

# Predicting environmental conditions to minimise salt damage at the Tower of London: a comparison of two approaches

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**Abstract** The paper compares the results of two studies of salt damage at the Wakefield Tower, Tower of London. The first, in 1993, was based on semi-quantitative salt analysis and hygroscopicity measurements. The second was based on quantitative analysis and a thermodynamic model, ECOS. Both studies aimed to predict environmental conditions that would minimise salt damage to the interior stonework. Their predictions are markedly different, and this paper examines the reasons for the differences. It is shown that hygroscopicity alone is not a reliable indicator of crystallisation from mixed salt solutions.

**Keywords** Stone · Conservation · Salt · Environmental control · Hygroscopicity

## Introduction

A paper was published in 1993 that made recommendations regarding the environmental conditions that would minimise salt damage in the Wakefield Tower, HM Tower of London (Price 1993). The recommendations were based on semi-quantitative analyses of the salts present in the masonry, and on the hygroscopic behaviour of mortar samples taken from the joints. The paper highlighted the limitations of this approach, and stressed the difficulty of predicting the behaviour of salt mixtures.

In the years that followed, considerable advances were made in modelling the crystallisation behaviour of salt mixtures and thereby predicting more reliably the environmental conditions that would minimise the damaging effects of salts (Price and Brimblecombe 1994, Steiger and Dannecker 1995, Price 2000). This approach requires reliable analytical data from traditional chemical techniques or from more recent techniques such as ion chromatography (IC) and inductively-coupled plasma atomic emission spectroscopy (ICP-AES). The 1993 approach, by contrast, required only very basic laboratory facilities, but was relatively labour intensive.

The purpose of the present paper is to make a critical comparison of the predictions of the two approaches. It does not set out to offer definitive recommendations for optimal environmental conditions at the Tower.

## The Wakefield Tower

The Wakefield Tower was built in the early thirteenth century. It consists of two large rooms, or chambers, one above the other. The external diameter is more than 15 m. Around 1970, the lower chamber was excavated down to its original floor level. The masonry that was thus revealed had been protected by infill since the late thirteenth century, and was then in excellent condition (Parnell 1993). It has since started to deteriorate. The stone in the interior of the upper chamber also shows serious decay in some parts. Further details are given by Brown and Curnow (1984) and by Price (1993).

The Wakefield Tower is built mainly of Kentish Ragstone (a dense, sandy glauconitic limestone from

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the Hythe beds of the Cretaceous Lower Greensand formation), with dressings of Reigate stone (a calcareous sandstone, from the Cretaceous Upper Greensand formation). Reigate stone, which has a porosity in the region of 30–40%, is well known for its poor resistance to weathering, and it has been the subject of a special study undertaken by Historic Royal Palaces (Sanderson and Garner 2001). It is the Reigate stone that gives the greatest cause for concern in the upper chamber; some areas are deteriorating much faster than others, and there is no obvious reason for the observed differences.

### The 1993 investigation

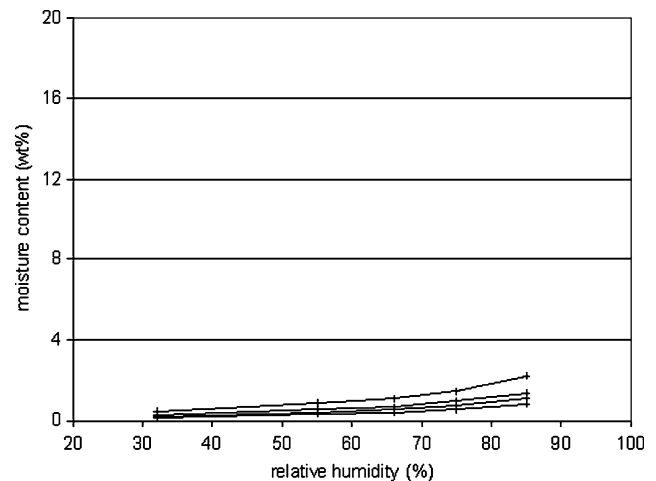
Price (1993) used Merckoquant test strips to determine the nitrate and sulphate content of aqueous extracts of stone and mortar samples, and an Aquamerck test kit for the chlorides. Forty three samples were taken, mostly by drilling with a 10 mm masonry drill. Successive drillings were taken at different depths, in order to determine the variation of salt content with depth. Full experimental details, together with sample locations, are given in the 1993 paper.

Price reported significant differences between the upper and lower chambers:

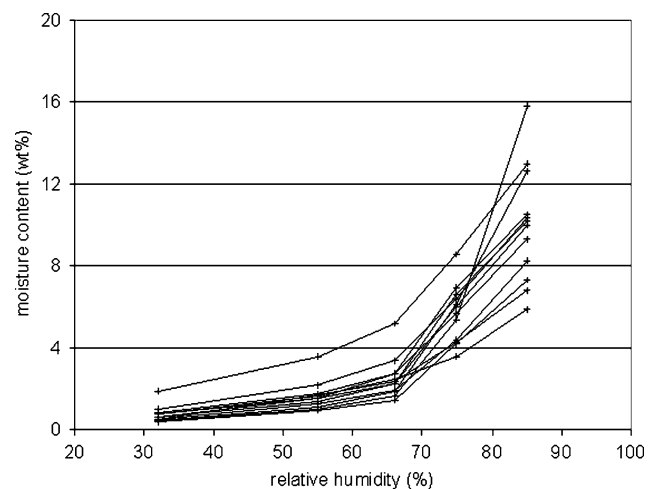
- the salt content of samples taken from the upper chamber was generally two to three times higher than that of samples from the lower chamber;
- the higher salt content of the upper chamber was largely attributable to chlorides and nitrates; and
- no sulphates were detectable in the upper chamber, whereas sulphate levels were high in the lower chamber.

These differences were reflected in the hygroscopic behaviour of the mortar samples, which are shown in Figs. 1 and 2. The samples were exposed, until they reached constant weight, at relative humidities of 32, 55, 66, 75 and 85%, achieved by the use of saturated salt solutions in sealed containers. Whilst the data do not rule out the possibility that the hygroscopicity of the samples from the lower chamber is due to the substrate alone, the influence of salts on the hygroscopicity of the samples from the upper chamber is clearly to be seen.

On the basis of these results, Price recommended that the relative humidity (RH) in the upper chamber should be kept below 66% at all times, and that the relative humidity in the lower chamber should be kept above 75%. The intention was to keep the humidity in the upper chamber at a low enough level to keep the



**Fig. 1** Hygroscopic behaviour of mortar drillings from the lower chamber



**Fig. 2** Hygroscopic behaviour of mortar drillings from the upper chamber

salts in the solid state at all times. He argued that “the moisture content of almost all the samples from the upper chamber showed a marked increase between 66 and 77% RH. It is likely that much of the absorption takes place at the top end of this range, as the equilibrium RH of sodium chloride is 75% and for sodium nitrate it is 76%.” It was on this evidence, discussed further below, that he recommended that the RH should not exceed 66%. By contrast, the intention was to keep the salts in the lower chamber in solution at all times. This was because the masonry of the lower chamber is in contact with groundwater, and dry conditions in the lower chamber would simply serve to draw up more moisture and salts from the soil, causing continuous crystallisation damage. The choice of 75% RH as a lower limit was a compromise between a

preferred value of 85% and realism, practicability, and the danger of biological growth.

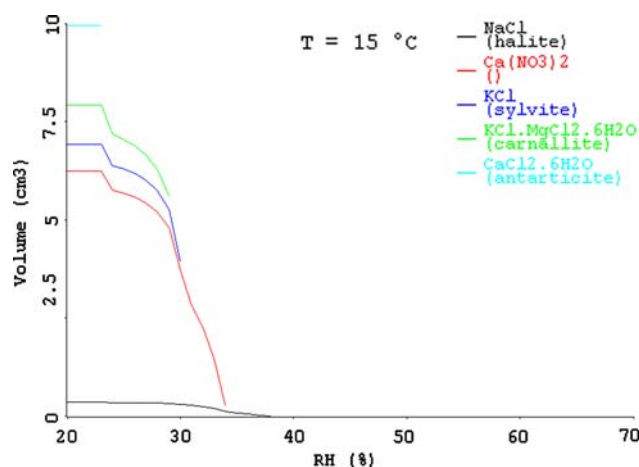
In the event, these recommendations were never implemented, due in part to the untimely death of Martin Caroe, the architect who commissioned the study.

### Predictions based on thermodynamic modelling

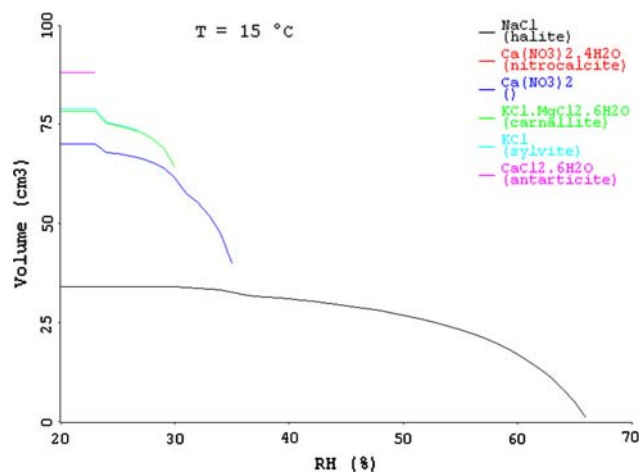
Collaborative work between Steiger, Brimblecombe, Clegg and Price, funded by the European Commission, led to the development of a computer program ECOS (Environmental Control Of Salts) that is capable of predicting the crystallisation behaviour of salt mixtures (Price 2000). The user is required to input the concentrations of a range of ions that are present in an aqueous extract of the salt-contaminated material in question. The program is then able to predict which minerals will exist in the solid state under specified conditions of relative humidity and temperature. This enables the user to determine ‘safe’ ranges of relative humidity and temperature, in which phase changes and consequent volume changes are kept to a minimum. Bionda subsequently developed an improved user-interface, whilst retaining the fundamental model and data of the ECOS program. His program, which can be downloaded from <http://www.ftk.id.arch.ethz.ch/software.php?lang=de>, is known as RUNSALT, and it has been used to produce the charts shown below.

Church (2003) carried out a further study of the deterioration of the Reigate stone in the Wakefield Tower. As part of her study, she undertook quantitative analysis (using IC and ICP-AES) of the same samples that had been taken in the 1993 study. Her analyses were entered into the RUNSALT program, using the autobalance facility to compensate for charge imbalance. The resulting charts fell into two very clear groups, representing the upper and lower chambers of the Tower. Typical examples are shown in Figs. 3 and 4, which show the relative volumes of the minerals that are in thermodynamic equilibrium at 15°C across a range of relative humidities<sup>1</sup>. The corresponding analytical data (in wt % of dry sample, and in  $\mu\text{eq/g}$ ) are given in Table 1, which also gives the data from the 1993 investigation.

<sup>1</sup> Steiger has queried the detailed ECOS predictions, noting that anhydrous calcium nitrate is stable only below about 10% RH, and that ECOS seemingly omits the double salt  $\text{KNO}_3 \cdot \text{Ca}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ . Nonetheless, he does not consider that the main conclusions are affected (Steiger, Institut für Anorganische und Angewandte Chemie, Universität Hamburg, personal communication, 2006).



**Fig. 3** Typical sample from the lower chamber (location B, 40–80 mm). The crystallisation sequence of the soluble salts as a function of RH is calculated using RUNSALT’s autobalance facility



**Fig. 4** Typical sample from the upper chamber (location F, 20–40 mm). The crystallisation sequence of the soluble salts as a function of RH is calculated using RUNSALT’s autobalance facility

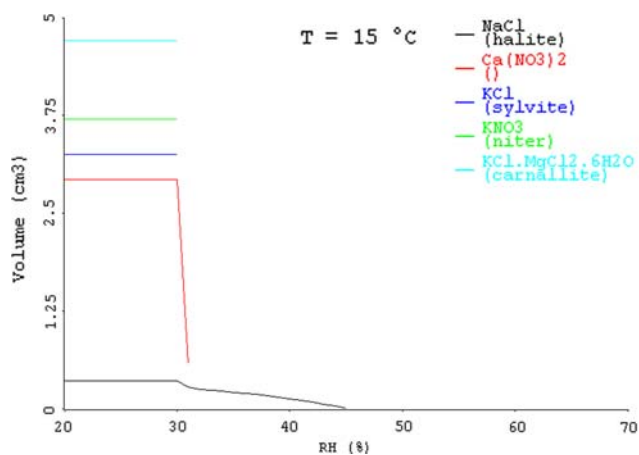
It should be borne in mind that the ECOS autobalance facility may be misrepresenting the true conditions: in each case, the cation charge exceeds the anion charge, suggesting the presence of carbonates or hydrogen carbonates which are not covered by the ECOS program. The program was therefore run again, balancing the charges solely by removal of calcium. This is equivalent, in practice, to regarding the deposition of calcite from the evaporation of a calcium hydrogen carbonate solution as a separate process from the crystallisation of other minerals from the solution. The resulting charts, shown in Figs. 5 and 6, are comfortingly and perhaps surprisingly similar in broad outline to those of Figs. 3 and 4.

**Table 1** Ionic analyses (wt %) of samples plotted in Figs. 3, 4, 5, 6. (The numbers in brackets are expressed in  $\mu\text{eq/g}$ )

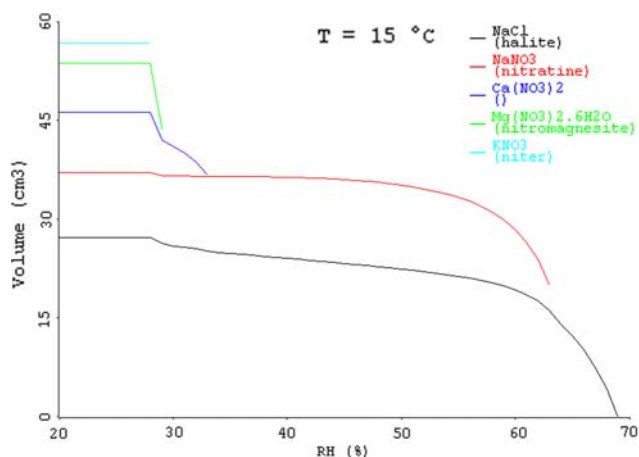
Sample location	Ca <sup>2+</sup>	Na <sup>+</sup>	Mg <sup>2+</sup>	K <sup>+</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>
B, lower chamber (Church)	0.42 (210)	0.02 (6.9)	0.01 (5.9)	0.05 (12)	0.07 (20)	0.28 (45)	0.49 (101)
B, lower chamber (Price)	NM	NM	NM	NM	0.4	<0.1	0.4–0.6
F, upper chamber (Church)	1.04 (517)	1.17 (510)	0.05 (38)	0.10 (25)	1.44 (405)	1.73 (280)	0.11 (23)
F, upper chamber (Price)	NM	NM	NM	NM	1.4	1.0	<0.1

NM not measured

The implications of Figs. 3 and 5 for the lower chamber are clear: that little or no salt will crystallise if the ambient relative humidity remains above 40–45%,



**Fig. 5** Typical sample from the lower chamber (location B, 40–80 mm). The crystallisation sequence of the soluble salts is calculated using RUNSALT, having first balanced the charges by removal of calcium



**Fig. 6** Typical sample from the upper chamber (location F, 20–40 mm). The crystallisation sequence of the soluble salts is calculated using RUNSALT, having first balanced the charges by removal of calcium

and crystallisation only starts in earnest when the RH falls as low as 30%. This differs markedly from Price's previous recommendation, that the RH should fall no lower than 75%. The discrepancy is due in part to the removal by ECOS of calcium sulphate, leaving no remaining sulphate; and in part to the very marked suppression of the relative humidity at which sodium chloride crystallises (ca. 40%, as opposed to 75% for the pure salt).

The removal of sulphate by the ECOS program is necessitated by the model's inability to handle calcium and sulphate simultaneously. This limitation is overcome by the program's removal of gypsum from the system, on the assumption that gypsum's solubility is so low that it will not contribute significantly to crystallisation damage (Price 2000). Whilst this might appear a weakness on first sight, it is illuminating to consider that this may indeed be what happens in practice. If so, Price's (1993) concern about the crystallisation of sodium and magnesium sulphates at high relative humidities was unfounded.

If the ECOS prediction is correct, then there should be little risk of crystallisation damage in the lower chamber at any time, provided that no heating is installed. The ambient relative humidity in London is usually well above 40%, and it rarely falls below 30%. In 2005, for example, the relative humidity fell below 40% on 43 days (all between April and September, except for one in February), but fell below 30% on only 7 days (<http://www.wunderground.com>)<sup>2</sup>. There is little data relating to the Tower itself, for Historic Royal Palaces has only recently started a programme of continuous monitoring. However, data for a 45-day period in April and May 2006 show twelve occasions on which the humidity in the lower chamber fell below 40%, a frequency that is very comparable to the 2005 data, above (Hallett, Historic Royal Palaces, personal communication, 2006). This raises the question of whether salt crystallisation is indeed the primary cause of the deterioration that has

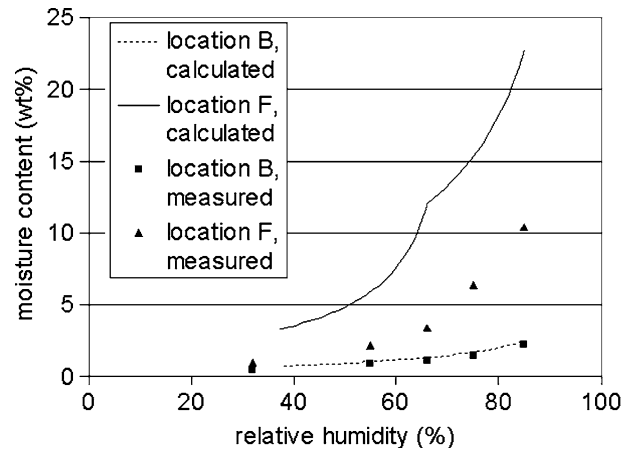
<sup>2</sup> data from London City Airport, 9 km from the Tower and, like the Tower, beside the River Thames.

taken place in the stonework of the lower chamber since it was exposed by excavation around 1970, and it suggests that salt damage might be minimised by the modest use of humidifiers during the summer months. It would be much easier to keep the humidity above 40–45% than to keep it above the previously recommended minimum of 75%.

Whilst the previous recommendations for the lower chamber do not appear, with hindsight, to have entailed any risk of damage to the stonework, the same cannot be said for the upper chamber. Figures 4 and 6 suggest that no damage can occur above 70% RH, for all salts are then in solution, and that there is a plateau of relative safety between about 35 and 50%, where the total volume of salts is relatively steady. The region between 50 and 70% RH is definitely to be avoided, since much crystallisation occurs within that range. This is in contrast to the earlier recommendation that the relative humidity in the upper chamber should be kept below 66% at all times, and it is a regime that would be extremely difficult to achieve in practice. Monitoring in the upper chamber, which is heated, has shown that the relative humidity falls in this region much of the time (Hallett, Historic Royal Palaces, personal communication, 2006).

With hindsight, it can be seen that the use of hygroscopicity measurements to predict safe ranges was unsatisfactory and that their interpretation was incorrect. Too much attention was paid to the equilibrium RHs of individual salts and to the RH ranges at which the samples were picking up large quantities of moisture. It was not appreciated that crystallisation from a salt mixture takes place over a range of relative humidities, and that individual minerals may crystallise out at relative humidities far lower than their equilibrium RH as a single salt, as was demonstrated empirically by Arnold and Zehnder (1991). Both these factors would serve to lower the supposedly safe RH ranges that were predicted on the basis of hygroscopicity measurements and the equilibrium RHs of individual salts.

Figure 7 shows the predicted hygroscopicity behaviour of the samples charted in Figs. 3, 4, 5, 6. The calculations were performed by Steiger, using the Pitzer model on which ECOS is based. There is very strong correlation between predicted and observed behaviour for one of the samples, suggesting that the hygroscopicity of the substrate itself plays only a minor role; the figures for the other sample differ by a factor of about two, and this has yet to be accounted for. The discontinuity at 66% RH in the predicted behaviour for the sample from the upper chamber corresponds with the onset of crystallisation (cf.



**Fig. 7** Comparison of observed and predicted hygroscopicities of samples charted in Figs. 3, 4, 5, 6

Figs. 4, 6), and the absence of any discontinuity above 40% RH for the sample from the lower chamber is consistent with the prediction that all salts will be in solution above that RH. But the most important observation in this context is that the discontinuity at 66% RH is not so marked that it could reasonably have been picked up by practical measurement of hygroscopicity, using saturated salt solutions which give stepped changes in relative humidity. Hygroscopicity measurement cannot therefore be regarded as a practicable predictor of crystallisation from mixed salt solutions.

## Conclusions

Price in 1993 was aware of the limitations of his approach, and foresaw the need for a model that would provide more secure predictions of conditions needed to minimise salt damage in the Wakefield Tower. Such a model, now that it is available, confirms that hygroscopicity measurement alone cannot provide an adequate indicator of crystallisation from mixed salt solutions, and that previous recommendations for environmental control at the Tower were unfounded.

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