IMPLICATIONS OF USING METEOROLOGICAL RECORDS TO ASSESS THE ENVIRONMENTAL RISK OF SALT CRYSTALLIZATION CYCLES IN STONE

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KEYWORDS

Salt, Cycling, Environmental risk, Meteorological data, Stone, Conservation

ABSTRACT

Salt crystallization and dissolution cycles can significantly contribute to the degradation of stone, brick and mortar. One year of meteorological observations is used to evaluate the environmental risk using a threshold approach for a NaCl single salt. We illustrate the effect of boundary conditions such as the averaging timeframe and the minimum RH on the determined number of salt phase transitions, which are informed by droplet experiments. Additionally, the mitigating effect of crystallization within a porous substrate is explored using heat-air-moisture simulations. The results provide information on regional and seasonal differences in the environmental risk of salt weathering. Which in turn provide a better understanding of salt damage mechanisms over time to establish proper management strategies.

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1 INTRODUCTION

Salt crystallization and dissolution cycles can significantly contribute to the degradation of lithic materials as stone, brick and mortar. Degradation can manifest as physical and aesthetical material decay, the degree depending on the amount and type of salt mixture present in the porous substrate, the material properties of that substrate and the ambient environment [1]–[3]. The environmental risk of salt related decay can be understood as the intensity and magnitude of climatic changes, such as fluctuations in relative humidity (RH) that drive salt crystallization and dissolution.

Such environmental risk has already been assessed by parameterizing climate data [4] or analysing meteorological records [5], with the intention to estimate the number of potential salt phase transitions (crystallization-dissolution cycles). Several authors have used a threshold approach for single salts: the number of potential salt phase transitions is calculated by the number of RH crossings across the relative humidity equilibrium of a single salt (RH_{eq}), which is known for common single salts. In the case of NaCl, a RH_{eq} of approximately 75.5% RH is commonly accepted, although some authors prefer a critical relative humidity about 10% lower [6], which could be more realistic in the case of salt mixtures [7]. The RH_{eq} of $Na_2SO_4(.nH_2O)$ is more dependent on temperature, and hence its RH threshold is set as a function of temperature [6]. With the help of thermodynamic data or models such as ECOS-RUNSALT [8], [9], this threshold approach has also been performed for more complex salt mixtures [10], [11]. Typically, the timeframe in which RH fluctuations must occur are set as boundary condition. This strongly relates to the time resolution of the parameterized climate dataset or the resolution of meteorological observations. Different timeframes, from six- to twelve hours over 24h to 48h have been used. Experiments with NaCl in small stone samples have shown that, under specific conditions, significant amounts of salt can crystallize within the first 6h of drying experiments [12]. In the end, these outcomes are used as guidelines for climatic control in preventive conservation, or to compare the relative risk of different climates or climate change scenarios.

In this paper, we look at the influence of the spatial-temporal resolution of meteorological data on the environmental risk assessment with a threshold approach for the simplest case of NaCl. Therefore we use meteorological data from the urban environment in Gent and its rural surroundings [13] with a high temporal resolution. We evaluate how dependent the resulting number of salt phase transitions areon the chosen timeframe and on the dynamics of the ambient environment. This is compared with preliminary results from accompanying laboratory experiments [7] to improve estimations. Additionally, the meteorological data are used to produce model data of the hygrothermal conditions of Savonnières limestone with heat-air-moisture simulations in Delphin. These results are used to evaluate the buffering effect of the substrate in response to environmental drivers. Finally, information of seasonal and regional variability of salt phase transitions is retrieved.

2 MATERIALS AND METHODS

2.1 Meteorological data

The meteorological data are retrieved from a high-accuracy urban climate monitoring network (MOCCA) in Ghent (Belgium, Cfb climate) [13]. The data of two stations is used: (1) Melle, representing a rural environment in the surroundings of Ghent, is used as standard climate data, (2) Sint-Bavo school in the centre of Ghent to retrieve information on the effect of urban conditions on salt phase transitions. One full year (1 July 2016 – 30 June 2017) of data at high temporal resolution (1-minute interval) is used. This is rather exceptional and allows us to perform this study. Given the low dependency of the RH_{eq} of NaCl on temperature, only RH data was used in this study. RH was measured using a HC2S3 probe with an accuracy of 0.8% in a passively ventilated radiation shield.

2.2 Methodology and experimental data

The data were initially analyzed for NaCl as a single salt, for which 75.5% RH was taken as the RH_{eq}. The number of salt phase transitions was counted as negative crossings (from RH ≥ 75.5% to RH < 75.5%) using the average RH in different averaging timeframes (1 min, 5 min, 10 min, 30 min, 60 min, 720 min (12 h), 1440 min (24 h)). The number of salt phase transitions was considered equivalent as the number of salt crystallization-dissolution cycles. A cycle was only accounted for when no other cycle was recorded within the averaging timeframe before the moment it was identified (e.g. for 1-minute timeframes a cycle can be registered every next minute, whilst for the 24 h-timeframe a cycle can only be registered in the succeeding 24 h period). Other $RH_{eq,}$ for examples in salt mixtures, are not considered in this paper.

Additionally, preliminary experimental results on the crystallization and dissolution kinetics of NaCl droplets were used to inform the environmental risk assessment. This set of experiments was performed by recording the solution/crystal in a controlled climatic chamber (GenRH), using RH cycles from 90% down to respectively 70%, 60%, 50%, 40%, 30%, 20%, and intermittently back to 90%. These data show a high dependency of the crystallization and less of the dissolution on the magnitude of the RH cycle [7]. Here, we conceptualize the results to include in the meteorological data analysis. A timeframe of 30 minutes is kept constant for the dissolution stage; whilst we set forward a crystallization time of 30 minutes for RH cycles that went down to 70%; 10 min for cycles that went down to 60% RH and 5 minutes for cycles where the minimum RH reached levels below 40%.

2.3 Hygrothermal model data

Heat, air and moisture (HAM-) simulations were performed using Delphin 6.1 (Bauklimatik Dresden) to simulate hygrothermal conditions in Savonnières limestone using in-house defined properties and the MOCCA data used for the meteorological analysis. A 1D construction (500 x 500 mm) with planar horizontal transport in the x-direction was assumed. The inside climate was assumed to be constant with a temperature of 20 °C and relative humidity of 50%. Simulations were run for 6 years (repeating the same yearly record), 5 years for preconditioning of the stone and the final year for studying hygrothermal conditions at the stone surface, at 1.1 cm and 2.0 cm depth. From this, the RH in the (pores of) the stone was retrieved for analysis. The data were reported with a time resolution of 5 minutes.

3 RESULTS AND DISCUSSION

3.1 Environmental risk using a 75.5% RH threshold

The meteorological data of Melle (rural) was tested on negative crossings of 75.5% RH, based on the chosen RH_{eq} of NaCl, for different averaging timeframes. The result is shown in Figure 1 for the entire year of observations (annual total) and illustratively for the periods July-September (summer) and December-February (winter). The annual number of potential salt phase transitions is rather high and strongly depending on the averaging timeframe. 1823 negative threshold crossings occur on the high-resolution data (1 minute) throughout the entire year. This number is irrelevantly high, as we assume salt phase transition kinetics to be much slower.

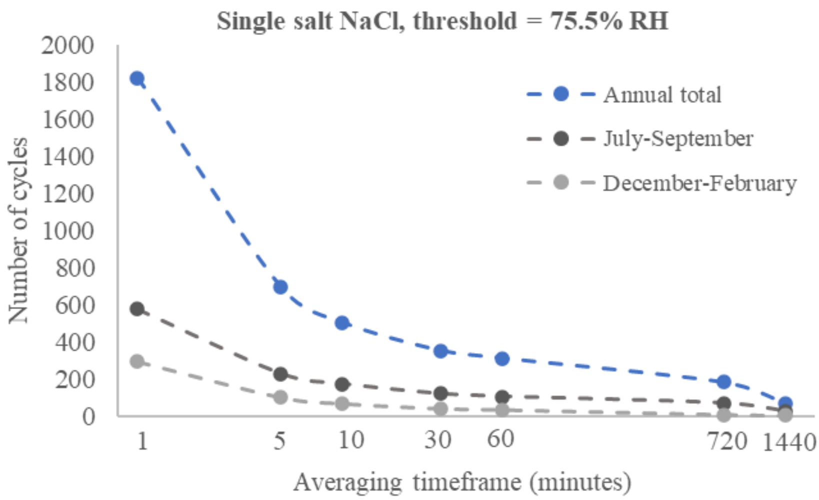

Figure 1: Total number of negative 75.5% RH crossings for the meteorological observations in Melle (rural) when considering RH averages over different time windows (1, 5, 10, 30, 60, 720 and 1440 min) in the period July 2016 – June 2017. The total number over the entire year is given in blue. The total number over three months in summer (July-September) and winter (December-February) are given in gray.

However, when looking at the data of 5 minutes and 10 minutes, the number of estimated events drops to 698 and 507 crossings respectively. This illustrates that many of the crossings are short term fluctuations around 75.5% RH during a more general RH rise. More interesting for considering salt phase transitions is the number of potential salt phase transitions when averaging the RH over a timeframe of 60 minutes, with 315 occurrences per year, and frames of 12 h and 24 h with 185 and 73 cycles, respectively. This means that there are 73 occasions where the average RH was below 75.5% for 24 subsequent hours during which no previous cycle was registered, followed by a period of 24 subsequent hours where the average is above 75.5%. This number is remarkably high. Though, being less than half the number of cycles compared to a 12 h timeframe, it shows the high dependency of the environmental risk assessment on the choice for the length of the timeframe.

3.2 Minimum RH as additional boundary condition

Preliminary experiments with fluctuating RH cycles on NaCl droplets give information on the speed of dissolution and crystallization. This is an oversimplification in the context of stone weathering, as kinetics in capillaries are generally slower. Nevertheless, this straightforward approach is used to incorporate the magnitude of RH jumps in the threshold approach as an additional constraint to the averaging timeframe. The results of the droplet experiment [7] were used to inform the analysis of the climate date, which in turn should inform the design of more realistic experiments in the future.

Based on the droplet experiments, the simplified (and conceptual) assumption is made that dissolution always takes 30 minutes, and that crystallization takes 60 minutes, unless the minimum RH during the crystallization phase was ≤ 70%, for which the crystallization time was set at 30 minutes; ≤ 60% for which the crystallization time was set at 10 minutes; and ≤ 40% for which the crystallization time was set at 5 minutes. In other words, the number of threshold crossings was now calculated with the average timeframe of dissolution always set at 30 minutes; whilst a minimum RH condition was applied to the average timeframe of 5, 10, 30 and 60 minutes.

In this case, we get a more realistic response from the analysis, shown in Table 1. For Melle, a total number of 619 cycles complying to the condition of 60 min of crystallization time occur throughout the year. Only 193 comply to a timeframe of 30 minutes, 5 to a timeframe of 10 minutes and 1 to a timeframe of 5 minutes. A cycle where the RH rises from 60% to above 75.5% within 10 minutes (and is counted in this timeframe), will also comply (and hence be counted) to the timeframe of 1 h. In theory, it is possible that additional cycles are identified at this high time resolution, but in practice this is not the case as such high fluctuations follow a single upward trend (i.e. multiple extreme oscillations do not occur within 1h). More interesting is that, although the RH regularly drops to low values, the drop below 75.5% RH is almost never fast enough and sufficient in magnitude to facilitate cycles at the time resolution of 5 minutes.

This data informs us about interpreting meteorological data at high temporal resolution. From the 507 negative threshold crossings observed throughout the year with a timeframe of 10 min, five potentially correspond to a salt phase transition if we rely on the (simplified) kinetic data of the droplet experiments. Of the 356 annual crossings with a 30-minute timeframe, 193 could correspond to a potential phase transition in the same reasoning.

	5 min. (RH ≤ 40%)	10 min. (RH ≤ 60%)	30 min. (RH ≤ 70%)	60 min (RH ≤ 75.5%)
Melle (rural)	1	5	193	619
St Bavo (urban)	0	0	150	649

Table 1: Number of negative 75.5% RH crossings for averaging timeframes of 5, 10, 30 and 60 minutes, applying a minimal RH within that timeframe as additional boundary condition for counting a salt phase transition. Based on the meteorological records from Melle (rural) and St Bavo school (urban) in the period July 2016 – June 2017.

3.3 Hygrothermal data of substrates

The conditions in porous media differ from the unconfined conditions of droplet experiments. Experiment in capillaries of different size and with solution/crystals at different depths can inform us about the differences in kinetics [7]. Additionally, the surface conditions on and in stone substrates might differ from bulk atmospheric conditions generally measured in meteorological records. Therefore, we adopt an alternative approach from the perspective of the climate data. Through HAM-simulations, a record of RH at different depths within the porous substrate is created in function of the meteorological observations. This record is tested in the same way as the meteorological data. Because of the computing time necessary for generating the record, we focus on analyzing the record for 1 representative month (July 2016) instead of the entire year (after 5 years of preconditioning to create a realistic record).

Table 2 shows the potential number of salt phase transitions (similar to Table 1) for 1 month based on meteorological and hygrothermal RH records. As expected, changes in RH are mitigated deeper within the stone, where equilibration with the ambient conditions is depending on capillary action and vapour diffusion. There is a small decrease in potential cycles at the surface of the stone compared to the meteorological conditions. However, the short-term cycles do occur near the surface. The number of potential salt phase transitions is further decreasing in depth where no short-term cycles occur and the overall number is reduced by more than a tenfold at 2 cm depth.

	5 min. (RH ≤ 40%)	10 min. (RH ≤ 60%)	30 min. (RH ≤ 70%)	60 min (RH ≤ 75.5%)
Meteo.data	0	1	26	73
Surface	0	1	21	67
1.1 cm depth	0	0	1	22
2 cm depth	0	0	0	6

Table 2: Number of negative 75.5% RH crossings for averaging timeframes of 5, 10, 30 and 60 minutes, applying a minimal RH within that timeframe as additional boundary condition for counting a salt phase transition. Meteo.data represents the outcome of direct observations from meteorological data. Surface, 1 cm depth and 2 cm depth are based on the hygrothermal RH conditions on and within a Savonnières limestone, based on hygrothermal modelling using the meteorological observations for Melle. Data for July 2016.

3.4 Seasonal and regional variations in environmental risk

Some interesting interpretations can be drawn from comparing the RH fluctuations at different locations (urban vs. rural) and during different seasons. Of course, these only apply to the hypothetical case of NaCl as single salt, and not in mixtures where the threshold can be different. Figure 2 shows boxplots of the monthly variation in RH (July 2016 – June 2017) for an urban and a rural location.

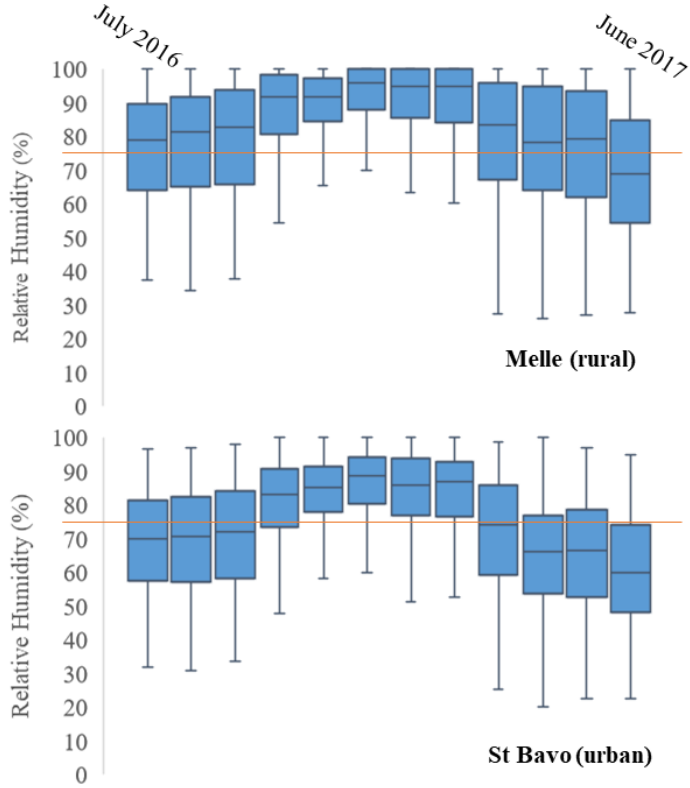

Figure 2: Boxplot showing the RH variations in Melle (rural) and St Bavo school (urban) for 12 consecutive months in the period July 2016 (left) to June 2017 (right). The orange line depicts the 75.5% RH threshold. It can be observed that the RH fluctuates more around the threshold in spring and summer, implying a higher NaCl single salt environmental risk during this period, compared to winter and spring.

First, it can be seen that monthly fluctuations in RH are much closer to the RH threshold of 75.5% during spring and summer. During fall and winter, the RH in general is much higher, resulting in less negative crossings of the 75.5% threshold. This can also be observed in Figure 1, where the total number of negative thresholds crossings is higher for the three-month periods July-September and lower for December-February. Hence, for the Cfb climate in Ghent, the outdoor environmental risk for NaCl-related weathering is higher in spring and summer, and less in winter.

It can also be observed that the RH is in general lower throughout the year in an urban environment compared to a rural environment, and the intensity of its fluctuations is also mitigated in an urban environment. As a result of the latter, the conditions for short term (< 1h) salt phase transitions as observed in the droplet experiments are less relevant in the urban environment.

4 CONCLUSIONS

A meteorological record with high time resolution can provide an accurate estimation of on the number of times the RH_{eq} is surpassed. Compared to records with low time resolution, averaging timeframes in a high-resolution dataset can yield a more correct number of cycles. Information on the kinetics of crystallization and dissolution can improve the evaluation of the environmental risk. Using short averaging timeframes yields an irrelevant high number; but in combination with RH constraints and simple experimental observations, this number is drastically reduced. Within the limits of this study, it can be proposed that few short-term cycles (<1 h) can occur, although these are also counted when using timeframes of 1 h. The outlook of more detailed experimental data will probably put more restrictions on the conditions and therefore further reduce the number of cycles. HAM-simulations have the potential to further improve environmental risk assessment. However, the approach for analyzing the data still needs to be validated and would benefit of experimental research on confined crystallization and dissolution in capillaries.

Analyzing the meteorological data shows that the risk of NaCl weathering in outdoor environments is highest in spring and summer for the meteorological record under observation. Significant differences also occur between rural and urban environment, but the impact is dependent on the chosen RH_{eq} and on the season under investigation. The results of such analysis can be used as guidelines for climatic control in preventive conservation, or to compare the relative risk of different climates or climate change scenarios.

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