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1 Durability of stone-repair mortars used in historic buildings from Paris

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10 ABSTRACT

11 Stone-mortar specimens manufactured with limestone and commercial stone-repair
12 hydraulic mortars have been exposed to accelerated ageing tests, salt crystallisation
13 (mixture of sodium chloride, sodium sulphate, calcium sulphate and potassium nitrate)
14 and acid vapours exposure (sulphurous and nitric acid) besides, their corresponding
15 control specimens were exposed to deionized water imbibition cycles and water vapour,
16 respectively. Large amounts of soluble nitrates and minor amounts of sulphates
17 precipitate on the surface of both stone and mortar materials after the acid test. This
18 mainly affects the surface of the specimens rather than the stone-mortar interface.
19 Whereas parallel fissures to the interface show up due to salt mixture crystals
20 precipitation after the salt crystallisation test.
21 A remarkable yellowing of the stone takes place after both tests. The mortar with closest
22 properties to Euville limestone, with no phyllosilicates is the most resistant. It shows
23 good mechanical properties but bad hydric interactions with the stone that could prove
24 damaging in the long run. The mortar with the highest porosity and better hydric
25 behavior but poor mechanical interaction with the stone is highly affected by the ageing
26 tests, acting more as a sacrificial mortar rather than as a repair mortar.

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28 **Keywords:** stone-repair mortars; natural stone; historic buildings; durability; salt
29 crystallisation, sulphurous-nitric acid vapours exposure

30 **1. Introduction**

31 Stone-repair mortars involve those materials that can be used for superficial repairing or
32 reconstruction of damaged stone. A missing part of the original material is modeled by
33 the new material that is adapted into the required shape [1]. The size, shape,
34 mineralogical and chemical composition of binder and aggregates are some properties
35 that determine the final behavior of a stone-repair mortar. These are critical for a good
36 compatibility with the damaged stone and to assure a long durability of a restoration
37 intervention. Besides, salt crystallisation processes greatly affect the durability of
38 building materials. As an example, the use in the eighteen-century of incompatible
39 restoration mortars in a historic monastery, lead to the ruins of this building that remain
40 in a bad state of conservation, until partial collapse, due to the too long lack of
41 maintenance [2]. Furthermore, even though international standards about additives for
42 masonry mortars allows the presence of clays in a maximum concentration of 3%, clay-
43 rich sands are frequently used by restorers because these can increase workability and
44 match the right colour for the restoration mortar. However, the main effect of clay fines
45 ($<63 \mu\text{m}$) in aggregates is an increase of the water demand, due to their high surface
46 area, for a constant mortar consistency. As a consequence, the higher water/binder ratio
47 causes a strong decrease of the mortar quality regarding to mechanical, hydric and
48 durability properties [3]. On the contrary, the durability properties of concretes
49 containing fly ash and nanosilica as partial replacement of cement are superior to
50 ordinary concrete containing 100 % cement [4].

51 Nowadays, there is a high increase of NO_x gases present in the atmosphere compared to
52 past SO₂ pollution from coal burning and consequent formation of gypsum crusts on

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limestone, which is a very well know phenomena. NO_3^- is the final product of the oxidation–hydration process of NO_x present in the atmosphere [5]. Oxidation of NO_x in the presence of water can result in nitric acid formation, which can react with calcareous stone. Although the lack of nitrates within the stone, or in runoff water, has led to the general conclusions that NO_x species have little effect on stone. For a mixture of gaseous pollutants (i.e. SO_2 plus NO_2), NO_2 enhances adsorption of SO_2 by the stone in a synergistic manner [6]. There is still a lack for testing the physical properties and durability of mortars according to European Standards, especially in terms of compatibility with stone, and the long-term behavior of the repaired stone-mortar interface [7].

The aim of this research is to determine the physic-chemical durability of both mortar and stone of stone-mortar specimens manufactured with the most frequently used commercial stone-repair mortars in restoration works of historic buildings in Paris. Their particular interactions of these mortars with a very common limestone used in the basements of these buildings have been assessed after accelerated ageing tests involving salt crystallisation and nitric and sulphurous acid exposure to achieve this goal.

2. Materials and methods

2.1. Characteristics of selected materials

Three commercial ready to mix stone-repair mortars were selected on the basis of their application on some damaged surface areas by salt crystallisation processes of the same type of stone (Euville limestone) in different historic buildings of Paris repaired in the same period of time (2008-2010).

2.1.1. Stone-repair anhydrous mortar raw materials

Three commercial mortars, ready to mix with water according to recommendations of the manufacturers, are called in this research *Lit*, *Art* and *Alt* mortars. These are

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78 mixtures of natural and hydraulic lime mortars (NHL, HL) with or without aerial lime
79 (CL). Three ready to mix with water commercial stone-repair mortars, are called in this
80 research Lit, Art and Alt mortars. These are mixtures of natural and hydraulic lime
81 mortars (NHL, HL) with or without aerial lime (CL). Lit mortar (Lithomex Light) is a
82 material produced by Chaux et Enduits St. Astier (CESA, France), based on a St. Astier
83 natural hydraulic lime binder. According to the manufacturer, this is a pure natural
84 hydraulic lime, defined as a natural lime, hydraulic binders, sand and specific additives.
85 It has a bulk density of 1325-1360 kg/m³ and gran-size distribution from 0.8 down to
86 0.08 mm. The mineralogical composition of Lithomex and Lithomex Light mortars has
87 been analysed by means of X-ray diffraction by other authors [8,9]. According to
88 Torney et al., 2015 [8], Lithomex mortar contains the following components (expressed
89 as percentage of binder): calcium hydroxide 20%; hydraulic binder (Portland cement)
90 20%; filler (vermiculite) 5%, fine grained quartz and calcite aggregates and talc filler.
91 Under the execution of Dimppa Project [9] it was determined that the Lithomex Light
92 mortar type (Lit mortar) does not have aerial lime (calcium hydroxide, i.e. portlandite),
93 which is present in the other two mortars (Art and Alt mortars). The technical data sheet
94 of Art mortar (Artopierre TM by Parexlanko, France) indicates that this is mainly aerial
95 lime (CL, binder, 70% in volume) with hydraulic, mineral and organic additives and
96 mineral pigments. Aggregates are mainly calcareous and siliceous with grains up to 1.5
97 mm. Other specified characteristics are an open porosity >30%, compression strength of
98 4MPa and soluble salts 0.12%. Finally, according to the manufacturer Alt mortar
99 (Altar® Pierre by ECP, France) contains quartz grains, calcium carbonate, hydraulic
100 binder, additives and mineral pigments. The mortar displays a porosity of 15 up to 45%,
101 adherence higher than 0.3 MPa, flexural strength 0.7 up to 9.5 MPa and compressive
102 strength values of 6 up to 45 MPa. The main mineralogical difference among these three

103 mortars containing as a hydraulic binder, di-calcium silicate (larnite), is the absence of
104 phyllosilicates in Alt mortar, which are present in Lit (chlorite and talc) and Art
105 (vermiculite) mortars [9].

106 2.1.2. Stone

107 Euville limestone (Oxfordian, Late Jurassic) is a crinoïdal grainstone almost completely
108 composed of calcium carbonate (98%) with a coarse-grained texture and a syntaxial
109 cement of calcite. The outcrops of this limestone are situated near Commercy
110 (Département de la Meuse, France). The Euville limestone has often been used as a
111 replacement stone in many historic buildings [10,11]. Even though this is a quite low
112 total porosity limestone (porosity amounts between 12 and 18 % probably due to local
113 changes in compaction), salt crystallisation processes have frequently weathered it. As a
114 restoration measure, some areas built with this stone have been repaired with several
115 stone-repair commercial mortars. The stone specimens used in this work were supplied
116 by ROCAMAT quarry in Euville, France. Stone specimens parallel to the bedding with
117 dimensions 2x4x16 cm were cut to apply a 2 cm layer of each mortar in order to
118 manufacture 4x4x16 cm stone-mortar specimens. For the water permeability test
119 cylindrical specimens 1 cm thick and 4 cm diameter were prepared. Besides, additional
120 stone-mortar specimens with 1 cm thick mortar layer applied on stone slabs of 4 x 22 x
121 22 cm were prepared to carry out the adherence test.

122 2.1.3. Stone-mortar specimens manufactured in the laboratory

123 The water-powder mortar ratios were prepared in the lab following as much as possible
124 the recommendations of the manufacturers according to their respective technical data
125 sheets. The manufacturer's preparation guidelines of Lithomex state that the materials
126 should be mixed (mechanically or by hand) for between three and five minutes, with
127 water content of 4.5–5.5 L of water per 25 kg of dry material. We used 5L of water (W)

128 per 25 kg of dry material (M), i.e. W:M=0.20. In this case, the restorer who used this
129 type of mortar in a historical building from Paris recommended us to add a handful of
130 sand to the mixture. According to grain size distribution analyses performed in the
131 project [9] and in order to have a similarity with the other two mortars, 150 g of
132 normalized sand (Ultibat, EN 12620: 2002 + A1: 2008 [12]), fraction 1.25-0.63 mm,
133 was added to 2000g of mortar. To prepare Art mortar, between 6 and 7 L of water per
134 30 kg of dry material is required. We used 6.5L per 30 kg of dry mortar powder mortar
135 (W:M=0.22). To manufacture Altar® Pierre mortar, from 4 up to 6 volume of dry
136 material per 1 volume of water is recommended. To prepare the specimens we used 5
137 volume of dry material per 1 volume of water (W:M= 0.13). In the case of this mortar,
138 before the application of the mortar layer onto the stone, the manufacturer
139 recommended to use a more diluted mixture to apply a first thin layer (some
140 millimeters) of mortar over the stone to ease the adherence. Since they did not quantify
141 this amount of water and they just follow traditional criteria, we set this amount in a
142 10% more diluted mixture.

143 A mechanical mixture was performed in the three mortars using an industrial mixer
144 (Controlab, France) during 4 minutes, starting at low speed rotation (62 rpm) during 1
145 min and finishing at high speed (125 rpm). Then, a jolting apparatus was also used
146 according to the standard test UNE-EN 196-1:2006 [13] applying 25 blows to
147 homogenize the mixture and avoiding the formation of air bubbles.

148 For hardening the mortar-stone specimens (28 days curing) we tried to simulate the
149 average annual weather conditions of Paris. The specimens were introduced into a
150 climatic chamber at 11°C and 85% relative humidity (RH)) during 7 days, 11°C and 65
151 % RH during other 7 days, followed by laboratory conditions in a climatic room at 20°C
152 and 50% RH during 14 days, under environmental CO₂ (400 ppm approx.)).

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153 To prepare the stone-mortar specimens, stone with dimensions 2x4x16 cm (for hydric
154 and mechanical tests) and 4x22x22 cm (for the adherence test) were prepared to attach
155 afterwards 2 and 1 cm mortar layer, respectively, to constitute the stone-mortar
156 specimens (4x4x16 and 5x22x22 cm respectively).

157 Seventy-two stone-mortar specimens (4x4x16 cm), twenty-four for each mortar, were
158 prepared to perform hydric and mechanical tests (Fig.1a). Six stone-mortar specimens
159 of each mortar type were manufactured to be subjected to a salt crystallisation
160 accelerated ageing test. Another six stone-mortar specimens were prepared in order to
161 be submitted to an accelerated acid exposure test. Six additional stone-mortar control
162 specimens were used for each ageing test (Fig.1b). To perform the adhesion test, ten
163 specimens were prepared one for each type of mortar and each condition of accelerated
164 ageing test (2 ageing and 2 control specimens). In the case of Art mortar, none Art
165 stone-mortar specimens could be prepared to carry out the adhesion test after the second
166 ageing test due to the lack of raw material at that moment. For this reason, only ten
167 specimens were prepared. None *Art* stone-mortar specimens with these latter
168 dimensions could be prepared to carry out the second ageing test due to the lack of raw
169 material at that moment. To carry out water vapour permeability tests, plastic molds
170 filled with each mortar mixture were used. These were big enough to prepare by cutting
171 at least two mortar circular specimens (50 mm diameter x 10 mm thickness) for each
172 type of mortar and condition of ageing test. The mortar mixtures were desmoulded after
173 two days and cured during 28 days under the same environmental conditions than the
174 other specimens used for the rest of the tests.

175 All the stone-mortar specimens were submitted to accelerated ageing tests right after the
176 28 days of curing and then immediately characterized at the end of these tests.”

177 2.1.4. Accelerated ageing tests

178 To assess the durability of the stone-mortar specimens, two different types of
179 accelerated ageing tests were performed involving different types of salt precipitation.

180 2.1.4.1. Salt crystallisation ageing test

181 The composition/nature and concentration of salts used in this test was based on the
182 XRD, MEB-EDS and IC results obtained in the mortar and stone samples collected
183 from the buildings. A solution with a mixture of salts of sodium chloride (NaCl),
184 sodium sulphate (Na₂SO₄), calcium sulphate (CaSO₄) and potassium nitrate (KNO₃)
185 was prepared. Their relative concentration was approximately three times higher than
186 the obtained by ion chromatography in the samples from the buildings under the
187 execution of Dimppa Project [9]. Three groups of six stone-mortar specimens
188 manufactured with each mortar type were placed in separate closed plastic containers
189 (30 litres volume). The stone part of each stone-mortar couple was placed on the base
190 with a separation distance of 3cm between each specimen. Twelve litres of salt solution
191 with a concentration 0.1% CaSO₄, 3% Na₂SO₄, 1% KNO₃ and 1% NaCl was spilled
192 inside each container covering the stone parts until reaching 0.5 cm below the stone-
193 mortar interfaces. All specimens were weighted before and after 3 hours immersion in
194 this solution. Then, these were placed in a stove at 60°C and left to dry for 18h. After
195 that, all the specimens were cooled down under laboratory conditions (19±1°C and 45±5
196 % relative humidity) for 3 hours and then each specimen was weighted and pictured in
197 all their angles. These steps make one cycle and they were repeated until reaching
198 eleven cycles, renewing each salt solution at the end of each cycle.

199 The same procedure was done with other three groups of six stone-mortar specimens
200 but instead of salt solution de-ionized water was spilled in another three different plastic
201 containers. Finally, at the end of the cycles (salt and water cycles) all the specimens
202 were submerged in deionized water during 24 hours to leave rid out the accumulated

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203 salts. These stone-mortar specimens are named as aged 1 and control 1 specimens,
204 respectively.

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206 2.1.4.2. Acid exposure ageing test

207 The second accelerated ageing test consisted in the exposure of the specimens to SO₂
208 and NO_x gases. As a reference, the standard test UNE-EN 13919:2003 [14] was used,
209 which involve sulphurous acid (H₂SO₃) diluted in water, leaving the solution evaporate
210 inside a closed container where the specimens are stored during 21 days. In our test,
211 besides the sulphurous acid (6% concentration), a nitric acid (HNO₃) solution was used
212 (78% concentration). Each group of stone-mortar specimens was placed in closed
213 plastic containers, with the stone part in the base. Three groups of six stone-mortar
214 specimens were placed in three different containers together with 400 ml of both acid
215 solutions introduced in glass beakers of 100 ml, two beakers containing the nitric acid
216 (200 ml) and the other two beakers containing the sulphurous acid (200 ml). These were
217 disposed in the four corners of the plastic containers. Control groups of six stone-mortar
218 specimens were exposed to water vapour by placing them in three different containers
219 together with four 100 ml glass beakers of de-ionized water (400 ml), creating a high
220 relative humidity atmosphere. The samples were pictured and weighted before and after
221 20 days inside the closed containers at a room temperature of 20°C. These stone-mortar
222 specimens are named as aged 2 and control 2 specimens, respectively.

223 The same curing conditions were applied to additional plastic containers used to
224 introduce the specimens set aside for the adherence and water vapour permeability tests.

225 2.2. Analytical techniques and experimental test methods

226 2.2.1. Mineralogical and chemical analyses

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227 Polarizing light optical microscopy was used to study the texture, grain size, porosity
228 and the main mineralogical constituents of the specimens. The samples were
229 impregnated with epoxy resin mixed with blue dye to fill the porosity in order to be
230 easily recognized under the microscope with parallel nicols. The thin sections were
231 studied with an Olympus BX50 polarized light microscope fitted with an Olympus
232 digital camera.

233 Scanning electron microscopy (SEM) coupled with Energy dispersive X-ray
234 spectroscopy (EDS), was carried out to study changes in morphology and precipitation
235 of salts on the surface of the mortar specimens. A SEM microscope Leica S430i was
236 used. The elemental composition of some selected components was qualitatively
237 determined by means of EDS microanalyses by means of a Bruker micro-analyzer
238 spectrometer. Nickel-sputtered fragments from the samples were studied in secondary
239 electrons mode.

240 Ion chromatography (IC) was performed to quantify the soluble salts present in the
241 stone and mortar samples after the ageing tests. Some anions (Cl^- , NO_3^- , PO_4^{2-} and
242 SO_4^{2-}) and cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+} and NH_4^+) were determined. Approximately
243 0.1 g of sample was dissolved in 10 ml of Milli-Q ultrapure water and placed it for 5
244 min in an ultrasonic bath at room temperature. The solution with the solid residue was
245 then left to settle down with a minimum rest period of 24 h. The soluble salts were
246 quantified on a Dionex ICS-900 ion chromatograph.

247 2.2.2. Physical tests

248 To determine hydric and mechanical properties, together with physical characteristics
249 such as density, porosity and colour parameters, the following tests were carried out in
250 the stone-mortar specimens.

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251 Water absorption by capillarity was carried out to evaluate the liquid water transfer into
252 the materials (placing the stone part on the base in contact with the water layer) and to
253 calculate their capillary coefficients. The standard test AFNOR NF EN 1925:1999 [15]
254 was followed; the results are expressed in $\text{g}/(\text{m}^2 \cdot \text{s}^{1/2})$ instead of $\text{kg}/(\text{m}^2 \cdot \text{s}^{1/2})$. Once
255 capillarity saturation was reached, the water drying (evaporation) was performed under
256 laboratory conditions (45 ± 5 % RH and $20 \pm 1^\circ\text{C}$) to calculate the desorption coefficient
257 (water loss) by weighing the specimens at several intervals of time

258 Water vapour permeability test was carried out in the stone and mortar specimens aged
259 by salt crystallisation and their corresponding control groups. The standard test AFNOR
260 EN 15803:2010 [16], with wet cup was followed to calculate the water vapour
261 permeability ($\text{Kg}/(\text{m}^2 \cdot \text{s})$). External relative humidity of 50% and temperature of 23°C
262 were achieved introducing the specimens in a climatic chamber, setting these conditions
263 during all the duration of the test. The first two millimeters of the mortar discs
264 specimens were removed to avoid the presence of laitance (surface coating developed
265 during hardening [8]).

266 Flexural strength measurements were carried out with a maximum load of 8 kN and
267 load rate of 0.05 kN/S. Compression strength measurements were performed in the four
268 fragments obtained from the breaking of two specimens after the flexural test. A
269 maximum load of 250 kN and load rate of 2.4 kN/S was applied until breaking,
270 providing only values equal or above 7 MPa, since this is the minimum value detected
271 by the compression cell. The mortar part of each stone-mortar couple was place on the
272 base of a Quantech 3R press (QuantiumTM, Researchers & Realisations, France)
273 following the standard test AFNOR EN196-1: 2006 [12].

274 Adhesion test was performed in the stone-mortar specimens (5x 22 x 22 cm) after the
275 accelerated ageing tests and their corresponding control groups. The standard test

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276 AFNOR EN 1015-12:2001 [17] was followed to determine the bonding strength (kN)
277 with a James bond tester®. The bond test was carried out performing two measurements
278 on the surface (22 x 22 cm) of each stone-mortar specimen.

279 Total Hg-porosity and pore size distribution was carried out by mercury intrusion
280 porosimetry. Small fragments (1x1x1cm approx.) were cut individually from the stone
281 and the mortar located at the stone-mortar interface. Readings were taken at pore radius
282 of 0.003–200 µm under measuring conditions ranging from atmospheric pressure to 228
283 MPa on a Micromeritics Autopore IV 9500.

284 Colour parameters were measured on the surface of stone and mortar of the stone-
285 mortar specimens. L* parameter i.e. luminosity, hue and saturation i.e. Chroma, C), a*
286 (red-green) and b* (blue-yellow) coordinates were obtained. Total colour difference
287 ΔE^* was provided as a result of the formula $\Delta E^* = ((\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2)^{1/2}$.
288 Measurements were performed with a spectrophotometer Konica MINOLTA CM-
289 2300d using the CieLab colour space, standard illuminant D65 and observer angle 10°.

290 **3. Results**

291 The accelerated ageing tests carried out in the stone-mortar specimens have
292 significantly changed the physico-chemical properties of both mortar and stone
293 materials. The mortars are more affected by the salt crystallisation and acid exposure
294 tests compared to the stone. Observations made with the naked eye, show the
295 development of salt efflorescences on the surface of Art mortar and in a lower extent
296 over Alt mortar surface after the salt crystallisation test. Lit mortar is not affected by
297 salt efflorescences, however powdering of the surface took place after the acid exposure
298 test. Control specimens from each test, exposed to deionized water imbibition (control
299 1) and water vapour (control 2) respectively, only have affected Art mortar surface. A
300 surface crust (or laitance layer) with signs of cracking and detachment has been

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301 developed on the surface of Art mortar, especially after the salt test. The hydric and
302 mechanical properties have also changed due to modification in the pore size system. A
303 great change in the colour parameters of the specimens can also be observed, especially
304 in the stone-mortar specimens subjected to the acid exposure test.

305 3. 1. Mineralogical and chemical characterization

306 A clear difference can be observed at the interface between Lit mortar and Euville stone
307 in both control and aged stone-mortar specimens. In some parts of the interface the
308 mortar penetrates in the larger pores of the stone. The most abundant aggregates with
309 smaller sizes embedded in the lower crystalline matrix of the mortar compared to
310 Euville are observed in Lit mortar which also displays higher porosity (Fig.2a and 2b).
311 There are almost no differences between specimens from control 1 (submerged into
312 deionized water) and control 2 (exposed to water vapour); except the higher quantity
313 and larger porosity located in some parts at the interface of the former specimens. The
314 interface of Lit-Euville specimens subjected to both ageing tests show clear differences
315 between them. Whereas the interface of the specimens subjected to ageing 2 (exposed to
316 acid vapours) are quite similar to their corresponding control specimens. Those
317 specimens subjected to ageing 1 (submerged in the salt solution) display fissures
318 parallel to the interface. In this case, the pores (filled with the blue resin) are spotted due
319 to the salt crystals that remained after the test (Fig.2c and 2d). There are almost no
320 differences in the surface of Lit mortar from the two types of control specimens (both
321 display cracking of feldspars).

322 Art mortar-Euville stone control specimens show greater textural differences between
323 the limestone and the mortar compared to the other stone-mortar specimens. A thin
324 fissure developed along the interface of both materials can be observed while some
325 larger pores of the stone are filled with this mortar. This type of fissure can be clearly

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326 distinguished in the control 1 specimens (Fig.3a and 3b), being this even more
327 continuous along the interface of the aged 1 specimens. Salt crystals can be observed
328 embedded in the blue resin that fills the pores of both mortar and stone. The fissure at
329 the interface of both materials is not observed in the specimens exposed to water and
330 acid vapours. Alteration of phyllosilicates, cracking of feldspar grains and
331 disaggregation of limestone aggregates is observed in the mortar surface of the aged 1
332 specimens (Fig.3c and 3d).

333 The interface between Alt mortar and Euville stone can be easily recognized by the
334 different grain sizes of both materials. Although there is a high compaction degree of
335 this mortar with almost no visible porosity (Fig.4a and 4b) in some areas higher
336 porosity close to interface is observed only in the control 1 and aged 1 specimens. The
337 main difference found in the surface of the mortars from the two control groups is the
338 higher quantity and larger porosity in the specimens from control 2. The surface of the
339 aged specimens shows cracking of feldspars, especially in the specimens from ageing 1.
340 Higher porosity and larger pores compared to their corresponding control specimens are
341 observed at the surface of aged 1 specimens. Salt crystallisation can be observed at the
342 edges of the pores located at the surface of aged 2 specimens (Fig.4c and 4d).

343 The SEM observations together with the EDS chemical analyses indicate the salt
344 crystallisation of sodium chloride, sodium and calcium sulphates with some minor
345 amounts of potassium on the surface of the specimens after the salt crystallisation test.
346 Sodium chloride (NaCl) was assigned to those crystals only with Na and Cl elements in
347 their composition. Calcium and sodium sulphates crystals (CaSO₄ and Na₂SO₄) were
348 identified only when Ca or Na, plus S and O were analyzed in the respective spectra.

349 Mainly calcium sulphate is crystallized after the acid exposure test (Fig. 5). Art-Euville
350 specimens are the most affected samples, which mortar surfaces show a higher degree

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351 of salt crystallisation, especially of sodium chloride (Fig. 5a). This sample also shows
352 acicular crystals that look like ettringite sulphates (Fig.5b). Plenty of portlandite re-
353 crystallisation is observed in Alt control 1 specimens, while calcium sulphate crystals
354 are observed close to calcium aluminosilicate (CAS) spherical particles with signs of
355 cracking after acid exposure ageing test (Fig.5c). Lit mortar also shows calcium
356 sulphate crystals on its surface. Sodium sulphate crystals, with some calcium and
357 potassium, are also observed in this mortar and especially in Art mortar surface after the
358 salt crystallisation test (Fig.5 d).

359 These salts are especially concentrated in the mortars and not in the stone, which just
360 display 0.1% Wt of chlorides and sulphates in the three cases (Table 1). Art mortar is
361 the one with the highest salt concentration after the test, displaying chlorides, sulphates
362 and nitrates (1.1% Wt), followed by Alt mortar (0.5% Wt) and Lit mortar (0.3% Wt).
363 Soluble nitrates are the most abundant salts precipitated after the acid vapours exposure
364 (Table 2). The highest values are obtained in Art mortar (488 ppm) followed by Alt
365 mortar (161 ppm) and Lit mortar (128 ppm). Precipitation of sulphates also occur in the
366 surface of the mortars, with the highest values obtained in Art mortar (198 ppm),
367 followed by Lit mortar (55 ppm) and Alt mortar (47 ppm). In the stone samples attached
368 to each mortar, the same soluble salts are detected in concentrations between 82 and 181
369 ppm of nitrates and between 26 and 108 ppm of sulphates. In general, both nitrates and
370 sulphates concentration is lower in the stone compared to the mortar. However, the
371 stone sample attached to Lit mortar contains higher concentration of nitrates than the
372 mortar, and the one attached to Alt mortar contains higher concentration of sulphates.
373 All the samples contain similar amounts of chlorides (circa 6 ppm). The concentration
374 of these salts in bulk material (only identified anions) represent percentages around 1.2

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2 375 % Wt in Alt mortar, 1.4 % Wt in Lit mortar and circa 5% in Art mortar. This
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4 376 concentration in the stone represents between 1.3 and 1.5 % Wt.

5 377 3.2. Physical characterization

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7 378 Looking at the physical properties of couple stone-mortar specimens after ageing tests
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9 379 several facts can be observed. On the water absorption capillarity curves, the slope of
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11 380 the first linear part can be considered as the capillarity coefficient (C) of Euville stone
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13 381 (bottom of the specimens). The slope of the second part corresponds to the capillarity
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15 382 absorption of mortars (upper part of the specimens). This split of the curve is based on
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17 383 our own observations and calculations from the results of the test. A different capillarity
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19 384 behavior between mortar and stone can be inferred from this split of the capillarity
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21 385 curves (Table 3). The capillarity coefficients (C) of Euville stone are much higher than
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23 386 those of the mortars. This difference is even higher in Euville-Lit specimens and lower
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25 387 in Euville-Art specimens. The ageing test has affected in a higher extent the stone in
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27 388 contact to Lit and Alt mortars, increasing their C values from 80 up to 93 g/m².s^{0.5} and
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29 389 82 up to 103 g/m².s^{0.5}. C values of the stone in contact to Art mortar have not changed
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31 390 after this salt crystallisation test. C values of the mortars slightly increase after the test
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33 391 except in Alt mortar that slightly decreases. In all the cases desorption coefficient after
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35 392 ageing tests is just slightly lower (in absolute value) than the control samples.

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37 393 Density values (apparent and bulk) are higher in all samples after ageing tests than in
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39 394 the control specimens (Table 4). Differences in porosity of Euville limestone between
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41 395 the control and the aged 1 samples depend on the mortar that was in contact with. A
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43 396 decrease in porosity is produced in Euville stone in contact to Lit mortar. However,
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45 397 there is an increase in porosity of Euville stone in contact to Art and Alt mortars.

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47 398 The pore size distribution (PSD) of Euville limestone is not very different from control
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49 399 1 to aged 1 specimens. However, the porosity values and PSD of control and aged
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400 mortar samples show many differences. Total Hg-porosity in aged mortars is slightly
401 higher in Lit mortars and much higher in Art mortar, but it decreases in Alt mortar
402 regarding to their corresponding control samples. There are small differences on the
403 main peaks positions of each porosimetry curve (Fig. 6), with the exception of Art
404 mortar curve that undergoes a great transformation. PSD is larger and more uniform in
405 aged samples than in the control samples.

406 The mechanical properties of the samples have slightly changed after the accelerated
407 ageing tests (Table 5). Flexural strength values do not really change after the ageing
408 tests. No differences between the results from both ageing tests can be observed, except
409 the in Euville-Alt specimens which flexural values are higher after control and ageing
410 test 1 compared to test 2 (7-9 MPa and 5-6 MPa respectively). This also happens with
411 Euville-Art specimens, however this is not representative since only one specimen
412 could be tested and there is no standard deviation in this case in this case. The flexural
413 values of Euville-Alt specimens are also higher compared to the obtained in those
414 manufactured with the other two mortars (between 2 and 4 MPa). Regarding to
415 compression strength results, the highest values are also obtained in Euville-Alt
416 specimens, especially after control 1 test (17MPa). The results are much lower in the
417 aged 1 specimens with similar values to those from test 2 (between 10 and 12 MPa).
418 The specimens manufactured with the other two mortars display compression values
419 below 10 MPa and below 7 (i.e. minimum value detected by the compression cell).
420 These latter lowest values are obtained in Euville-Lit specimens after test 1 and in
421 Euville-Art specimens after test 2. The second type of ageing has a lesser incidence on
422 the mechanical properties compared to the first one.

423 The results of the adhesion tests carried out in control and aged specimens are presented
424 in Table 6. The most significant result of this test is the bonding strength between Alt

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425 mortar and Euville that is stronger than in the other specimens. All the Alt mortar-
426 Euville stone couples have been broken though the stone. The bonding strength of Art-
427 Euville specimens is really weak; sometimes these are broken at the interface, even
428 when the steel disk to perform the test was glued to the surface of the mortar. The
429 strengths values obtained in Lit-Euville specimens mortar are more variable. Most of
430 the specimens were broken though the mortar but others though the interface or though
431 the stone. The bonding strength values are more or less the same in control and aged
432 specimens. However, this is higher in ageing test 1 compared to ageing test 2 in the Alt-
433 Euville specimens and the opposite in the Lit-Euville specimens. It was not possible to
434 perform the accelerated ageing 2 test for this adherence analyses.

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435 Table 7 and Table 8 show the colour parameters obtained on both stone and mortar
436 surfaces of the stone-mortar specimens. The highest total colour difference (ΔE^*)
437 between the stone and the mortars after both accelerated ageing tests and their
438 corresponding control specimens is produced between Alt mortar and Euville stone.
439 This difference is even higher between the control 1 specimens (ΔE^* 14) and it
440 decreases in the salt aged specimens (ΔE^* 9) (Table 7). Similar values are obtained in
441 the aged 2 and control 2 specimens (Table 8), being lower than aged 1 specimens (ΔE^*
442 around 7.5). Lit mortar, even though ΔE^* value is quite low in the control specimens
443 (circa 3 in control 1, and circa 6 in control 2) this value increases after both ageing tests
444 (above 7). Art mortar displays a ΔE^* value that greatly decreases from the control 1
445 specimens to the aged 1 specimens (from 6 down to 2). The ΔE^* between this mortar
446 and the stone subjected to control and ageing 2 is quite similar (around 5).

54 447 **4. Discussion**

56 448 4.1. Chemical and mineralogical characterization

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449 Observations made with the naked eye, show that the most meaningful results are the
450 development of salt efflorescences in Art mortar and in a lower extent in Alt mortar,
451 after the salt crystallisation test. Lit mortar is not affected by salt efflorescences,
452 however the powdering of the surface was detected after the acid exposure test. Control
453 specimens from each test exposed to deionized water imbibition (control 1) and water
454 vapour (control 2) respectively, only have affected Art mortar surface. A surface crust is
455 developed, showing signs of cracking and detachment of the surface of specimens
456 subjected to the salt crystallisation test. Large amounts of soluble nitrates were detected
457 by ion chromatography (which is not possible by SEM-EDS) together with minor
458 amounts of sulphates on the surface of the stone and mortar specimens after the acid
459 exposure test (ageing 2). This test has affected in a larger extent the surface of the
460 specimens rather than the interface stone-mortar specimens. The thin sections show a
461 clear difference at the interface between the mortars and Euville stone in both control
462 and aged specimens. Whereas the interface of the specimens exposed to acid vapours
463 are quite similar to their corresponding control specimens, those specimens submerged
464 in the salt solution, display parallel fissures to the interface. The cracking of feldspars
465 affected by hydrolysis is observed after the control and ageing tests (especially in those
466 exposed to salt crystallisation) in all the mortars (Figs.2-4). SEM observations show salt
467 crystallisation of sodium chloride, sodium and calcium sulphates with some minor
468 amounts of potassium after salt crystallisation. Crystallisation of calcium sulphate is
469 mainly observed after the acid test (Fig.5). Even though the specimens subjected to the
470 salt crystallisation test were submerged in deionized for 24 h at the end of the last cycle
471 of the test, these still contain salts that could not be ruled out. Art-Euville specimens are
472 the most affected samples, which surfaces show a higher degree of salt crystallisation,
473 especially of sodium chloride. Besides, acicular crystals that resemble ettringite

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474 sulphates could explain the development and cracking of the thin crust on the surface of
475 these specimens. The presence of gypsum and aluminate hydrate is a necessary
476 condition for ettringite formation, controlled by the amount of water and soluble CO₂,
477 which could both determine the conditions of ettringite precipitation [18]. Lit mortar is
478 the one less affected by this test, whereas Art mortar is the most affected due to a higher
479 absorption of salt solution and hence a further greater extent of salt crystallisation inside
480 the pores. This is related to its original composition, also prone to precipitate ettringite
481 crystals, together with higher capillarity rates that eventually cause larger modifications
482 of the pore system (PSD) and the increase of total porosity (Fig.6).

483 Plenty of micro to sub-micrometric portlandite crystals is observed in Alt control 1
484 specimens. Besides, calcium sulphate is observed close to calcium aluminosilicate
485 spherical particles with signs of cracking after the acid test. The large amount of nitrates
486 precipitated after the acid exposure test is especially concentrated on the surface of the
487 mortars and not in the stone. Art mortar is also the most affected by this test, with a
488 higher development of nitrates compared to the other mortars. In this case, Lit mortar is
489 also the less affected by nitrates but is similarly affected by sulphates as Alt mortar. In
490 general, concentrations of nitrates and sulphates are lower in the stone compared to the
491 mortar.

492 4.2. Physical characterization

493 The three mortars react in a different way under a same weathering agent/process, due
494 to their different water transfer properties. In all cases, the desorption coefficient after
495 the ageing tests is smaller (in absolute value) than the control specimens, so it can be
496 inferred that remains of salt crystals in the pores slow down water evaporation. The
497 capillarity coefficient of Lit mortar specimens increases of about 75%, assuming that
498 salt ageing test modifies its hydrophobic behavior. The content of clays in its

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499 composition and the possible presence of a water-repellent product in the mortar
500 composition could explain the formation of fissures. These fissures are observed both at
501 the interface of the stone-mortar repaired areas at the Grand Palais building [9] and
502 close to the interface in the stone-mortar specimens (Fig.2c).

503 The density values (apparent and bulk) are higher in all samples after the ageing tests
504 than in control specimens. This can be explained by the presence of salts, especially
505 Na_2SO_4 with a density of 2.66 g/cm^3 . One of the most remarkable results is the
506 appearance of a “patina” (colour change) on the surface of the three types of aged
507 mortars and the development of a crust on the surface of the aged Art mortar specimens.
508 This crust is linked to an important degradation of the surface. It shows cracking and
509 granular disintegration consequence of the saline solution transfer and mainly of the
510 dissolution/crystallisation cycles with also give rise to precipitation of ettringite
511 crystals. In the case of Euville-Lit mortar specimens, also a smooth coating is observed
512 on the surface of the mortar. This surface coating, ‘skin’ or ‘scum’ is known as laitance
513 and is formed when fine lime particles held in suspension migrate to the outer surface of
514 the wet material. This laitance is believed to hinder the vapour permeability of lime-
515 based materials and negatively impact upon the substrate beneath by causing
516 accelerated masonry decay associated with entrapment of moisture. Therefore, it is
517 recommended to remove this laitance from this type of restoration mortar, especially
518 when applied to permeable substrates [8].

519 The increase of capillary coefficients (C) of the stone in Euville-Lit and Euville-Alt
520 specimens after the salt crystallisation-ageing test (Table 3) could be explained by the
521 different capillarity behavior between the stone and the mortar. This is also related to
522 the different pore size distribution and connectivity of the pores of both materials. The
523 much lower C values of Lit and Alt mortars compared to the stone seems to have

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524 affected it by the retention and salt crystallisation in the pores of the stone. The pore
525 sizes and connectivity of pores of the stone have been modified after the test, affecting
526 so its capillarity behavior. In the case of Art mortar there are almost no modifications of
527 C parameter after the test suggesting a more similar hydric behavior between mortar and
528 stone. During the capillarity test, only Euville-Art mortars specimens show a continuous
529 water imbibition once water reach the mortar through the stone (its rate is almost
530 constant during the performance of the test). This can be explained by the high porosity
531 and pore size distribution of Art mortar (Fig.6). PSD is larger and more uniform in aged
532 samples than in the control samples. It might be that the pores have been partially filled
533 with salt crystals, increasing the amount of smaller pores and the generation of larger
534 pores caused by the salt crystallisation. In the other stone-mortar specimens the
535 imbibition is slowed down when water arrives to the mortars. Salt migration could be
536 promoted by the high porosity of the mortars. This together with capillary transfer
537 allows the evaporation of water and the crystallisation of salt in the stone or at the
538 interface cement– stone [19].

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539 There are almost no differences in compression and strength results, neither between the
540 two types of ageing tests nor between aged and control specimens. However, these
541 values are much higher in Euville-Alt mortar specimens than in the others. Compression
542 strength values are especially higher after control 1 test, suggesting that water
543 imbibition cycles have favoured carbonation and hydrolyses reactions of Alt mortar
544 giving rise a higher mechanical strength. Whereas salt crystallisation cycles provide
545 lower and similar compression strength results to those obtained in the specimens
546 subjected to the acid exposure test. The salt crystals inside the porosity that make the
547 strength to increase in a short-term can explain the slight higher values obtained in the
548 aged specimens. For Euville-Alt specimens the result is the opposite, since the

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549 crystallisation cycles have significantly reduced their compression strength. Some
550 authors found that the most relevant variable influencing the damage after durability test
551 of commercial ready-mix NHL mortars is the PSD [20]. A high amount of pores
552 concentrated in the range of fine diameters (below 0.01 μm) is generally associated to
553 major damage. In our case, Alt mortar display higher amount of pores bellow this size
554 compared to the other mortars. However, the lower porosity of salt aged Alt mortar
555 specimens compared to their control specimens (Table 4) could be due to salt crystals
556 filled the pores without breaking the structure. This may be related to its higher
557 hardness compared to the other mortars. It seems that the salts were not completely rid
558 out after washing the specimens at the end of the cycles, giving rise to lower porosity
559 values after the salt crystallisation test.

560 The acid test does not have a major incidence on the mechanical properties.
561 Experiments carried out in aerial and NHL mortars show that samples exposed to SO_2
562 give rise to gypsum formation that causes a strength drop, whereas an outstanding
563 strength increment is observed in hydraulic mortars [21]. Dissolution/precipitation
564 processes take place in the mortars after water imbibition/drying and salt crystallisation
565 cycles. The hydration of C2S (di-calcium silicates) produces CH (calcium hydroxide)
566 and some of new CH amount can also crystallize inside of the CSH (calcium silicate
567 hydrates) structure [22]. This fact has been related to the improvement of mortar
568 strength [23-25]. Besides, the addition of micro spherical calcium aluminosilicate
569 (CAS) particles (diameter of 50 μm) similar to fly ash is known to be effective in
570 forming a denser matrix leading to higher strength and better durability against sulphate
571 attack [26]. All these processes could contribute to the increase the strength of the
572 mortars, especially to compression of Alt mortar after water imbibition cycles.

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573 Euville-Art aged specimens have higher bonding strength values than their control
574 samples, which can be explained by their higher salt content. However, this was
575 determined in just one control specimen, which is not conclusive. The two other aged
576 stone-mortar specimens have lower bonding strength than the control specimens (Table
577 6). Euville-Alt mortar specimens have higher bonding strength values than the other
578 mortars. These specimens always break through the stone, while Euville-Lit mortar
579 specimens break through the mortar and Euville-Art specimens break through the
580 interface between mortar and stone. Therefore, Alt mortar has a better mechanical
581 interaction with the stone than the other mortars. Good adhesion (bond) between the
582 substrate and the repair mortar is commonly presented as one of the main measures of
583 success of repair and its long-term durability to assure the mechanical stability of the
584 façades. One important aspect is that the new mortar should not have a higher strength
585 than the stone. However, the water absorption and water vapour permeability of the
586 mortar must be of the same order of magnitude or greater than that of the other masonry
587 elements [27].

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588 Regarding to spectrophotometry results, the highest ΔE^* is produced between Alt
589 mortar and Euville stone after the accelerated ageing tests. This difference is even
590 higher in the control 1 specimens (ΔE^* 14) and it decreases in the salt aged specimens
591 (ΔE^* 9). According to suitability criteria used to assess conservation treatments, ΔE^*
592 values lower than 5 [28], and close to 3 [29,30] would not be visually detectable by the
593 human eye. This would not significantly affect the colourimetric parameters of the
594 substrate, but only Art mortar would fulfill this criterion after the accelerated ageing
595 tests. The increase in b^* parameter in all the stone-mortar specimens after the ageing
596 tests, means that colour turns to yellow. This especially affects the stone in contact with

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597 Lit and Alt mortars after the salt crystallisation test and the stone of Euville-Lit
598 specimens after the acid exposure test (Table 7 and Table 8).

599 We can say that Alt mortar is the most resistant mortar, with the best mechanical
600 interaction with Euville stone but with poor hydric compatibility. We can also point out
601 that Art mortar has a good hydric compatibility with the stone but a low mechanical
602 interaction being highly affected by the ageing tests.

603 This work is an essential step on the research concerning the compatibility and
604 durability of mortars and stone in masonry building restoration. In future research works
605 it should be necessary to work on the mineralogical composition and grain size
606 distribution together with the influence of different environmental curing conditions
607 which exert a great control on the pore structure and final physico-chemical properties
608 to assure a better hydric interaction with a specific type of stone.

609 **5. Conclusions**

610 Stone-mortar specimens were manufactured with Euville limestone and hydraulic
611 mortars (NHL, HL) containing Iarnite (di-calcium silicate, C2S) as the main hydraulic
612 component. After 28 days curing at similar environmental conditions of Paris region
613 these were exposed to accelerated ageing tests, salt crystallisation and acid vapours
614 exposure. Besides, additional control specimens were exposed to deionized water
615 imbibition cycles and water vapour exposure.

616 Regarding to chemical and mineralogical durability, the stone-mortar specimens
617 develop salt efflorescences after salt crystallisation (salt mixture solution) on those
618 mortars containing aerial lime (portlandite) in the anhydrous raw material (Artopierre
619 TM and Altar® Pierre commercial stone repair mortars, named as Art and Alt mortars,
620 respectively). Besides, those containing portlandite and phyllosilicates (Artopierre TM),
621 also develop a surface crust with cracking and detachment by ettringite crystals. Mortars

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622 with no portlandite, but with a high degree of larnite and phyllosilicates (chlorite and
623 talc) i.e. Lithomex Light mortar (named as Lit mortar) are not affected by salt
624 efflorescences, but powdering of the surface occurs after the acid vapour exposure test
625 (sulphurous and nitric acids). Large amounts of soluble nitrates and minor amounts of
626 sulphates precipitate on the surface of both stone and mortar materials after this test.
627 Whereas parallel fissures to the stone-mortar interface show up in specimens subjected
628 to the salt crystallisation test (salt mixture solution).
629 Regarding to physical durability, evaporation and water vapour permeability is slowed
630 down in all the salt aged specimens. The mortar with portlandite and vermiculite (Art
631 mortar) with higher porosity and quicker permeability and capillarity
632 absorption/evaporation coefficients, is the most affected by the ageing tests and hence
633 the less durable compared to the others. However, it displays a better hydric interaction
634 with the stone with a continuous imbibition once water through the stone reaches the
635 mortar.
636 Water imbibition cycles favor carbonation and hydrolyses reactions of Alt mortar (made
637 with portlandite and no phyllosilicates) giving rise to higher mechanical strengths.
638 However, these salt crystallisation cycles have reduced the compression and bonding
639 strength of this mortar which displays higher amount of pores bellow 0.01 μm ,
640 compared to the others. In spite of this, it displays higher strength values than the other
641 stone-mortar couples, with bonding break through the stone, while the others break
642 though the mortar or through the interface. A great change in colour takes place with a
643 remarkable yellowing of the stone, especially in the stone-mortar with chlorite and talc
644 specimens subjected to both aging tests. Only the stone-mortar specimens with
645 vermiculite (Art mortar) fulfill suitability conservation criteria, with a total colour
646 difference not visually detectable by the human eye.

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647 Even though the mortar Alt mortar that also has 50 μm spherical calcium
648 aluminosilicate particles is the most resistant mortar with the best mechanical
649 interaction with Euville stone, its worst hydric compatibility could damage the stone in
650 a longer term. Art mortar has a good hydric behavior but poor mechanical interaction
651 with the stone, being highly affected by salt crystallisation and acid vapours exposure
652 which would act more as a sacrificial mortar rather than s tone-repair mortar.
653 The physico-chemical composition and curing conditions play an important role in the
654 final pore system and hardness of these mortars. The hydric and mechanical properties
655 condition their durability also affected by the environmental conditions and other
656 current anthropogenic factors, especially remarkable in historical and touristic cities.
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748 **FIGURE CAPTIONS**

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2 749 **Fig.1.** a) Euville stone-mortar specimens (top: *Lit* mortar-stone; center: *Alt* mortar-stone;
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4 750 bottom: *Art* mortar-stone); b) Stone-mortar specimens: control (to the right) and aged by salt
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6 751 crystallisation accelerated test (to the left); top: *Lit*-stone specimens; center: *Alt*-stone
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8 752 specimens; bottom: *Art* mortar-stone specimens.

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11 753 **Fig. 2.** Thin section images of Euville stone-Lit mortar specimens, with porosity in blue colour:
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13 754 a) Euville in the bottom and Lit mortar in the top (control 2 specimen) showing the aggregates
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15 755 and rounded porosity; b) same former image in cross nicols, c) Lit mortar (aged 1 specimen)
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17 756 displaying a fissure close to the interface; d) same former image in crossed nicols.

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21 757 **Fig. 3.** Thin section images of Euville stone-Art mortar specimens, with porosity in blue colour:
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23 758 a) Euville in the bottom and Art mortar in the top (control 1 specimen) with a fissure along the
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25 759 interface; b) same former image in cross nicols; c) Art mortar surface (on the top) of aged 1
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27 760 specimen showing aggregates with signs of salt weathering; d) same former image in crossed
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29 761 nicols.

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32 762 **Fig. 4.** Thin section images Euville stone-Alt mortar specimens, with porosity in blue colour. a)
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34 763 Euville in the bottom and Alt mortar in the top (aged 1 specimen) with almost no porosity; b)
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36 764 same former image in cross nicols; c) Alt mortar surface (on the top) of aged 2 specimen with
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38 765 salt crystallisation in the edges of the pores; d) same former image in crossed nicols.

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41 766 **Fig. 5.** SEM images of salt crystals after accelerated ageing tests performed on the stone-mortar
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43 767 specimens. a) sodium chloride (NaCl) crystals on the surface of Art mortar after salt
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45 768 crystallisation test; b) acicular crystals of ettringite crystals on the surface crust of Art mortar
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47 769 after salt crystallisation; c) calcium sulphate (CaSO₄) crystals and calcium aluminosilicate
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49 770 (CAS) spherical particles after acid exposure test of Alt mortar; d) sodium sulphate (Na₂SO₄)
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51 771 crystals, with some calcium and potassium, on the surface of Alt mortar after salt crystallisation
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53 772 test.

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57 773 **Fig. 6.** Connected porosity and pore size distribution (PSD) obtained by mercury intrusion
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59 774 porosimetry in control 1 and aged 1 specimens of: (a) Euville in contact with Lit mortar (from
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775 Euville-Lit specimen); b) Lit mortar (from Euville-Lit specimen); c) Euville in contact with Art
 776 mortar (from Euville-Art specimen); d) Art mortar (from Euville-Art specimen); e) Euville in
 777 contact with Alt mortar (from Euville-Alt specimen); f) Alt mortar (from Euville-Alt specimen).
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 779 **Table 1.** Ion chromatography results (anions) carried out in the mortar (Lit, Art and Alt) and in
 780 stone (Euville, EU) in contact to these mortars of each couple of stone-mortar specimens after
 781 salt crystallisation ageing test (ageing 1).

Soluble salts		Anions (ppm)			Wt (%)
Specimen	Sample	Cl ⁻	SO ₄ ²⁻	NO ₃ ⁻	
Lit-Euville	Lit-ageing 1	9.5	39.9	0.9	0.3
	EU-Lit-ageing 1	8.1	12.5	0	0.1
Art-Euville	Art- ageing 1	41.1	84.3	46.3	1.1
	EU-AR- ageing 1	7.6	10.9	0.0	0.1
Alt-Euville	Alt- ageing 1	31.5	11.4	33.7	0.5
	EU-AL- ageing 1	7.2	12.6	0	0.1

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 783 **Table 2.** Ion chromatography results (anions) performed in the mortar (Lit, Art and Alt) and
 784 stone (Euville, EU) of each couple of stone-mortar specimens after acid ageing test (ageing 2).

Soluble salts		Anions (ppm)			Wt (%)
Specimen	Sample	Cl ⁻	SO ₄ ²⁻	NO ₃ ⁻	
Lit-Euville	Lit-ageing 2	6.1	55.3	128.4	1.4
	EU-Lit- ageing 2	6.1	25.5	181.4	1.5
Art-Euville	Art- ageing 2	6.4	198.1	487.7	4.7
	EU-AR- ageing 2	5.8	46.5	145.3	1.3

Alt-Euville	Alt- ageing 2	5.6	23.2	161.2	1.2
	EU-AL- ageing 2	6.0	108.0	81.9	1.3

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787 **Table 3.** Hydric results of stone-mortar specimens after control 1 and ageing 1

	Specimens	Euville-Lit	Euville-Art	Euville-Alt
Capillary coefficient (g/m ² .s ^{0.5}) (first part)	Control	80.28	40.45	82.09
	Ageing 1	92.90	39.86	102.87
Capillary coefficient (g/m ² .s ^{0.5}) (second part)	Control	1.81	10.83	5.55
	Ageing 1	3.08	11.96	4.42
Desorption coefficient (g/m ² .s ^{0.5})	Control	-5.32	-9.42	-5.88
	Ageing 1	-4.97	-7.11	-5.55
Water vapour permeability (Kg/ (m ² .s))	Control	4.77E-06	1.59E-05	7.95E-07
	Ageing 1	4.77E-06	3.97E-06	3.18E-07

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789 **Table 4.** Results from mercury intrusion porosimetry analyses obtained in both mortar and
790 stone samples from each couple of stone-mortar specimen (control 1 and ageing 1).

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	Specimens	Euville-Lit		Euville-Art		Euville-Alt	
		Euville	Lit	Euville	Art	Euville	Alt
Apparent density (g/cm ³)	Control 1	2.17	1.74	2.17	1.63	2.17	1.94
	Ageing 1	2.23	1.77	2.13	1.65	2.17	1.99
Bulk density (g/cm ³)	Control 1	2.55	2.34	2.55	2.14	2.55	2.41
	Ageing 1	2.59	2.41	2.59	2.28	2.61	2.36
Hg-porosity (%)	Control 1	14.91	25.39	14.91	23.90	14.91	19.21

	Ageing 1	13.75	26.25	17.83	27.50	16.91	15.71
Mode pore radius (μm)	Control	0.60	1.34	0.60	0.48	0.60	0.07
	Ageing 1	0.43	1.50	0.85	0.28	0.60	0.03
Median pore radius (μm)	Control	0.82	1.15	0.82	0.47	0.82	0.07
	Ageing 1	0.58	1.16	1.32	0.72	0.96	0.03

792 **Table 5.** Flexural and compression strength of stone-mortar specimens after the ageing tests.

Test	Euville-Lit		Euville-Art		Euville-Alt	
	Flexural (MPa)	Compression (MPa)	Flexural (MPa)	Compression (MPa)	Flexural (MPa)	Compression (MPa)
Control 1	3.59 ± 0.81	< 7	4.03 ± 0.54	$8.87 \pm 0.52^*$	7.04 ± 3.40	17.02 ± 2.29
Ageing 1	3.99 ± 0.63	< 7	3.91 ± 1.23	10.02 ± 1.18	9.32 ± 0.15	11.70 ± 0.67
Control 2	3.22 ± 1.25	$9.30 \pm 0.36^*$	1.58^{**}	< 7	5.26 ± 0.13	10.35 ± 1.57
Ageing 2	3.16 ± 1.86	9.99 ± 0.62	2.78^{**}	< 7	5.68 ± 0.52	12.06 ± 2.94

793 * Only two specimens, in the other specimens Compression was <7; ** Only one specimen

794 **Table 6.** Results obtained from the adherence test in the stone-mortar specimens

Specimens	Test	Bonding strength (KPa)	Breaking
Lit-Euville	Control 1	814.87 ± 72.03	Mortar
	Ageing 1	738.47 ± 108.04	Mortar
	Control 2	916.73 ± 72.03	Mortar**
	Ageing 2	763.94 ± 360.13	Interface**
Art-Euville	Control 1	254.65^*	Interface
	Ageing 1	713.01 ± 72.03	Interface
	Control 2	N/P	N/P
	Ageing 2	N/P	N/P
Alt-Euville	Control 1	1324.17 ± 216.08	Stone

	Ageing 1	1158.65 ± 90.03	Stone
	Control 2	967.66 ± 144.05	Stone
	Ageing 2	993.13 ± 252.09	Stone

795 * Only one specimen; ** one break through the mortar or through interface and the other through
796 the stone; N/P Not performed.

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799 **Table 7.** Colour parameters (L*, lightness; a* and b* colour coordinates; C*, Chroma; ΔE*, total colour difference)
800 of the surface of stone and mortar on the stone-mortar specimens (control 1 (C1), exposed to water imbibition, and
801 aged 1 (A1), exposed to salt crystallisation test.

Material	L*	a*	b*	C* (D65)	Hue (D65)	ΔE*
L-C1	80.33 ± 0.48	2.00 ± 0.07	10.20 ± 0.42	10.39 ± 0.42	78.92 ± 0.18	2.90
Eu-L_C1	83.00 ± 1.87	2.75 ± 0.83	11.08 ± 1.55	11.43 ± 1.67	76.24 ± 2.73	
L-A1	80.37 ± 0.69	1.91 ± 0.11	9.78 ± 0.24	9.96 ± 0.24	78.95 ± 0.57	7.30
Eu-L-A1	78.73± 2.72	4.10± 0.62	16.55 ± 1.18	17.06 ± 1.27	76.16 ± 1.28	
Art-C1	88.33 ± 0.46	0.54 ± 0.09	7.02 ± 0.31	7.04 ± 0.30	85.58 ± 0.97	6.28
Eu-Ar-C1	82.61 ± 1.45	2.32 ± 0.80	8.91 ± 1.06	9.22 ± 1.21	75.78 ± 3.42	
Art-A1	83.09 ± 0.63	1.62 ± 0.24	11.46 ± 0.15	11.57 ± 0.18	81.99 ± 1.11	1.85
Eu-Art-A1	82.20 ± 1.35	3.02 ± 0.52	12.28 ± 1.72	12.65 ± 1.79	76.27 ± 0.59	
Alt-C1	69.05± 1.18	2.25 ± 0.20	10.08± 0.34	10.33 ± 0.37	77.41 ± 0.91	14.15
Eu-Alt-C1	83.15± 1.34	2.32 ± 0.48	8.91± 0.60	9.21 ± 0.71	75.51 ± 1.86	
Alt-A1	72.77± 0.40	1.95 ± 0.09	9.96± 0.03	10.15 ± 0.03	78.94 ± 0.54	8.99
Eu-Alt-A1	80.97 ± 1.58	3.41 ± 0.22	13.35 ± 0.45	13.78± 0.41	75.64 ± 1.20	

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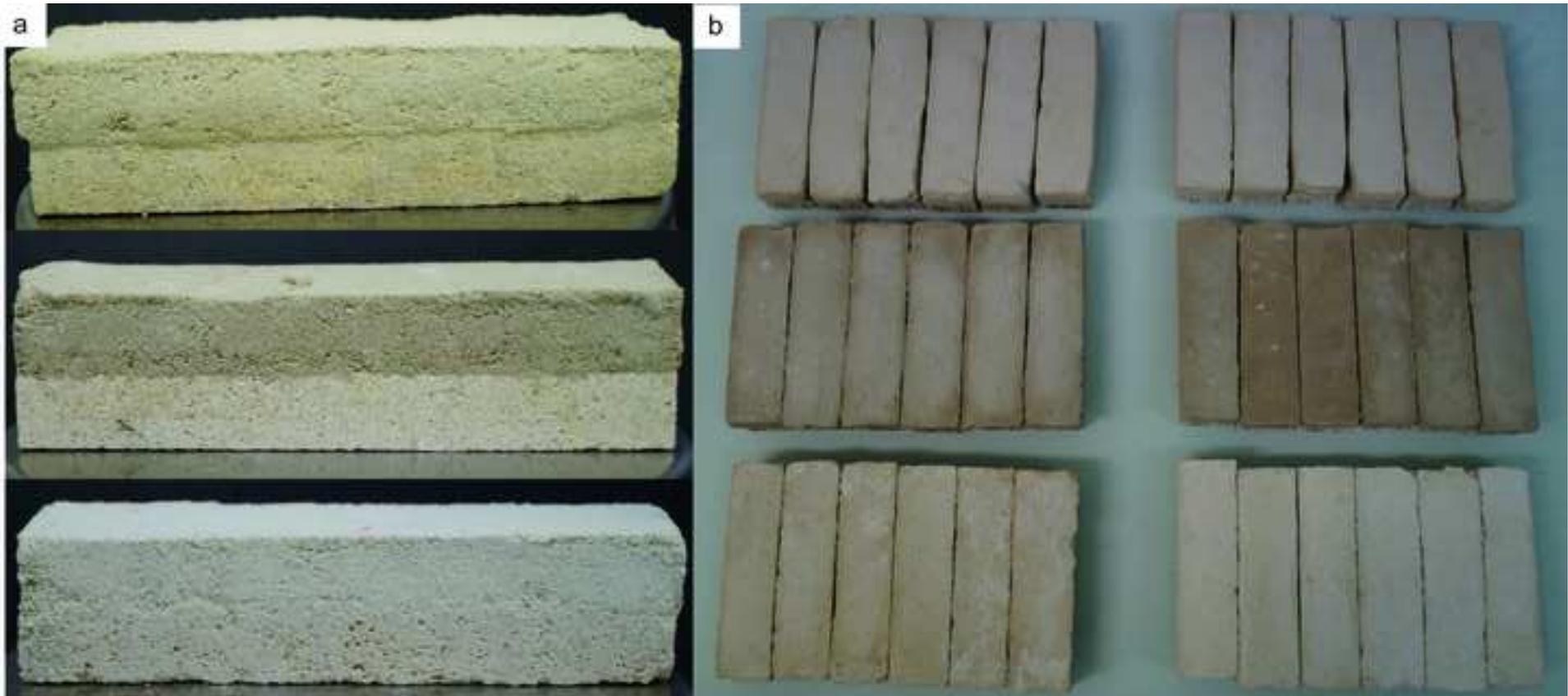
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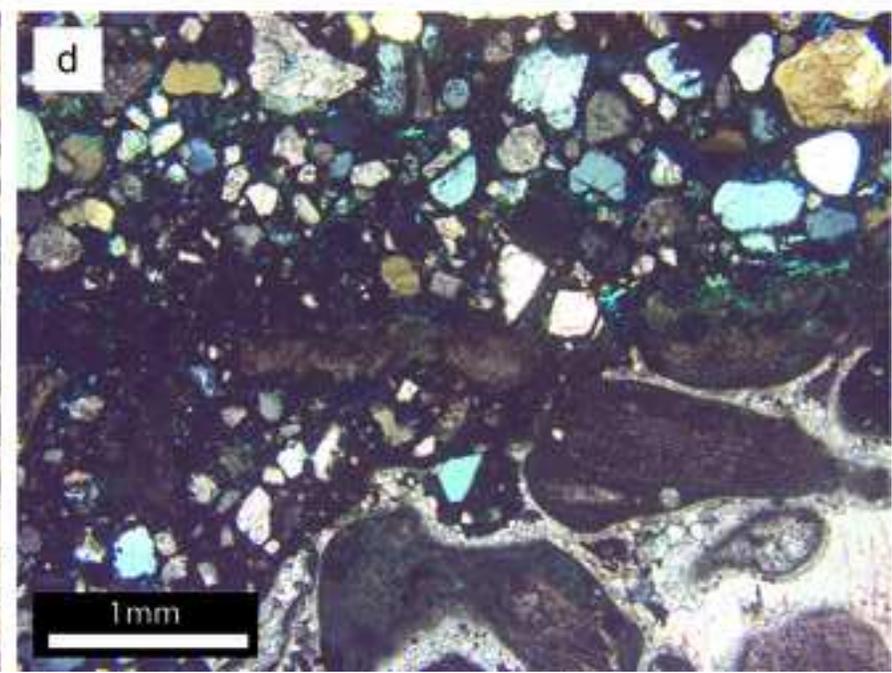
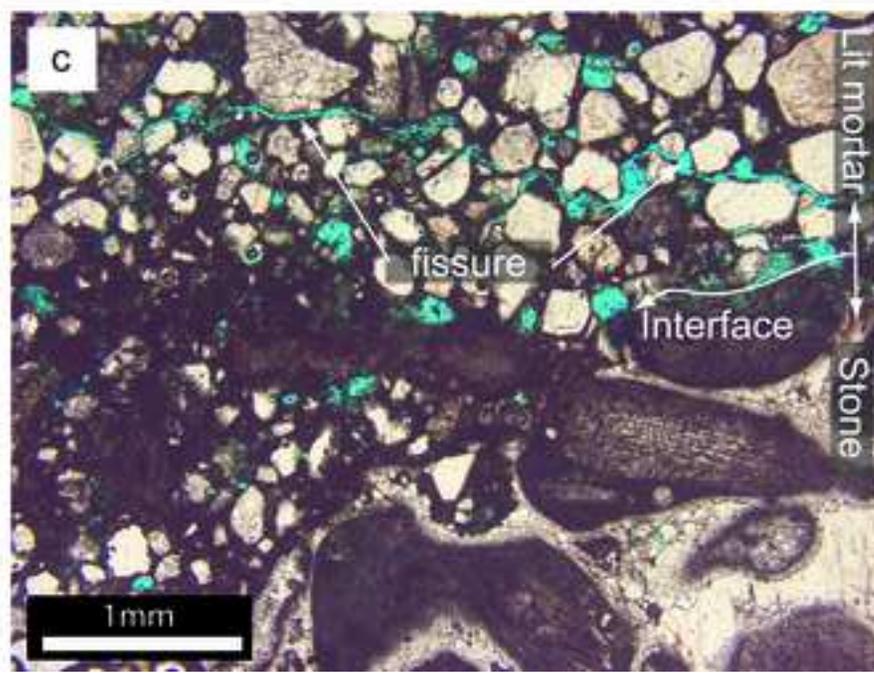
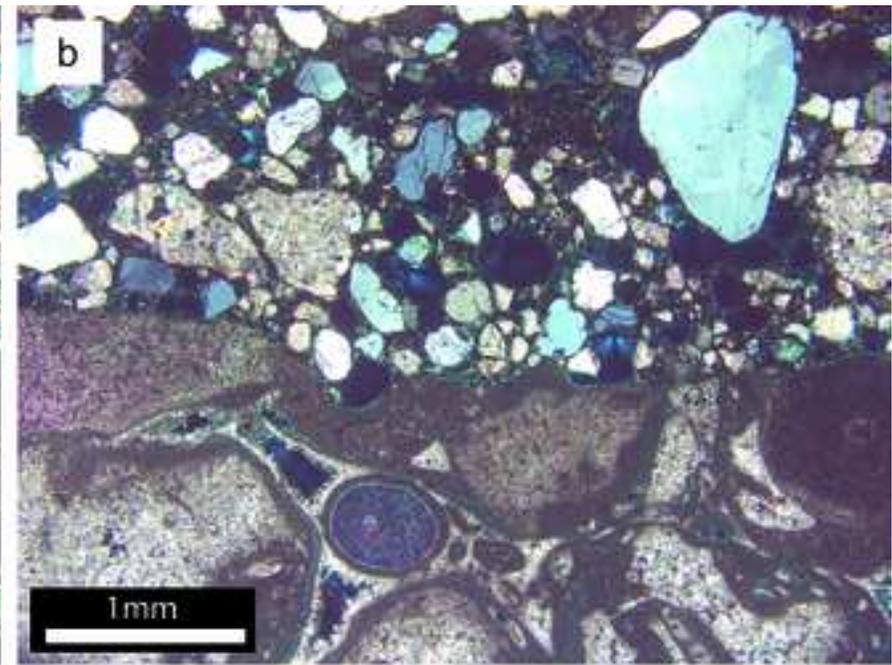
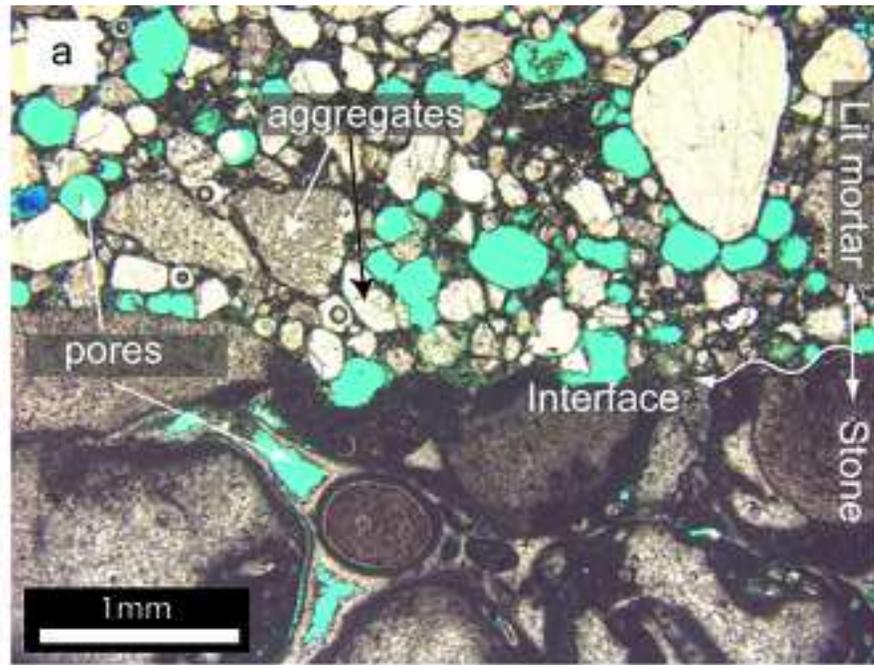
810 **Table 8.** Colour parameters (L*, lightness; a* and b* colour coordinates; C*, Chroma; ΔE*, total colour difference)
 811 of the stone-mortar specimens (control 2 (C2), exposed to water vapour, and aging 2 (A2) exposed to acid vapours
 812 exposure).

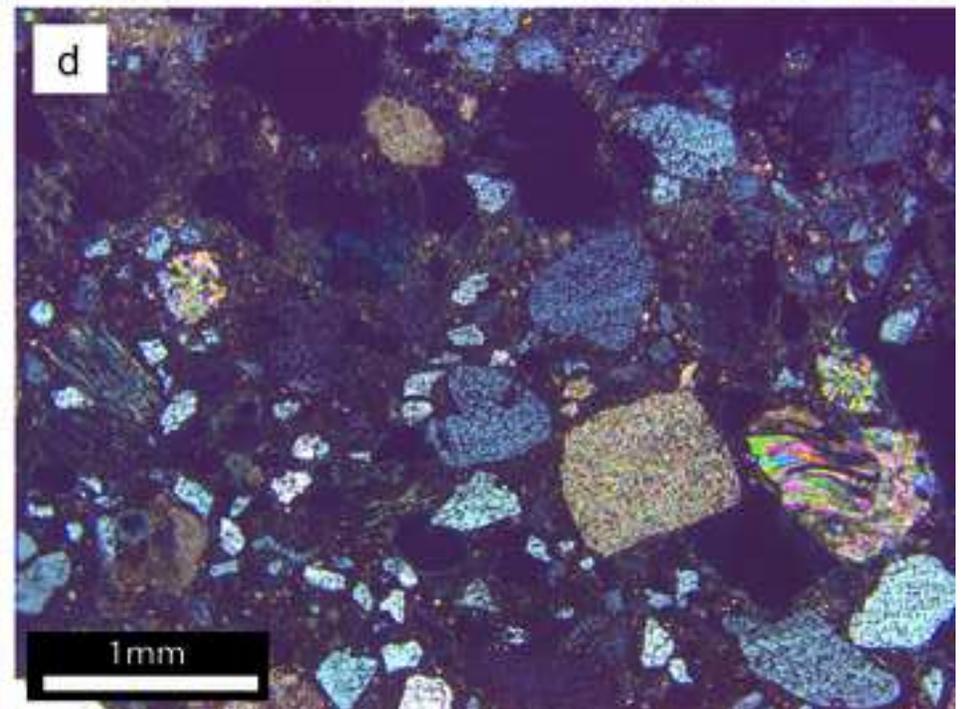
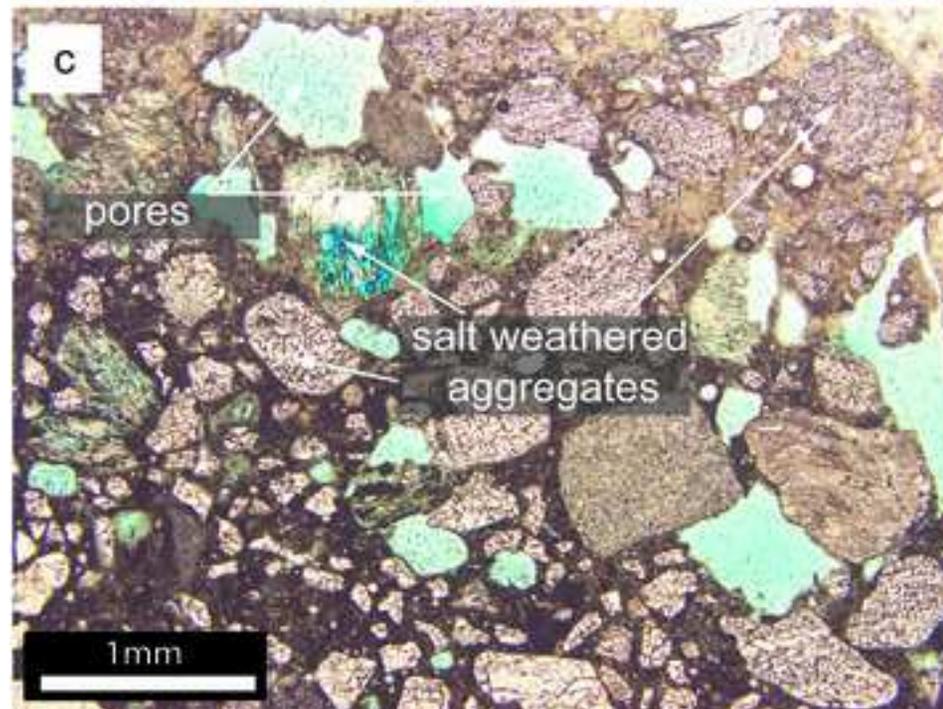
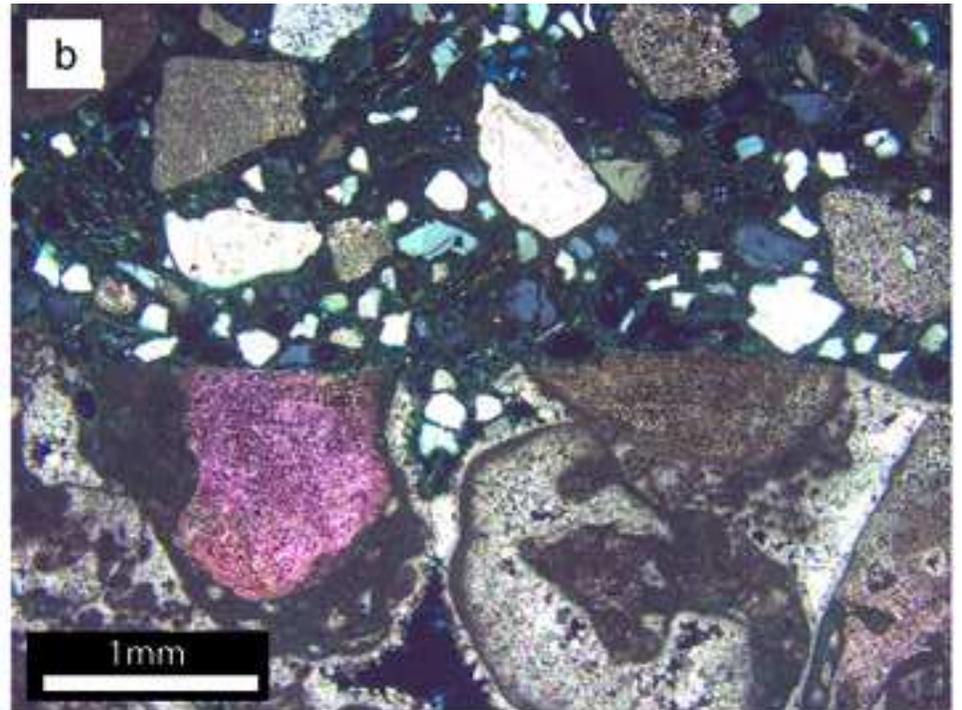
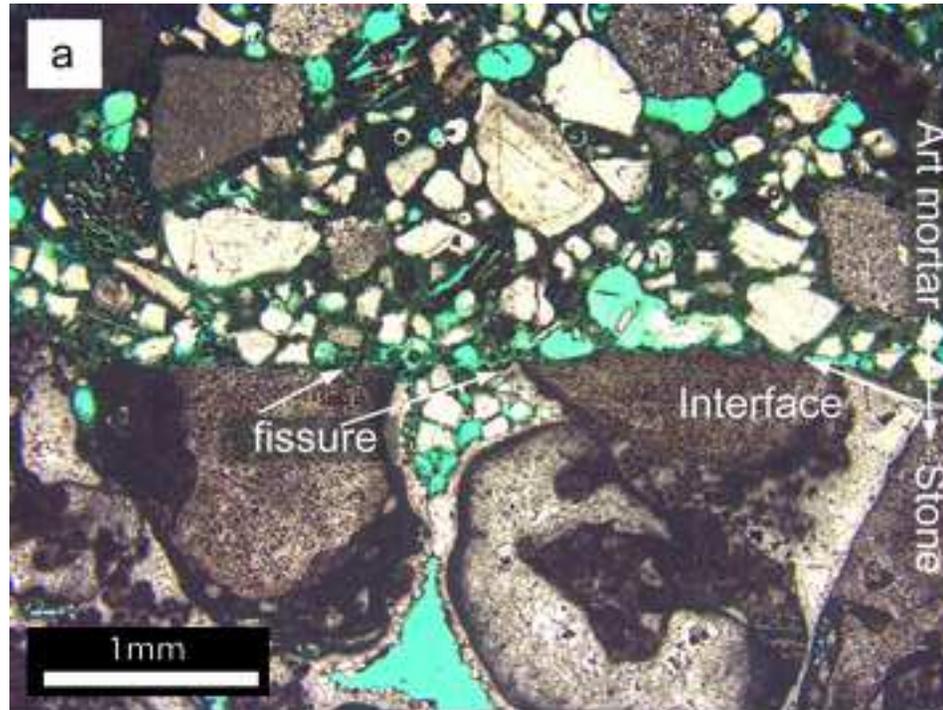
Material	L*	a*	b*	C* (D65)	Hue (D65)	ΔE*
L-C2	79.50 ± 0.12	2.31 ± 0.04	11.23 ± 0.27	11.47 ± 0.27	78.38 ± 0.14	5.93
Eu-L-C2	84.46 ± 2.26	2.02 ± 0.59	7.98 ± 1.81	8.24 ± 1.90	75.93 ± 0.89	
L-A2	72.72 ± 0.05	3.52 ± 0.03	17.58 ± 0.07	17.93 ± 0.06	78.68 ± 0.14	7.58
Eu-L-A2	78.95 ± 3.71	3.41 ± 0.84	13.26 ± 2.28	13.69 ± 2.42	75.73 ± 0.99	
Art-C2	88.04 ± 0.71	0.81 ± 0.06	6.37 ± 0.53	6.43 ± 0.53	82.76 ± 0.15	5.02
Eu-Art-C2	83.79 ± 0.78	1.98 ± 0.10	8.78 ± 0.97	9.00 ± 0.95	77.21 ± 1.14	
Art-A2	85.66 ± 0.66	1.41 ± 0.10	8.98 ± 0.10	9.09 ± 0.12	81.08 ± 0.49	5.73
Eu-Art-A2	80.52 ± 0.70	2.80 ± 0.10	11.10 ± 0.49	11.45 ± 0.49	75.84 ± 0.13	
Alt-C2	75.64 ± 0.08	1.94 ± 0.06	10.39 ± 0.26	10.57 ± 0.27	79.44 ± 0.08	7.88
Eu-Alt-C2	83.50 ± 2.12	2.22 ± 0.24	10.03 ± 0.96	10.27 ± 0.97	77.50 ± 1.17	
Alt-A2	73.63 ± 0.92	2.15 ± 0.29	11.81 ± 1.23	12.01 ± 1.26	79.73 ± 0.32	7.48
Eu-Alt-A2	81.08 ± 0.37	2.65 ± 0.36	11.89 ± 0.52	12.18 ± 0.57	77.48 ± 1.29	

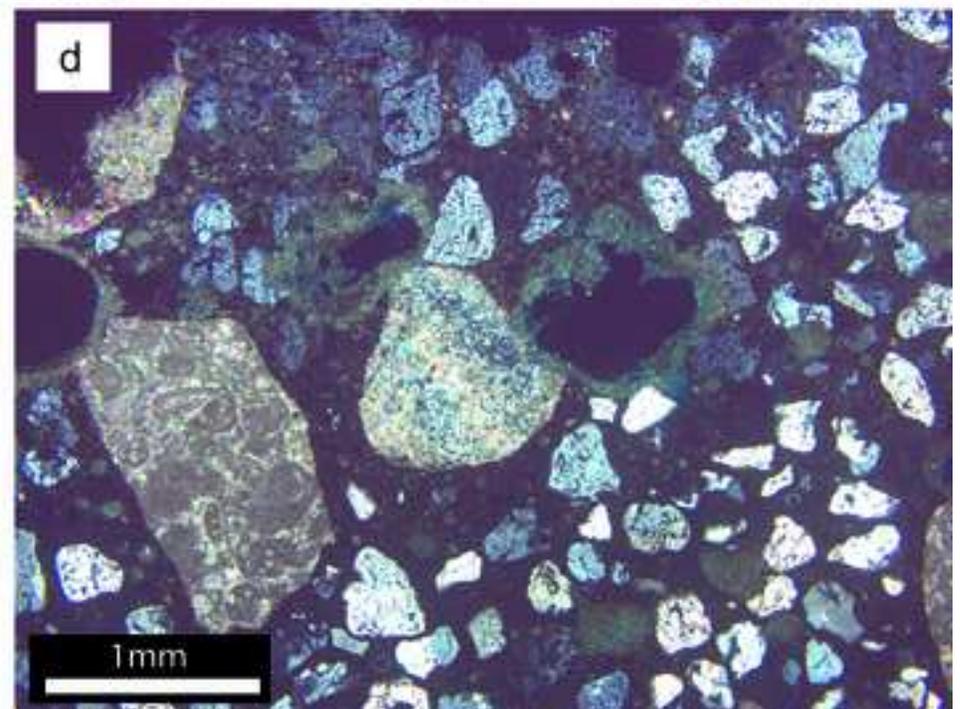
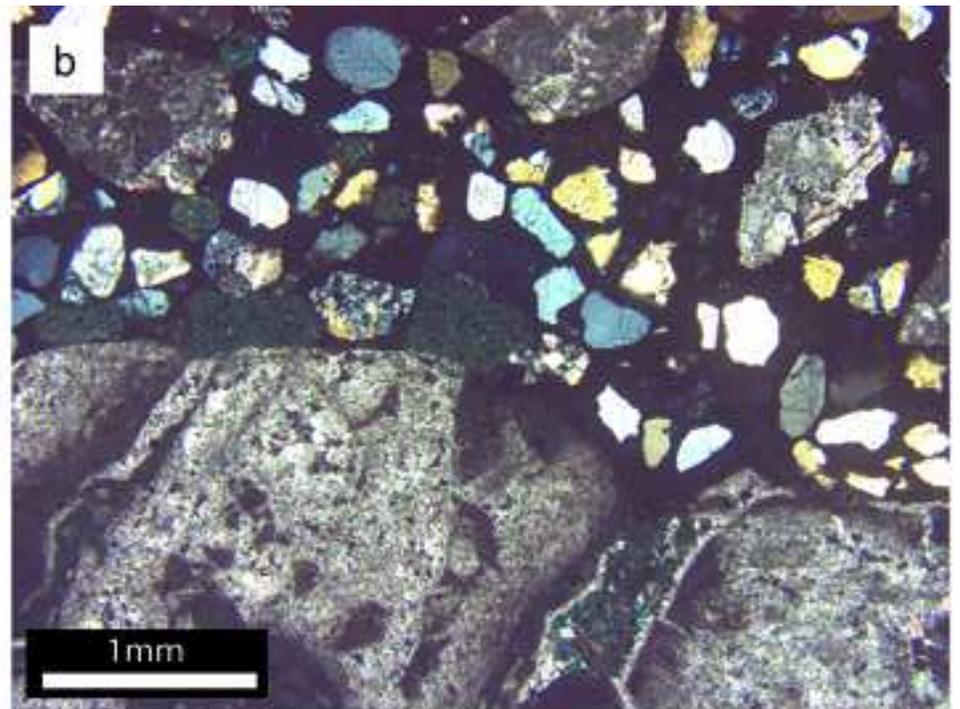
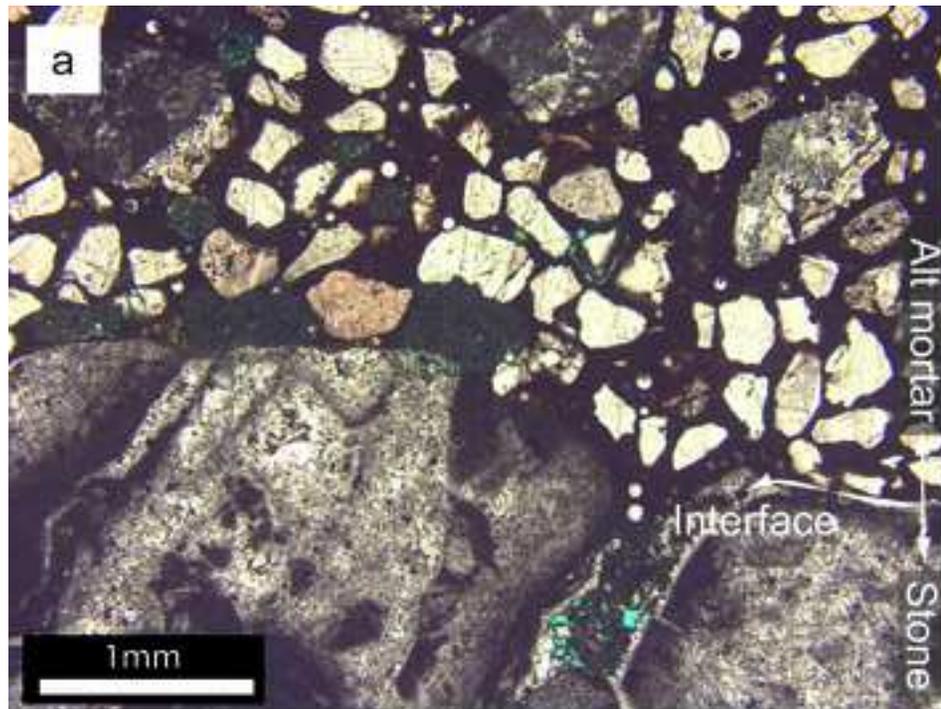
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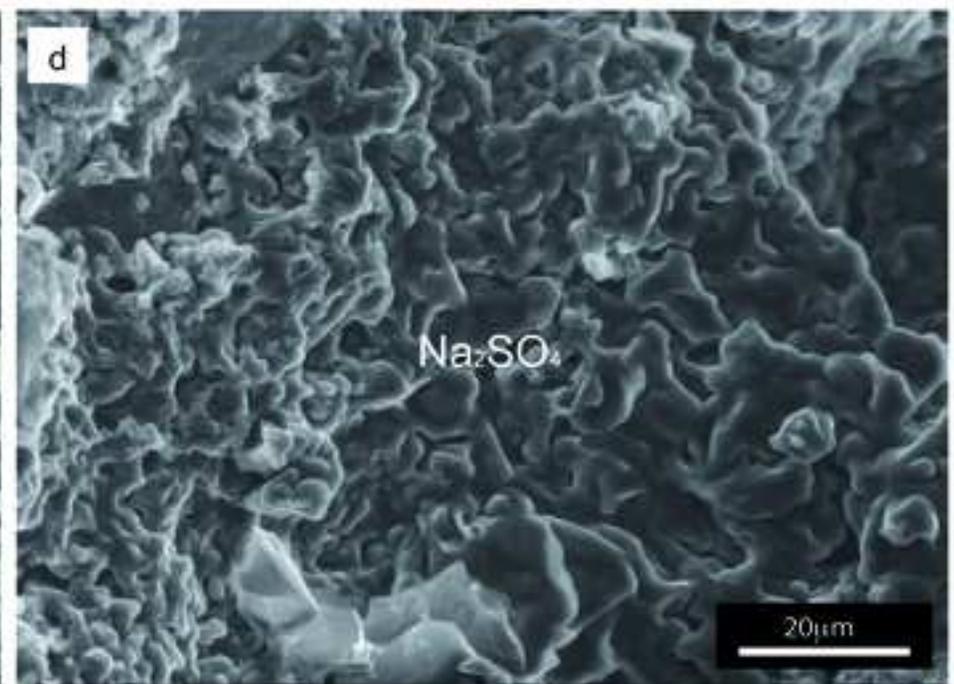
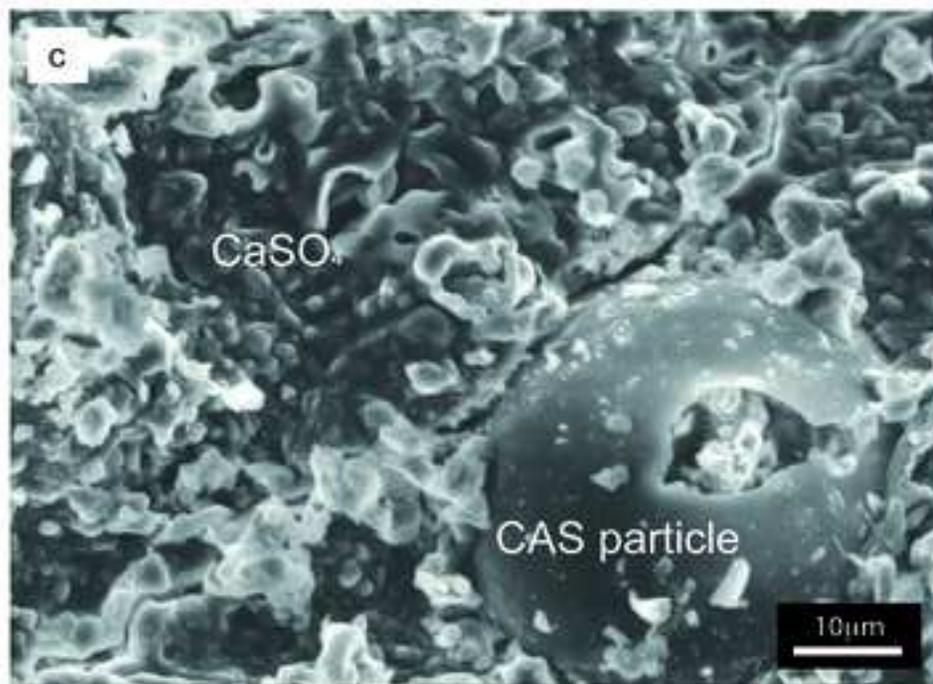
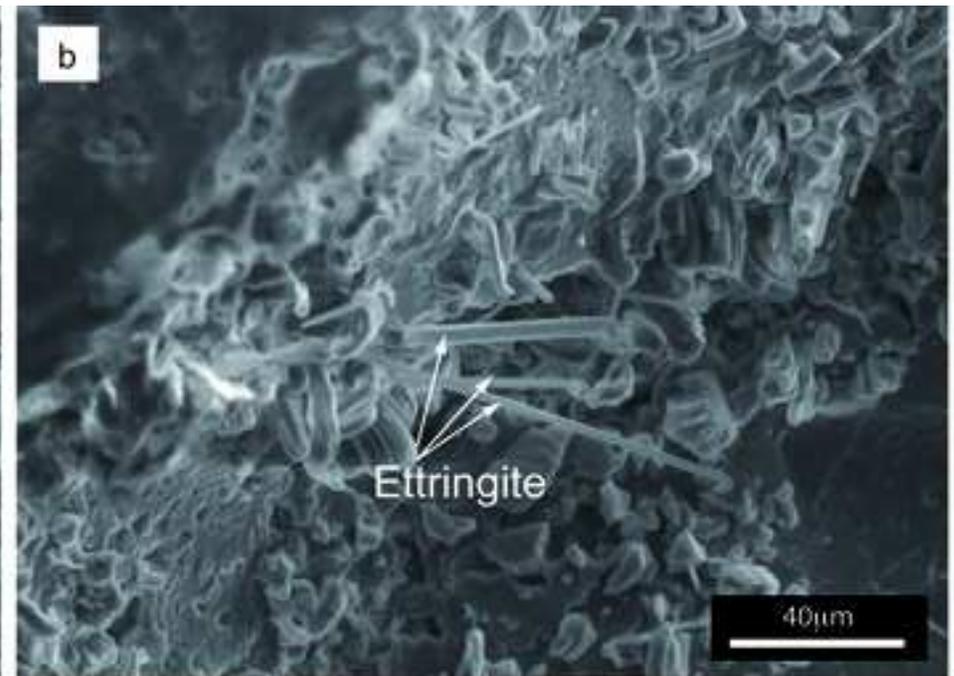
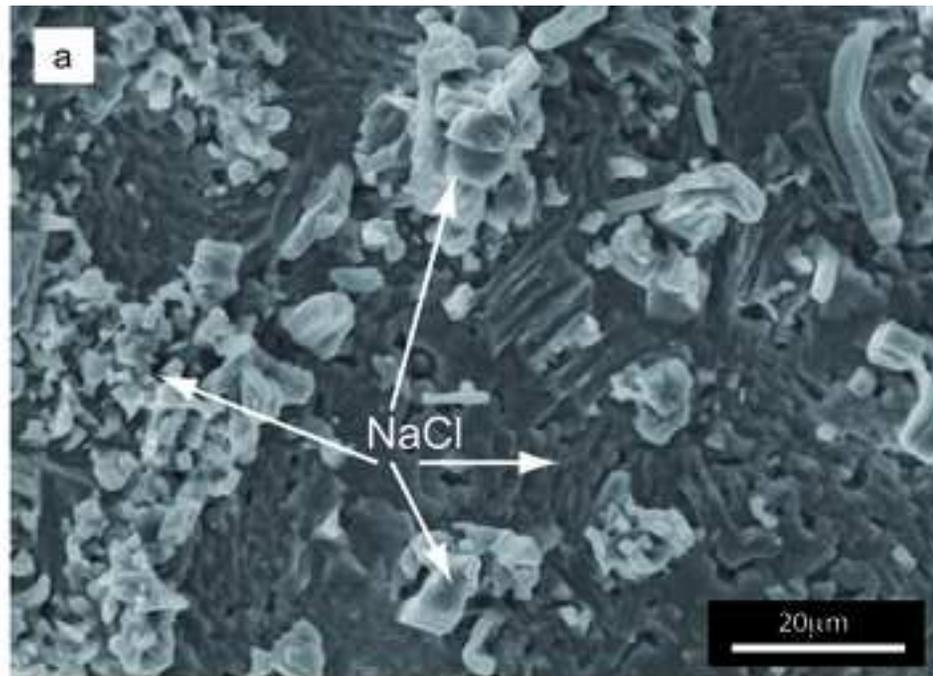
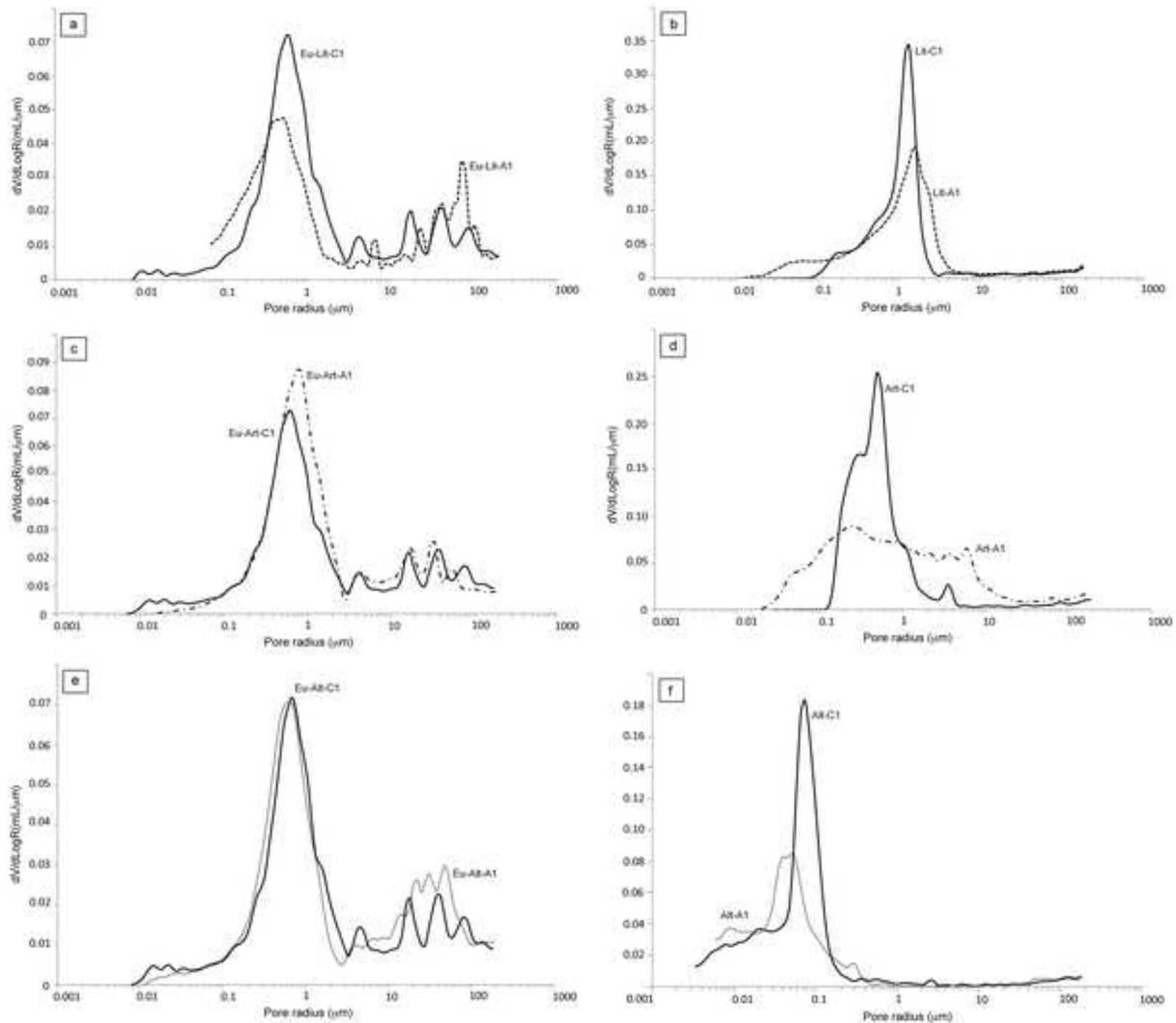


Fig.6



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