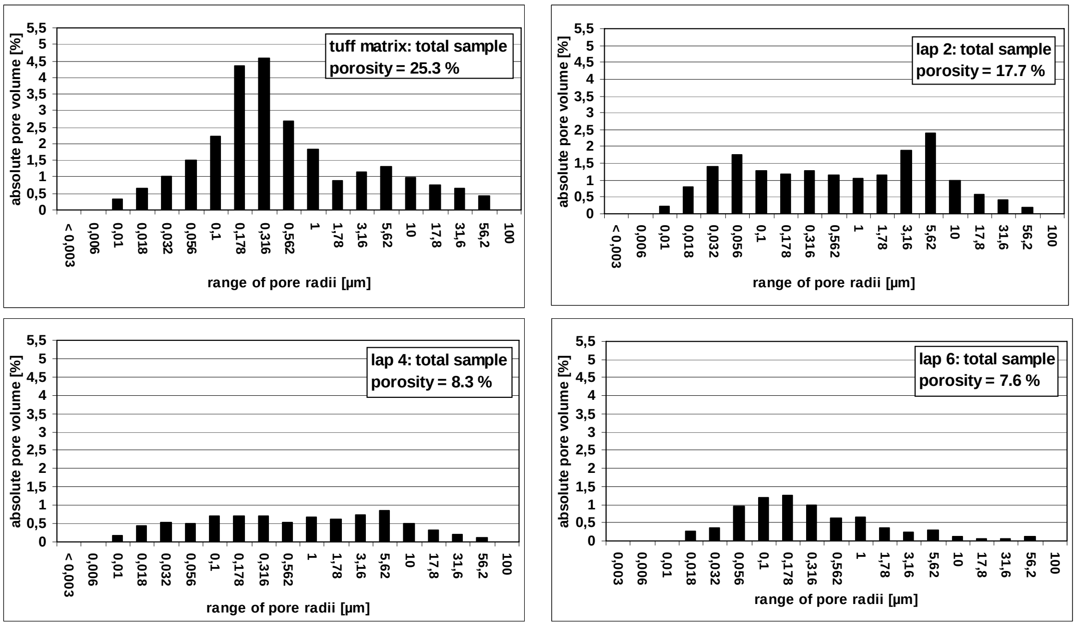

Figure 5: Pore size distribution in the matrix of rhyolite tuff and in lapilli inclusions

4.3 Investigations of soluble salts

The results of the analyses of soluble salts in drill cores are shown in Figure 5. Spatial distribution of soluble salts suggests that they have come from the ground with rising damp. In the zone close to the ground (h = 19 cm) only gypsum occurs near the surface. At a height of 80 cm, very high contents of gypsum are found with maximum values in a depth of 0.5 to 1 cm (sulphate 5.2 to 10.1 wt%, calcium 2.1 to 3.9 wt%). In this zone, contour scaling is the dominant weathering form (cf. fig 3 and Siedel 1998).

Sodium and potassium contents are below detection limit in both profiles. The salt load of the zone with incipient alveolar weathering (h = 120 cm) is characterized by a multi-ion system including remarkable contents of nitrate and chloride as well as magnesium sulphate beside gypsum.

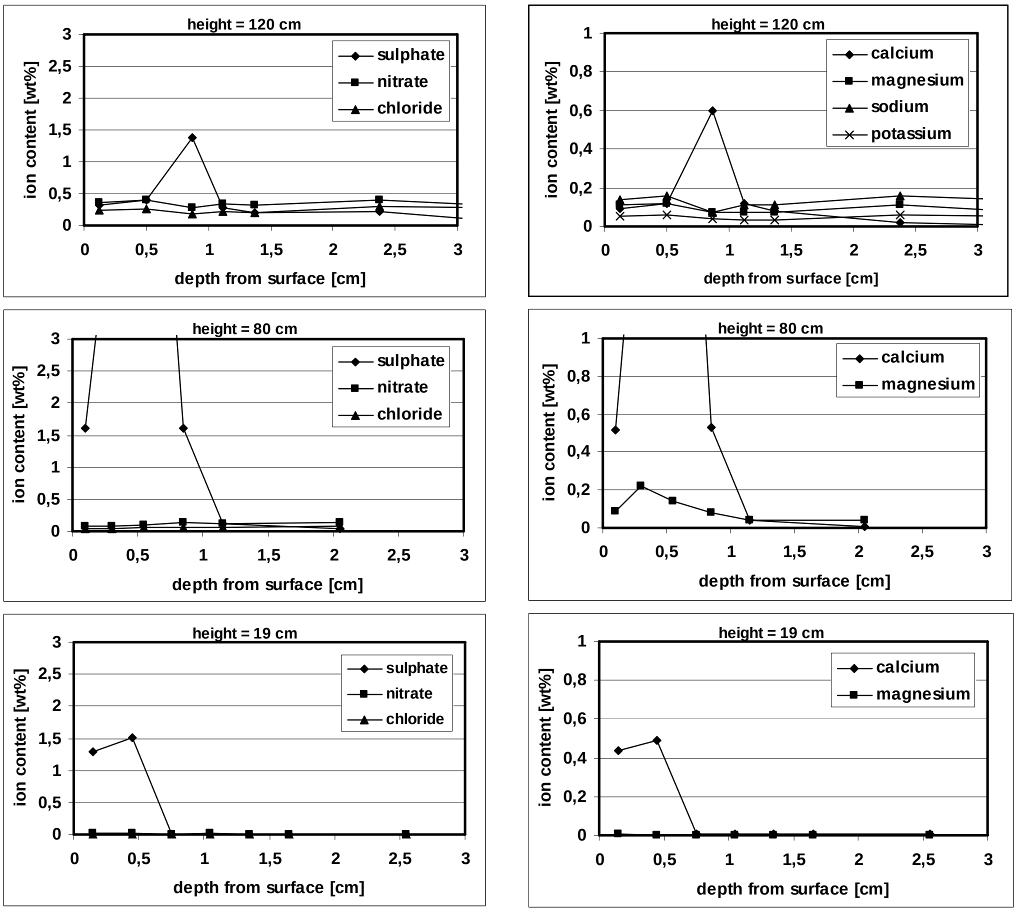

Figure 5: Results of salt analyses of drill cores from the church of St. Kunigunden taken at several height levels (cf. Figure 3).

Since the drill core at the height of 1.20 m was taken from a stable surface, more detailed salt investigations were carried out in an area with active alveolar weathering at the same level. A profile of drill powder samples was taken from the surface to depth on a ridge as well as at the bottom of the neighbouring hole. Results are given in Figure 6. Sulphate contents are very high near the surfaces of the ridge and of the hole as well. Nitrate and chloride, correlating with sodium and potassium, show the typical, more or less even distribution of these highly soluble salts over the whole profile depth in both ridge and hole. The spatial distribution of the cations calcium and magnesium correlates with the sulphate content, but is different for ridge and hole. Calcium (bound in gypsum, $CaSO_4 \cdot 2\ H_2O$) is concentrated near the non-weathered surface in the ridge whereas its content near the surface in the hole is much lower. In comparison, magnesium (bound in magnesium sulphate, $MgSO_4 \cdot n\ H_2O$) is highly concentrated near the surface in the hole and shows significantly lower contents near the non-weathered surface of the ridge. This is in accordance with the results of XRD analyses: efflorescing salts in holes are mainly magnesium sulphates, sometimes together with traces of gypsum.

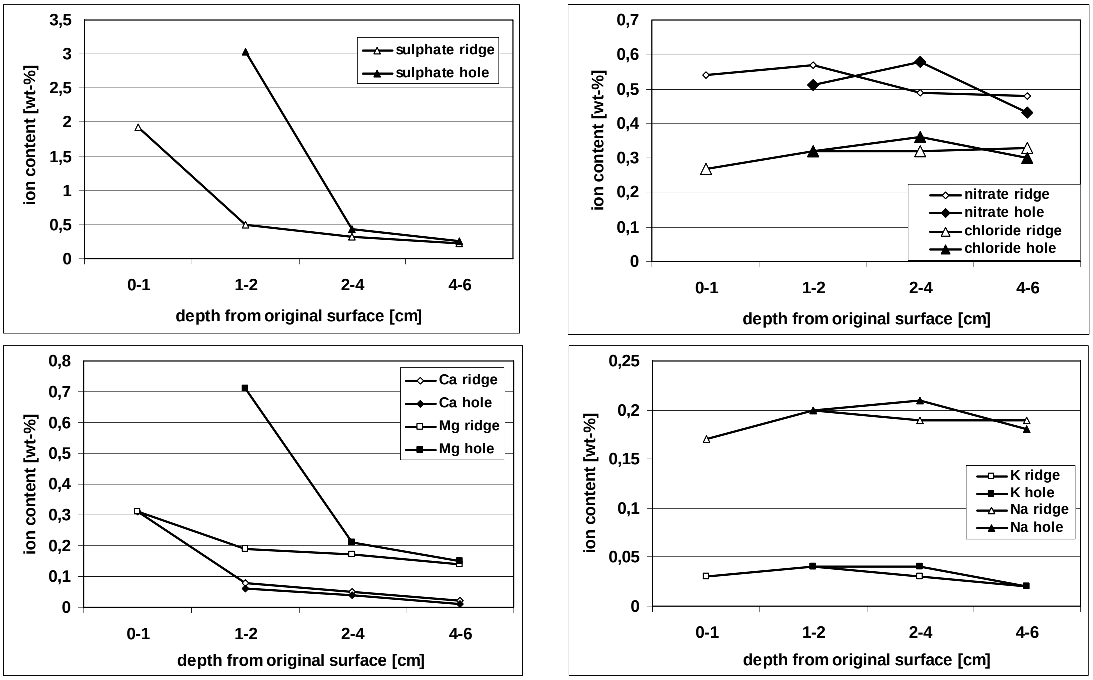

Figure 6: Salt distribution in a hole and in a ridge (cf. Figure 4, lower right).

The results of salt analyses demonstrate that alveolar weathering does not occur in the zone where only gypsum is the dominant salt compound even if the the highest salt concentration at all was found there. The presence of better soluble, mobile salts beside gypsum seems to be a precondition for this weathering process. In the profiles from the alveolized area as well as in efflorescence taken from the holes, magnesium sulphate is the dominant salt. This is in a good accordance with results obtained from investigations of alveolar weathering on sandstone buildings in Saxony [Siedel 2008]. The presence of magnesium sulphates is due to accelerated weathering of the dolomitic joint mortars under the environmental conditions of the last century with high pollution of air and rain water by sulphur [Klemm & Siedel 2002].

Initial pits caused by weathering out of single lapilli components from the matrix play a crucial role in the development of alveolar structures. Moisture and the most soluble salts will concentrate behind the pit when the stone dries slowly after a wet period [Huinink et al. 2004]. The original, non-weathered surface zone dries much faster, and less soluble salts (gypsum) are preferentially precipitated there. The salt distribution found in hole and ridge is the result of repeated moistening-drying events at the stone surface, leading to different spatial accumulation of magnesium sulphate and gypsum.

Once developed, the hole shelters magnesium sulphate from rainwash. Dependent on the climatic situation while sampling, the hydrated form found in the efflorescence was hexahydrite ($MgSO_4 \cdot 6H_2O$). Magnesium sulphates are among the most dangerous salts due to their changes in volume with the phase transition between hexahydrite and epsomite ($MgSO_4 \cdot 7\ H_2O$). According to Steiger [2000], the molar volume increases by nearly 10 % with this transition. Hydration and dehydration occur under the rapidly changing climatic conditions that are quite normal to building surfaces and cause expansion and shrinking of the stone in the affected zone [Juling *et* *al.* 2004]. These volume changes, induced by changes in relative humidity and/or temperature, also proceed in positions totally sheltered from direct rain attack. Since these processes are only dependent on the changing relative humidity of the air in the hole and not on contact with liquid water, they can work even in dry periods without rain events. Weathering is limited to the outermost zone at the bottom and on the walls of the hole with high salt load near the surface (cf. Figure 6), gradually moving deeper each time after loss of the surface material. Bigger holes can finally coalesce (cf. fig. 4, lower left). The different spatial distribution of salts in holes and ridges as well as the development of holes of greater dimension indicate that weathering dynamics becomes more and more independent of the primary, heterogeneous stone texture in the course of time.

5. Conclusions

The results of this study have shown that investigations on a façade can give useful hints towards the mechanism of alveolar weathering on a building stone. The different maturity of alveolization on different ashlars facilitates the description of distinct stages of the process. The weathering starts with the accumulation of magnesium sulphate in and behind lapilli components with a pore structure different from the tuff matrix. Material loss in these small areas leads to the formation of incipient holes. The preferential accumulation of magnesium sulphate in the shelter of the holes and its ability to change volume with climate changes force a further, permant loss of material on the surface of the holes. The deepening and widening of the holes become a self-perpetuating process at this stage, finally leading to coalescence of single holes and to the development of mature alveolar structures. The heterogeneous structure of the building stone as well as the high content of magnesium sulphate in the affected zone are preconditions for alveolar weathering in the case investigated in this study.

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