1997

# The Short-Term Effects of Mortars on Salt Movement in Stone

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## Recommended Citation

Perry, S. H. and Duffy, A. P. (1997) 'The short-term effects of mortars on salt movement in stone' in *Atmospheric Environment*, Pergamon,Vol. 31, No. 9, pp. 1297-1305. doi:10.1016/S1352-2310(96)00290-7

Pergamon

*Atmospheric Environment* Vol. 31, No. 9, pp. 1297–1305, 1997

1352–2310/97 $17.00 + 0.00

PII: S1352–2310(96)00290–7

# THE SHORT-TERM EFFECTS OF MORTAR JOINTS ON SALT MOVEMENT IN STONE

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(*First received 3 January 1995 and in final form 9 September 1996*)

**Abstract**—Salt-induced decay processes are one of the most important reasons for the decay of many stone buildings and monuments. This paper investigates the role played by masonry mortars in the movement of salts within Baumberger sandstone and Portland limestone. The methodology used involved the monitoring of stone units containing mortar joints which were exposed to a city-centre environment. Rain-water run-off was collected from the stone/mortar units during the exposure period; the units were subsequently destructively analysed. The methods of run-off and destructive sample analysis used included ion chromatography and atomic absorption spectrophotometry; stone and mortar samples were also analysed petrographically. Mortars were found to act as sinks of environmentally derived ions and as sources of mortar-derived ions in well washed surface environments. Mortar enrichment by sulphate ($SO_4^{2-}$), sodium ($Na^{2+}$) and chloride ($Cl^-$) ions, which may lead to salt damage of stone around joints, was observed. High levels of calcium ($Ca^{2+}$) ions, in the form of reprecipitated calcite, were recorded in the stone, leading to a reduction in the surface permeability of the stone, a common cause of delamination. © 1997 Elsevier Science Ltd. All rights reserved.

*Key word index*: Stone decay, stone durability, stone weathering, mortars, soluble salts, microcatchment units, Portland limestone, Baumberger sandstone.

## 1. INTRODUCTION

The term "building stone decay" refers to the breakdown of surface stone due to the action of external agents coming from the environment. The action of natural weathering parameters such as rain, wind and temperature, pollution is widely believed to accelerate stone decay processes. In particular, high air- and precipitation-borne sulphate levels in urban environments have led to extensive stone decay. The decay resulting from environmental agents manifests itself visually in many ways: granulation, flaking, scaling, rupture and delamination are examples and there is a similar diversity of processes which lead to these forms of decay. Stone minerals which are soluble in water dissolve in areas where water runs over the building's surface. The growth of biological species may also result in stone dissolution due to the secretion of acids. Moreover, physical processes, such as those resulting from the action of penetrating hyphae, may lead to stone decay. Mineral alteration occurs when chemical species, such as the hydrogen ion, react with the stone's minerals to form materials which are less durable, thus resulting in the decay of the stone. In some cases, stone minerals which have absorbed moisture exert stresses on the stone matrix, because of the associated volume increase, also resulting in decay. Similarly, the freezing of water in a stone exerts internal stresses and may eventually lead to the fracturing, spalling or rupture of the stone. Finally, salt-induced decay is caused when salts exert internal pressures which exceed the stone's strength.

These stone decay processes are dependant both on the material properties of the stone and the surface environment to which it is exposed. The surface environment describes the temperature, water chemistry, dry chemistry and quantity of water at the stone's surface. For example, a stone such as Portland limestone will dissolve more rapidly than one such as granite, due to the higher solubilities of the minerals comprising the former. However, the amount of dissolution is also dependent on the quantity and chemistry of water flowing over the stone. The chemistry of the aqueous surface environment is determined by the type of chemical species deposited from the general environment, such as gaseous or particulate sulphate, and by chemicals released from the material as a result of decay processes such as those described above. For example, in the case of Portland limestone dissolution, $Ca^{2+}$ and $CO_3^{2-}$ ions which are released into the surface environment may precipitate to form salts.

The above account of the causes of stone decay reflects the common emphasis on the stone as the primary source of materially-derived chemical species. However, mortars also play an important, if neglected, role in the types, levels and location of soluble salts in building stone and, hence, in building stone decay.

## 2. SALT-INDUCED STONE DECAY PROCESSES

Salt-induced stone decay processes are caused when salts exert internal pressures on stone through crystallization, hydration and thermal expansion. When these stresses exceed the compressive strength of the stone, disruption and decay occur (Lewin, 1989).

Salts crystallize out of either under-saturated or super-saturated salt solutions which may be present in the pore spaces of a stone. Precipitation of salts from under-saturated solutions, as in the case of the evaporation of the solvent, does not result in stresses being exerted on the stone. It can, however, result in pore blockage and a reduction in stone permeability, leading to water entrapment and accelerated decay due to internal stone dissolution. However, the precipitation of salts from super-saturated solutions, caused either by a decrease in temperature or evaporation, leads to a release of energy: Work is done and stresses are exerted on the walls of pores and cracks —this can result in stone decay (Lewin, 1989).

Salt hydration occurs when a salt adsorbs water as a result of surface wetting or increases in relative humidity. This change in the hydrated state of the salt is accompanied by a volume increase and results in pressure being exerted on the stone matrix, which may cause decay.

The thermal expansion of salts occurs when a salt is subject to an increase in temperature. The expansion coefficients of most salts are, however, small and this fact, coupled with the low temperature fluctuations for most buildings, means that the stresses exerted are relatively low in comparison with those caused by crystallization and hydration.

The types and concentrations of salts found on building stone vary greatly and depend on the stone type and the environment around the building. In general, the most common salts are sulphates, chlorides and nitrates. These include calcium sulphate ($CaSO_4$), sodium sulphate ($Na_2SO_4$), magnesium sulphate ($MgSO_4$), calcium chloride ($CaCl_2$), sodium chloride ($NaCl$), potassium chloride ($KCl$) and potassium nitrate ($KNO_3$); calcium and magnesium carbonates are also common (Amoroso and Fassina, 1981). Of these salts, calcium sulphate is generally found in the highest concentrations on stone buildings and is widely thought to be the most damaging to stone (Haneef *et al.*, 1989). This salt has a high hydration pressure relative to other salts, and thus hydration/dehydration decay mechanisms are important in its presence. Fluctuations in solvent temperature do not greatly affect the solubility of this salt relative to other building salts such as sodium sulphate—the solubility of $CaSO_4$ at 30°C is only 1.1 times that at 0°C; the corresponding figure is 7.0 for $Na_2SO_4$ (Gaudie, 1987). This suggests that $CaSO_4$ is not as prone to supersaturation as other building salts and is less likely to cause crystallization damage than hydration damage.

The sources of all salts causing salt-induced decay are therefore important, but sources of calcium and sulphate are of particular interest because they lead to the formation of calcium sulphate, which is the most damaging salt in buildings. Mortars contain high quantities of soluble calcium (Duffy *et al.*, 1993) and are thus a potential source of these damaging salts. Moreover, the permeable nature of many pointing and bedding mortars would suggest that they are subject to high levels of water movement by wetting, absorption and evaporation. Consequently, they may retain and/or release salts which are damaging to building stones.

The object of this paper it to determine how mortars affect the supply and movement of salts in building stones.

## 3. EXPERIMENTAL MATERIALS AND METHODS

Stone microcatchment units with mortar joints were exposed between November 1992 and August 1993 on the roof of Trinity College, in the centre of Dublin. Each microcatchment unit (Cooper *et al*, 1991; Fig. 1) consisted of a flat stone slab, 200 mm square and 50 mm deep, with a 20 mm wide flush mortar joint along one diagonal. The stone and mortar slab was surrounded by glass sides which projected 20 mm above the surface to retain the rain water which hit the unit during exposure. All stone/glass and mortar/glass joints in the units were sealed with an inert mastic to prevent water leakage. The microcatchment units were exposed with the mortar joint in a horizontal plane, and with the other diagonal at an angle of 15° to the horizontal. A drainage hole, to which a one litre plastic bottle was connected, was placed at the lowest corner; the run-off water which was collected was used for analysis.

Six such microcatchment units were fabricated—three each of Portland limestone and Baumberger sandstone which are common building stones in Britain and Germany respectively; in addition, three control units composed solely of glass (with no mortar joint) were fabricated. The Portland limestone used in the units is classified as a fine grained oolitic limestone containing shelly fragments. It has a porosity of 13% and is composed of calcite with small amounts of magnesium carbonate and quartz. The Baumberger is classified as a quartz bearing limestone composed predominantly of quartz and calcite with smaller amounts of feldspar, montmorillonite, mica and glauconite; it has a porosity of approximately 21%. The mortar used in the microcatchment units was made of siliceous sand, ordinary Portland cement and hydrated lime in the ratios of 6 : 1 : 1, respectively, as this, is a mortar mix which is commonly used in the conservation and building industries. The mortar was fabricated in accordance with BS 4551: 1980 and BS 5224: 1974 and was cured for 28 d before exposure.

Dublin is a city of about one million people located on the east coast of Ireland on the Irish Sea. The exposure site was located adjacent to one of the busiest traffic junctions in the city centre on a flat roof at an elevation of about approximately 18 m. Dublin city has a temperate climate with mean daily maximum and minimum temperatures of 19.5°C and 12.5°C in summer and 8°C and 3°C in winter. Annual mean rainfall is 699 mm, prevailing wind directions are westerly, southwesterly or southeasterly. The winter and summer median smoke concentrations in 1990/91 were 22 and 19 $\mu g\,m^{-3}$; the corresponding figures for $SO_2$ were 22 and 18 $\mu g\,m^{-3}$ (Dublin Corporation, 1991).

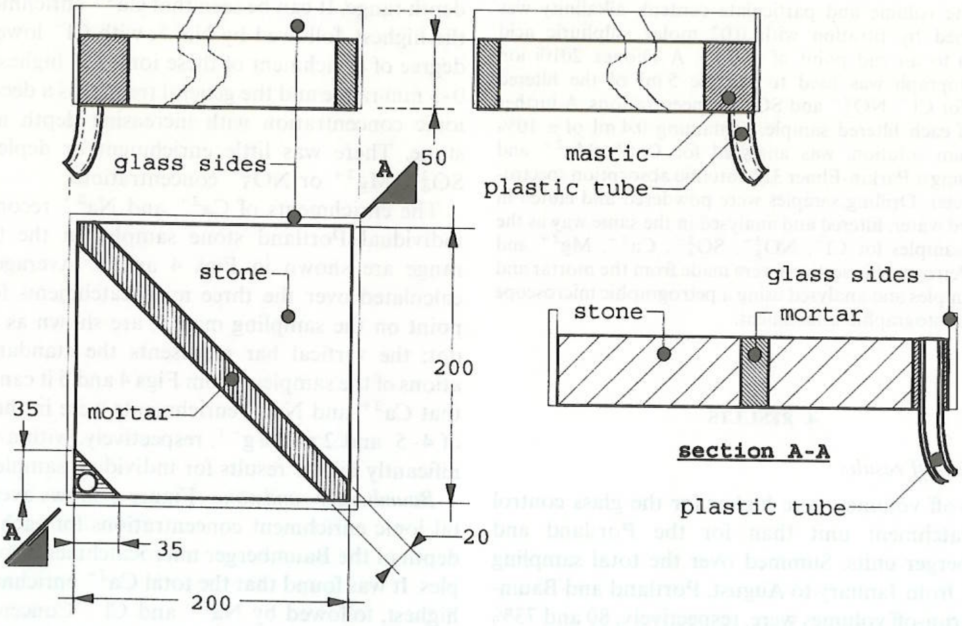

Fig. 1. Plan, elevations and section of the stone and mortar microcatchment unit. All dimensions are in millimetres.

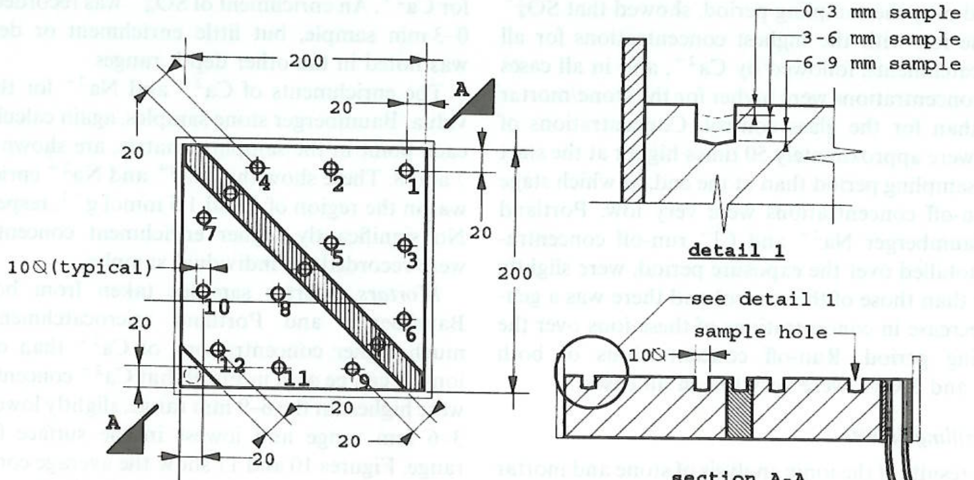

Fig. 2. Stone and mortar drilling sample points on the microcatchment units. All dimensions are in millimeters.

During the exposure of the microcatchment units, rain water run-off was regularly collected from the plastic collection bottles attached to each unit. Upon the completion of exposure, the stone and mortar of the microcatchment units were destructively analysed to determine the types and levels of soluble salts present. Drilled samples were taken from the stone and mortar in a grid pattern (Fig. 2). At each point of the grid, three samples were taken at successive 3 mm depths, as shown in Detail 1 of Fig. 2. In addition to the drilled samples, samples of stone were cut from the centre of the lower stone of the microcatchment unit, adjacent to the mortar joint, and mortar samples were cut from the joint itself for use in petrographic analysis.

The run-off from each microcatchment was analysed to determine its volume, particulate content, alkalinity and soluble chemistry. Samples were weighed and filtered to determine volume and particulate content; alkalinity was determined by titration with 0.02 molar sulphuric acid ($H_2SO_4$) to an end point of pH 4.5. A Dionex 2010i ion chromatograph was used to analyse 5 ml of the filtered sample for $Cl^-$, $NO_3^{2-}$ and $SO_4^{2-}$ concentrations. A further 10 ml of each filtered sample, containing 0.4 ml of a 10% lanthanum solution, was analysed for $Ca^{2+}$, $Mg^{2+}$ and $Na^{2+}$ using a Parkin-Elmer 3100 atomic absorption spectrophotometer. Drilling samples were powdered and eluted in deionized water, filtered and analysed in the same way as the run-off samples for $Cl^-$, $NO_3^{2-}$, $SO_4^{2-}$, $Ca^{2+}$, $Mg^{2+}$ and $Na^{2+}$. Petrographic sections were made from the mortar and stone samples and analysed using a petrographic microscope with a photographic attachment.

## 4. RESULTS

### 4.1. *Run-off results*

Run-off volumes were higher for the glass control microcatchment unit than for the Portland and Baumberger units. Summed over the total sampling period, from January to August, Portland and Baumberger run-off volumes were, respectively, 80 and 73% of those for the glass control unit. Regarding the alkalinity of the run-off, this was lowest for the control unit; although neither stone type showed consistently higher values than the other, nor did values show any trend over the sampling period. Ionic concentrations, totalled over the sampling period, showed that $SO_4^{2-}$ was the ion with the highest concentrations for all microcatchments, followed by $Ca^{2+}$, and in all cases these concentrations were higher for the stone/mortar units than for the glass control. Concentrations of $SO_4^{2-}$ were approximately 50 times higher at the start of the sampling period than at the end, at which stage the run-off concentrations were very low. Portland and Baumberger $Na^{2+}$ and $Cl^-$ run-off concentrations, totalled over the exposure period, were slightly higher than those of the control and there was a general decrease in concentrations of these ions over the sampling period. Run-off concentrations of both $Mg^{2+}$ and $NO_3^{2-}$ were very low in all cases.

### 4.2. *Drilling results*

The results of the ionic analysis of stone and mortar drilled samples from the Portland and Baumberger microcatchment units are illustrated in Figs 3–10. The ionic concentrations are expressed as enrichment values, calculated by subtracting unweathered stone values from the exposed stone values, to give ionic enrichment values for the exposed stone. No unweathered control has been used to adjust the drilled mortar sample ionic concentrations, which are expressed as total ionic concentrations. All ionic concentrations are expressed in millimoles per gram dry weight of sample.

*Portland limestone.* Average total ionic enrichment concentrations for each sample depth of the Portland stone are shown in Fig. 3. These averages are calculated over the values obtained at all 36 stone sample points on the three microcatchment units at a given depth range. It can be seen that $Ca^{2+}$ enrichment was the highest, followed by $Na^{2+}$, with $Cl^-$ lowest. The degree of enrichment of these ions was highest in the 0–3 mm range and the general trend was a decrease in ionic concentration with increasing depth into the stone. There was little enrichment or depletion of $SO_4^{2-}$, $Mg^{2+}$ or $NO_3^{2-}$ concentrations.

The enrichments of $Ca^{2+}$ and $Na^{2+}$ recorded for individual Portland stone samples in the 0–3 mm range are shown in Figs 4 and 5. Averages, here calculated over the three microcatchments for each point on the sampling matrix, are shown as a black dot; the vertical bar represents the standard deviations of the samples. From Figs 4 and 5 it can be seen that $Ca^{2+}$ and $Na^{2+}$ enrichments were in the region of 4–5 and 2 mmol $g^{-1}$, respectively, with a few significantly higher results for individual samples.

*Baumberger sandstone.* Figure 6 shows average total ionic enrichment concentrations for each sample depth of the Baumberger microcatchment stone samples. It was found that the total $Ca^{2+}$ enrichment was highest, followed by $Na^{2+}$ and $Cl^-$. Concentrations of $Ca^{2+}$ were highest in the 0–3 mm range with much less enrichment in the 3–6 mm range and a slight depletion in the 6–9 mm sample. Similar enrichments of $Na^{2+}$ and $Cl^-$ were recorded, but concentrations were distributed more evenly between all depths than for $Ca^{2+}$. An enrichment of $SO_4^{2-}$ was recorded in the 0–3 mm sample, but little enrichment or depletion was noted in the other depth ranges.

The enrichments of $Ca^{2+}$ and $Na^{2+}$ for the individual Baumberger stone samples, again calculated at each point in the sampling matrix, are shown in Fig. 7 and 8. These show that $Ca^{2+}$ and $Na^{2+}$ enrichment was in the region of 5 and 1.5 mmol $g^{-1}$, respectively. No significantly higher enrichment concentrations were recorded for individual samples.

*Mortars.* Mortar samples taken from both the Baumberger and Portland microcatchments had much higher concentrations of $Ca^{2+}$ than of other ions. It can be seen in Fig. 9 that $Ca^{2+}$ concentrations were highest in the 6–9 mm range, slightly lower in the 3–6 mm range and lowest in the surface 0–3 mm range. Figures 10 and 11 show the average concentrations of all other measured ions in the Baumberger and Portland mortar samples. Of these ions, $Cl^-$ and $SO_4^{2-}$ concentrations were highest in both microcatchment mortars. Concentrations of $SO_4^{2-}$ were highest in the surface 0–3 mm. $Cl^-$ concentrations were more evenly distributed throughout the three sampling depths. Concentrations of $Na^{2+}$ were highest in the 0–3 mm range, but overall concentrations were low. In all cases, $Mg^{2+}$ and $NO_3{}^{2-}$ concentrations were very low.

### 4.3. *Petrographic results*

A thin section analysis of the stone samples taken from below the mortar joints revealed few changes in the stone structure, although there was widespread evidence of reprecipitated calcite in the surface pores

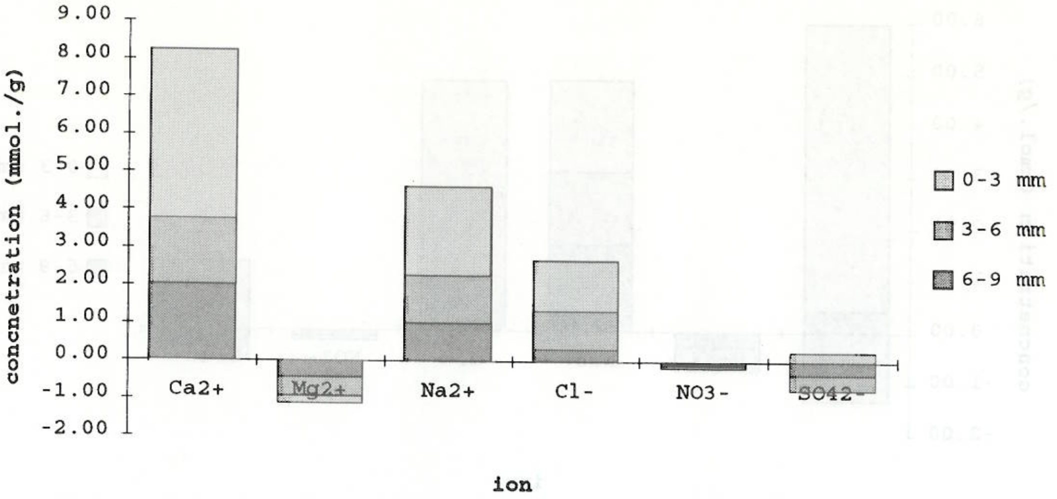

Fig. 3. Average total ionic enrichment concentrations at each depth for the Portland stone samples.

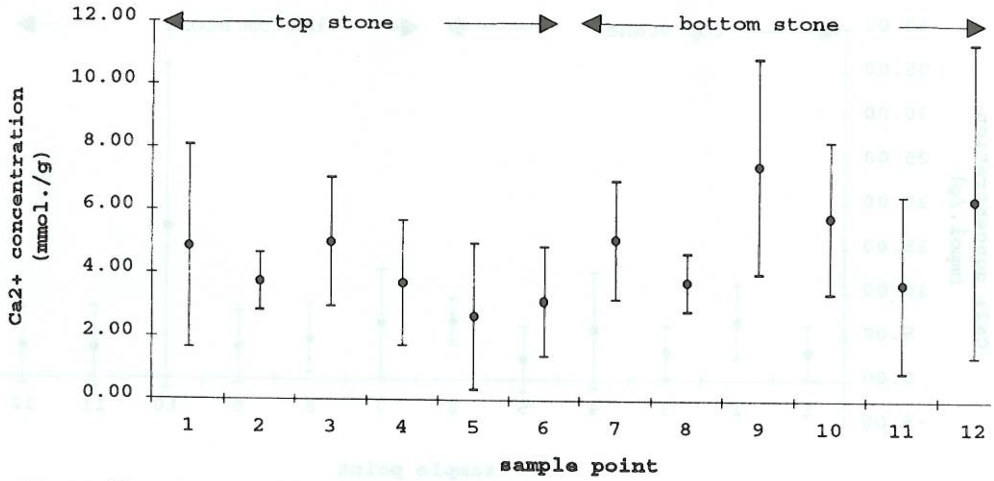

Fig. 4. $Ca^{2+}$ enrichment concentrations for the 12 Portland stone samples in the 0–3 mm range. The black dots are average values and the vertical bars represent standard deviations. For sample point positions, refer to Fig. 2.

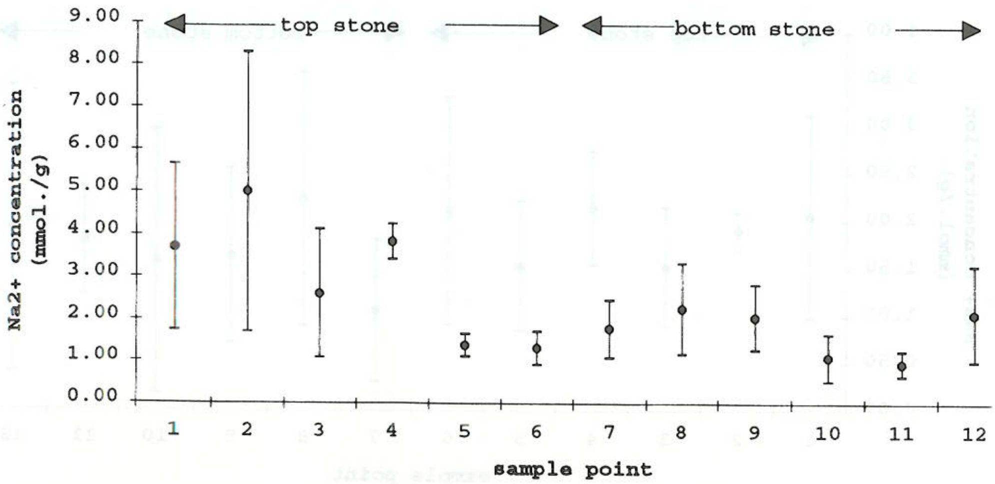

Fig. 5. $Na^{2+}$ enrichment concentrations for the 12 Portland stone samples in the 0–3 mm range. The black dots are average values and the vertical bars represent standard deviations. For sample point positions, refer to Fig. 2.

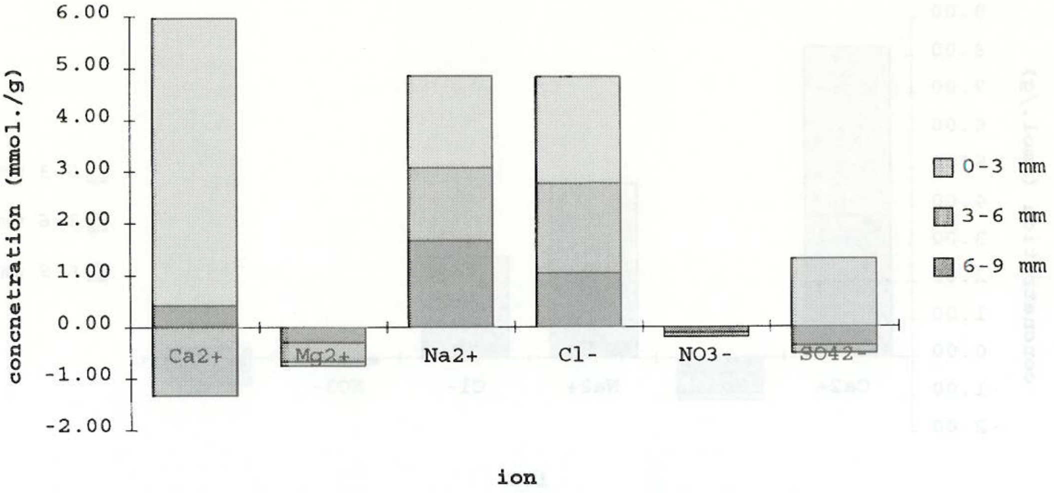

Fig. 6. Average total ionic enrichment concentrations at each depth for the Baumberger stone samples.

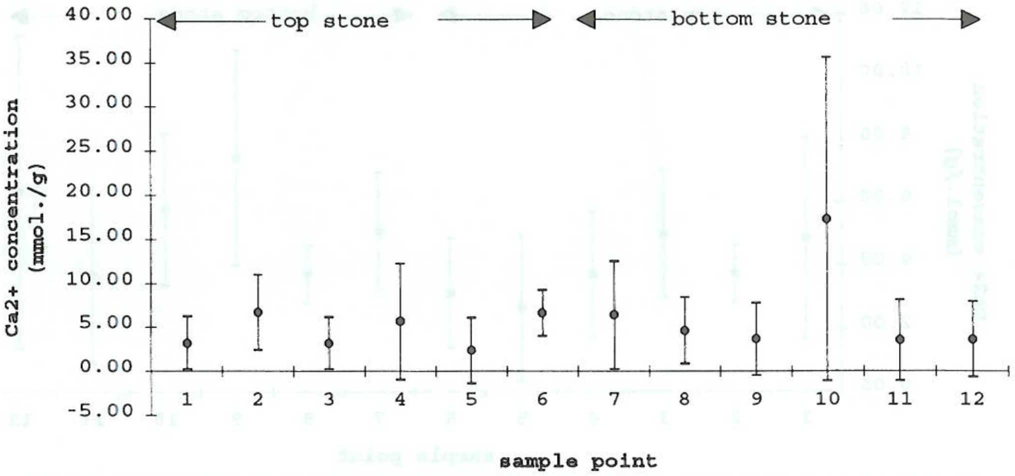

Fig. 7. $Ca^{2+}$ enrichment concentrations for the 12 Baumberger stone samples in the 0–3 mm range. The black dots are average values and the vertical bars represent standard deviations. For sample point positions, refer to Fig. 2.

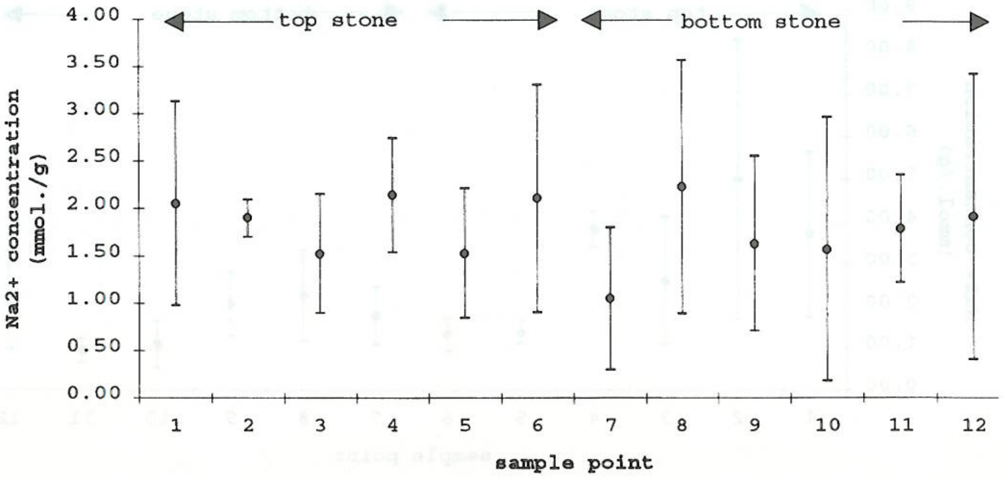

Fig. 8. $Na^{2+}$ enrichment concentrations for the 12 Baumberger stone samples in the 0–3 mm range. The black dots are average values and the vertical bars represent standard deviations. For sample point positions, refer to Fig. 2.

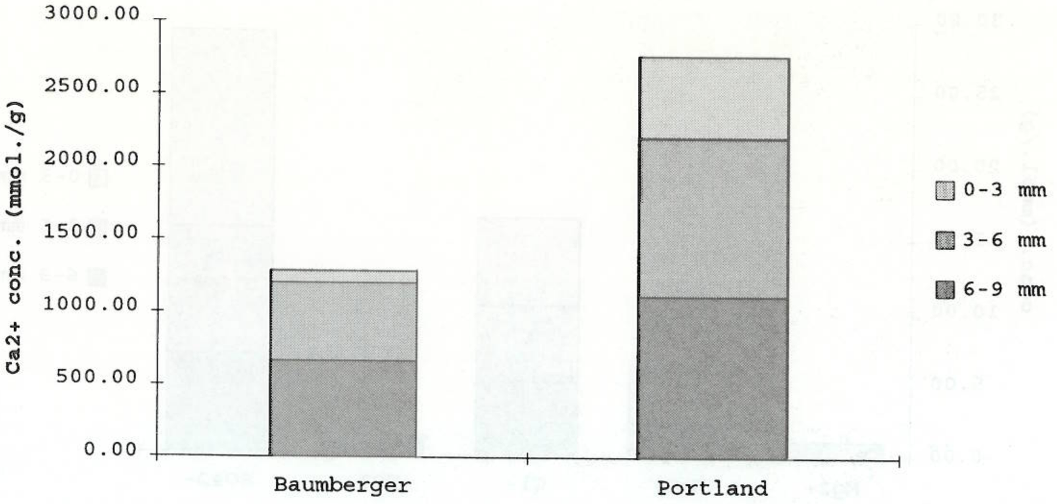

Fig. 9. Depth profiles of the average $Ca^{2+}$ concentrations in the Portland and Baumberger microcatchment mortars.

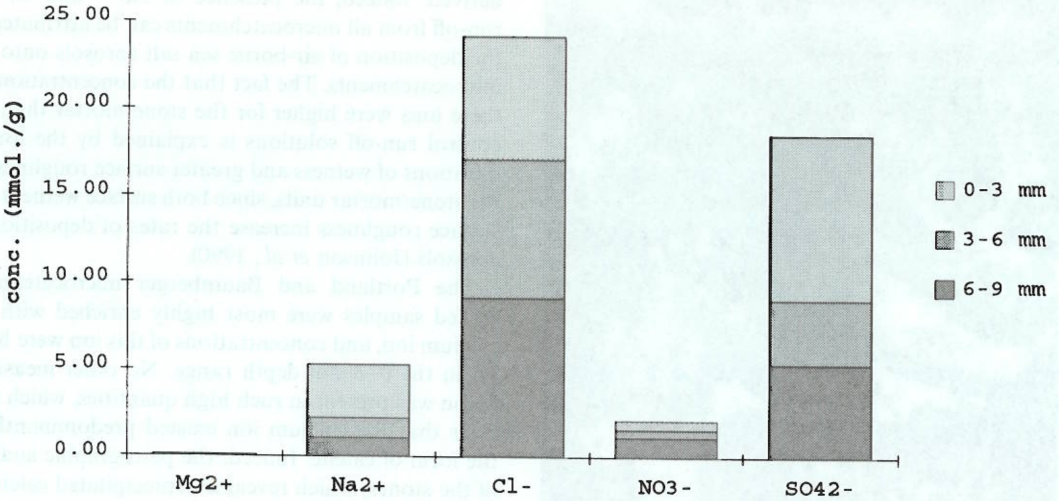

Fig. 10. Depth profiles of the average concentrations of all measured ions except $Ca^{2+}$ in the Portland microcatchment mortars.

of both stones (see Fig. 12). The petrographic analysis of the mortars revealed some surface loss of the cement/lime binder.

## 5. DISCUSSION

The fact that there was more surface run-off water from the glass control unit, and less from both the Portland and Baumberger stone/mortar units can be accounted for by the absorption of some water by the more porous stone/mortar units. This absorption of water—about 20 to 30% of all water which came into contact with the stone/mortar units—indicates a longer duration of wetness for these than for the glass microcatchment units. Water absorption further implies internal water movement and resultant stone and mortar dissolution processes. The differences between the control and the stone microcatchment run-off volumes must be accounted for by evaporation, which increases the likelihood of the presence of salts within the stone and mortar.

Run-off alkalinity was consistently higher for the stone/mortar units. This can be attributed to the dissolution of the calcium carbonate minerals in both stone types, leading to the release of $Ca^{2+}$ and $CO_3^{2-}$ into the run-off solution. Upon dissolution, these ions are available for the formation of reprecipitated calcite or, if $SO_4^{2-}$ is present, for the formation of gypsum.

The stone/mortar microcatchment run-off $SO_4^{2-}$ concentrations were high at the beginning of exposure.

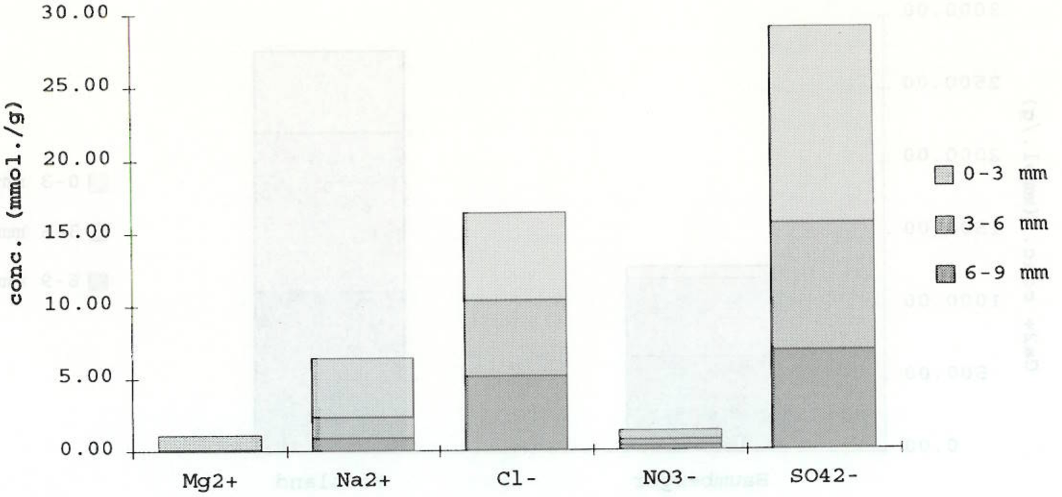

Fig. 11. Depth profiles of the average concentrations of all measured ions except $Ca^{2+}$ in the Baumberger microcatchment mortars.

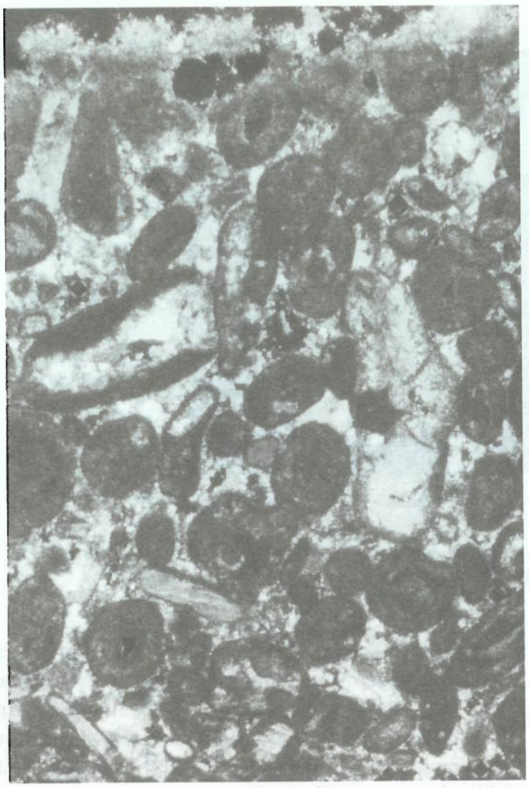

Fig. 12. A petrographic thin section showing reprecipitated calcite in the pores of one of the Portland limestone samples.

This is unusual and may be attributed to higher air- and rain-borne sulphate concentrations in winter months.

The concentrations of $Na^{2+}$ and $Cl^-$ were negligible in the stone and mortar before they were weathered, so their increased levels in the run-off suggest that these ions are largely environmentally derived. Indeed, the presence of $Na^{2+}$ and $Cl^-$ in run-off from all microcatchments can be attributed to the deposition of air-borne sea salt aerosols onto the microcatchments. The fact that the concentrations of these ions were higher for the stone/mortar than the control run-off solutions is explained by the longer durations of wetness and greater surface roughness of the stone/mortar units, since both surface wetness and surface roughness increase the rates of deposition of aerosols (Johnson *et al.*, 1990).

The Portland and Baumberger microcatchment drilled samples were most highly enriched with the calcium ion, and concentrations of this ion were highest in the 0–3 mm depth range. No other measured anion was present in such high quantities, which suggests that the calcium ion existed predominantly in the form of calcite. Indeed, the petrographic analysis of the stones, which revealed reprecipitated calcite at the surface of both microcatchment stones, supports this hypothesis. The low concentrations of $Ca^{2+}$ in the glass control run-off solution indicates a low concentration of this ion in the environment. Most of this $Ca^{2+}$ must therefore be stone and/or mortar derived.

Microcatchment stone enrichment concentrations of $Na^{2+}$ and $Cl^-$ were next highest but, unlike $Ca^{2+}$, their concentrations were similar for all sample depths. The fact that the concentrations of $Na^{2+}$ and $Cl^-$ were almost identical for the Baumberger samples, and varied by a factor of approximately 1.8 for the Portland samples, suggests that they existed predominantly in the form of NaCl. The similar concentrations of these ions at each sample depth reflects the highly soluble and mobile nature of this salt.

Little enrichment of $SO_4^{2-}$ was recorded for the stone drilled samples. It appears that any deposited sulphate was either washed directly away without reacting or formed salts, such as $CaSO_4$ or $Na_2SO_4$, which were washed away in subsequent rain events.

No significant spatial patterns of ion enrichment were observed for the matrix of stone samples studied. It appears that in a well washed surface environment, such as that of the microcatchment units, salts are transported evenly over the entire face of the stone, and no localized enrichment takes place.

The analysis of the drilled mortar samples showed that $Ca^{2+}$ concentrations were inversely proportional to depth into the mortar, whereas the opposite was the case for the stone samples. This indicates that $Ca^{2+}$ is being lost from the surface of the mortar, probably as a result of the dissolution of its binder. This in turn explains the high concentrations of $Ca^{2+}$ in the surface of the stone. Petrographic analysis of the mortar showed that a loss of surface binder had indeed occurred, further supporting the theory that $Ca^{2+}$ was redistributed from the mortar to the stone.

The mortars contained increased levels of $Na^{2+}$ and $Cl^-$, but the much higher levels of $Cl^-$ relative to $Na^{2+}$ suggests that the $Cl^-$ precipitated with another cation, possibly $Ca^{2+}$ to form $CaCl_2$. The concentrations of $SO_4^{2-}$ in the mortars were also high, especially in the surface 0–3 mm. As was the case for the $Na^{2+}$ and $Cl^-$ ions, the fact that $SO_4^{2-}$ was present in very low quantities in unweathered mortar (unweathered $Na^{2+}$, $Cl^-$, $SO_4^{2-}$ concentrations at 28 d were 0.020, 0.007 and 0.101 mmol $g^{-1}$, respectively), together with its location near the top of the mortar, suggests that this ion is environmentally derived — the majority of sulphate is probably deposited in gaseous or particulate form.

## 6. CONCLUSIONS

The high concentrations of $Ca^{2+}$ in the Portland and Baumberger run-off solutions, the enrichment of the stones by $Ca^{2+}$ and its depletion at the surface of the mortar indicates that $Ca^{2+}$ from the mortar was washed into solution and deposited onto the stone. This ion precipitates mainly as calcite in the surface cracks and pores of the stone and can lead to the blockage of surface pores, thus restricting water movement and possibly leading to decay. The enrichment of $Ca^{2+}$ in the stone was uniform over its surface. Therefore, mortar joints in well washed, inclined surface environments, such as those studied, lead to spatially uniform enrichment of $Ca^{2+}$ in the surrounding stone, both above and below the joint. The fact that the mortar microcatchment units are exposed after 28 d of curing and that $Ca(OH)_2$ carbonates quite quickly would suggest that, in real world situations — where the mortar is immediately exposed to the environment — more $Ca^{2+}$ would be deposited into the stone than in this experimental case.

As well as $Ca^{2+}$ enrichment, the stone contained increased levels of $Na^{2+}$ and $Cl^-$ due to the presence of air-borne sea salt at the exposure site. Little enrichment of $SO_4^{2-}$ took place despite the site's urban location.

Although mortars were depleted in $Ca^{2+}$ after their exposure, they were found to contain environmentally derived $Na^{2+}$, $Cl^-$ and $SO_4^{2-}$, the latter highest in the 0–3 mm range. These probably existed as NaCl, $CaCl_2$ and $CaSO_4$, respectively. Where the mortars and stones are frequently wetted and dried by rain water, these salts are dissolved and transported regularly into the adjacent stone where they can cause damage by salt crystallization and hydration. Indeed, stone around many mortar joints is badly decayed, probably as a result of this phenomenon.

To summarize, it can be stated that joint mortars act as sources of soluble ions such as $Ca^{2+}$, and sinks of environmentally derived ions such as $SO_4^{2-}$ and $Cl^-$. Mortars play a key role in the occurrence and location of salts, even in relatively soluble stones such as Portland limestone and Baumberger sandstone.

*Acknowledgements*—The authors would like to thank the Commission of the European Union for the funding which made this work possible; Tim Cooper, Director of the Stone Decay Research Team at Trinity College, Dublin; the Irish State Laboratory and the Buildings' Office, Trinity College, Dublin. The authors also wish to thank Emer Bell, Jim McDermott and Paul O'Brien.

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