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Chemo-mechanical behaviour of non-expansive clays accounting for salinity effects

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2	Chemo-mechanical behaviour of non-expansive clays
3	accounting for salinity effects
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28 Abstract

29 Changes in the chemistry of the pore fluid are known to impact on the hydro-mechanical 30 behaviour of clays. Experimental evidence collected in the last decades led to the formulation of 31 constitutive chemo-mechanical models for expansive soils used in engineering practice for the 32 containment of pollution, such as bentonite. Less attention has been paid to modelling the 33 chemo-mechanical behaviour of non-expansive clays, less frequently used for geoenvironmental 34 applications but equally exposed to chemical changes.

35 First key differences between the impact of salinity on the fabric of expansive and non-expansive 36 clays are pointed out. At the macroscopic scale, an increase in salinity causes a small translation 37 of the Normal Compression Line of non-expansive clays to higher void ratios, which in some cases 38 is also accompanied by an increase in compressibility. The opposite occurs for expansive clays. 39 These experimental evidences provide the basis for a chemo-mechanical model formulated in 40 the frame of elasto-plasticity with generalised hardening, whose yield surface expands with pore 41 fluid concentration. The model is validated against experimental results, both original and from 42 the literature. Simulation results compare very well with those of tests performed on 43 reconstituted, compacted and intact samples.

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⁴⁶ **1** - Introduction

Pore fluid chemistry is well known to influence the hydraulic and mechanical behaviour of clays (see e.g. the comprehensive summary in Sridharan, 1991). This influence is due to the effects that the pore fluid composition has on superficial forces, such as van der Waals and electrostatic interactions, which act between close particles (see e.g. van Olphen, 1977, Santamarina et al., 2001). van der Waals interaction is always attractive and mainly depends on the dielectric constant of the pore fluid, while it is practically independent on the type and concentration of dissolved ions.

54 Electrostatic forces are caused by the electrical charge of the particle surface, and are mediated 55 by the Diffuse Double Layer (DDL) of ions of opposite charge with respect to the one of the 56 particle surface. As the solute concentration increases, the thickness of the DDL decreases and 57 so do electrostatic forces. Particles of most clay minerals have a platy thin shape with large faces 58 and thin edges. The electrical charge on the particles face is mainly caused by the isomorphous 59 substitution in the crystal lattice of higher valence metals with lower valence metals (e.g. Mitchell 60 and Soga, 2005): it is negative and not very influenced by pore water composition. The electrical 61 charge on the particle edge is caused by the interruption of the crystal lattice, which exposes 62 metals and hydroxyl groups to the surface: it is usually positive (van Olphen, 1977), and it can be 63 influenced by the composition of the pore water because of the adsorption/desorption of 64 protons and other ions (Santamarina et al., 2001). This is particularly relevant for kaolinite: as the 65 pH grows the positive edge charge decreases and becomes negative at high pHs (e.g. 66 Sposito 1984).

67 The interplay between van der Waals attractive forces and electrostatic forces (which are 68 repulsive between particles faces, attractive between edges and faces) determines the type of 69 association of the particles in suspensions and the fabric imparted through sedimentation. 70 According to Santamarina et al. (2002), the ratio between the length and the thickness of the 71 particles determines the relevance of the different forces. Particles of expansive clays, such as 72 montmorillonite, are extremely elongated and thin. Edge charges have then very little relevance 73 on their interaction and in suspensions they tend dispose according to a face to face association 74 (i.e. they aggregate). This association dominates their fabric and as at high ionic concentrations 75 more cations are available to shield the face negative charge the thickness of the double layer 76 and the size of the pores in the aggregates reduces.

77 Particles of non-expansive clays, such as illite and kaolinite, have a smaller length to thickness 78 ratio (i.e. they are relatively thicker) and attraction between particles edges and faces, or even 79 between edges, can be relevant when they are in suspensions. The term flocculation is used to 80 describe single particles, or groups of aggregated particles, associated according to a face to edge 81 or edge to edge scheme. The association of kaolinite particles is ruled both by pH and ionic 82 concentration (Palomino & Santamarina, 2005). Aggregation takes place above a threshold ionic 83 concentration (NaCl molarity of the order of 10⁻¹ M): edge to face flocculation of aggregates 84 occurs at both very low and very high pHs, while it has not been observed for pHs ranging 85 between 5 and 7. Below the threshold ionic concentration aggregation occurs without 86 flocculation when the pH is smaller than 3, while edge to face flocculation occurs at higher pHs. 87 Not much attention has been paid to the role of pH for illite. As for the role of pore fluid salinity, 88 the experience gathered for Norwegian illitic quick clays suggests that sedimentation in brackish

or marine water promotes edge to edge flocculation (i.e. a cardhouse type of fabric), which
collapses after exposure to fresh water (see e.g. Rosenqvist, 1966). The same evidence has been
reported for non-expansive smectites from Japan (Ariake clay, Ohtsubo et al., 1985).

92 The fabric of a given clay soil depends on the relative magnitude of superficial, body and 93 mechanical forces when it is formed. While surface forces dominate on body and mechanical 94 forces for suspensions and slurries, the opposite holds for compacted soils. The relevance of the 95 soil formation stage, and in general of fabric, on the soil hydro-mechanical response has been 96 clearly highlighted by Collins and Mc Gown (1974) according to which, although the fabric of soils 97 "cannot be expected to be determined simply on basis of the depositional history and 98 environment, it has been found that there may be a dominant feature or set of feature present in 99 any one soil. This predominant microfabric can induce certain types of engineering behaviors, 100 such as sensitivity, collapse and expansion".

101 The impact of fabric on different aspects of the clay behaviour, and particularly on those related 102 to the pore fluid chemistry imposed during preparation of reconstituted or compacted 103 specimens, has been documented in many experimental works. The one-dimensional 104 compression curves of samples reconsolidated from slurry have been found to depend on the 105 pore fluid of preparation for kaolinite and bentonite saturated with fluids with different dielectric 106 constant (Sridharan & Rao, 1973) and smectitic soils saturated with brines (Barbour & Yang, 1993, 107 Di Maio, 1996). Barbour & Yang (1993) further remark that adding brine to montmorillonite 108 material prior to static compaction in unsaturated conditions causes the moisture-density 109 relationship to be altered due to physicochemical effects, which leads to compression curves 110 which depend both on the preparation procedure and on the pore fluid chemistry.

111 Modifications of pore fluid chemistry are also possible due to pore fluid replacement or of 112 diffusion of the solutes, and pore water chemistry is expected to change with time in many earth 113 structures devoted to pollution containment. For example, in compacted clay barriers for nuclear 114 waste storage, chemical changes can occur due to the inflow of water from the host rock and 115 because of the evaporation triggered by temperature increase induced by radioactive reactions. 116 In both cases, a variation of the concentration of the species dissolved in water is anticipated. In 117 clay barriers for municipal waste disposal, the pore fluid chemical composition changes with the 118 transport of domestic leachate, while cutoff walls encapsulating polluted sites can be 119 progressively permeated by the contaminants spilled underground. Pore fluid chemistry also 120 varies in natural environments, e.g. in clay formations that were deposited in marine 121 environments that emerged in relatively recent geological times. After emersion, salt diffusion 122 towards the boundaries of the formation or entrance of fresh water (i.e. leaching) are 123 anticipated.

124 As changes in the pore fluid chemistry modify the interaction between particles, deformations 125 even at constant total stress and impact on the hydro-mechanical properties are expected. 126 Volume change caused by pore fluid replacement under a constant vertical stress and their 127 effects on the following compression behaviour were studied among others by Sridharan & Rao 128 (1973), Torrance (1974), Di Maio (1996), Musso et al., (2003, 2013). Torrance (1974) leached with 129 distilled water undisturbed samples of Norwegian quick clays, originally filled with marine water: 130 this process induced shrinkage in normally consolidated samples and swelling of highly over-131 consolidated samples.

132	To account for these effects, elastic-plastic chemo-hydro-mechanical models (e.g. Hueckel, 1997;
133	Loret et al., 2002; Gajo & Loret, 2003; Liu et al., 2005; Guimaraes et al., 2013, Witteveen et al.,
134	2013, Della Vecchia & Musso, 2016, Yan, 2018, Della Vecchia et al, 2019) were formulated in the
135	paste decades Limited efforts have been devoted to modeling the behaviour of non-expansive
136	clays, since excluding Witteveen et al. (2013) all of these works make specific reference to very
137	expansive clays (bentonites, mostly relying on the experimental data from the seminal paper of
138	Di Maio, 1996).
139	This work aims at formulating a simple phenomenological elasto-plastic model for the chemo-
140	mechanical behaviour of non-expansive illitic and smectitic clays. First, the different effects of
141	salinity on the behaviour of expansive and non-expansive clays are explored. The paper focuses
142	then on non-expansive clays, exploring the relationship between pore fluid salinity, clay fabric
143	and void space. The experimental behaviour of oedometer reconstituted samples of a smectitic
144	non-expansive clay under the effects of mechanical loads imparted at different constant pore
145	fluid compositions and of salinity changes imparted at constant vertical stress is discussed. The
146	experimental results are exploited to formulate a constitutive model in the framework of elasto-
147	plasticity with generalized hardening. The model was then used to simulate the results of
148	experimental tests, both original and from the literature, performed on samples of different soils
149	and with different initial fabrics (reconstituted from slurry, undisturbed and statically compacted
150	samples). In all these cases, a very good agreement between model predictions and experimental
151	data was found.

153 2. Influence of pore fluid salinity on the liquid 154 limit of active and inactive clays

155

156 As recalled by Jang and Santamarina (2016), liquid limit determinations account for the mass of 157 water which is both adsorbed onto the particle surfaces and held within the fabric. Comparing 158 the liquid limits obtained using different saline solutions reveals then the impact of the pore fluid 159 composition on the fabric of a given clay in slurry state. However, expressing the results of the 160 liquid limit tests in terms of the ratio between the mass lost through heating in the oven to the 161 non-evaporated mass is here inappropriate, since dissolved salts don't evaporate and they would 162 be improperly considered as solid fraction. Furthermore, as the density of a solution depends on 163 the solute concentration and the fabric is related to the type and volume of voids in the soil, the 164 comparison between liquid limits at different salinities is more significant if expressed in terms 165 of void ratio $e_{\rm L}$ instead of gravimetric water content $w_{\rm L}$.

166 The effects of water salinity on the liquid limit was analysed for the soils listed in Table 1, where 167 mineralogical compositions are also provided. The corrections required to the measurements of 168 the mass lost in the oven were carried out accordingly to the procedure described in Noorany, 169 1983. The data set includes pure clays (kaolinite, illite and bentonite), as well as natural soils from 170 Italy, Norway and Japan. Both data from the literature and original determinations are 171 considered. The original determinations include a pure illite from Hungary (provided by BAAN 172 industrial materials, Formigine, Italy), a commercial sodium bentonite which was mixed with the 173 illite (provided by Laviosa Chimica Mineraria, Livorno, Italy), and Spigno Monferrato clay, a 174 natural clay proceeding from the Langhe region of Italy which has been the seat of diffuse slope

175 instabilities promoted by the dilution of the interstitial pore water (Musso et al., 2008; Musso et 176 al., 2017). Spigno Monferrato is a mixture of nontronite (a non-expansive ferrous smectite), illite 177 and chlorite. 178 Table 1 also reports the void ratio at liquid limit obtained mixing the powder of these clays with 179 distilled water, eL⁰, which ranges from 0.80 (Asrum clay) to 10.80 (Ponza Bentonite). The 180 normalised ratio $e_{\rm L}/e_{\rm L}^0$ is then used in Figure 1 to show in a single graph the effects of salinity on 181 the liquid limits of the different soils. Three different trends can be observed: eL decreases with 182 salinity in the case of pure expansive minerals (Ponza Bentonite) and of clays containing 183 expansive minerals (75 illite 25 bentonite, Marino clay); it remains about constant or increases 184 very mildly with salinity for Spigno Monferrato clay, illite and kaolinite; it increases sharply in the 185 case of quick clays from Norway (Asrum and Drammen clay) and Japan (Ariake clay).

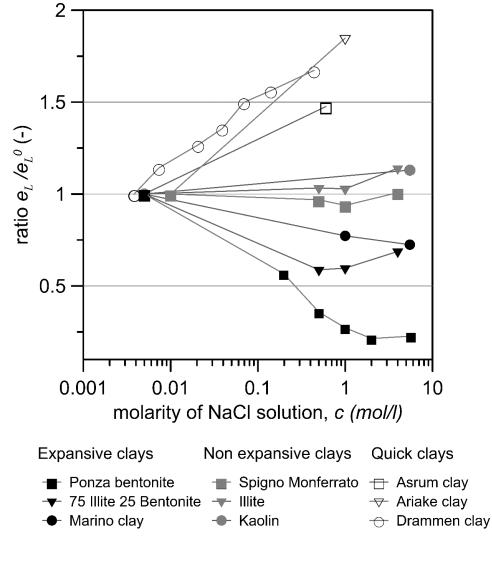


Figure 1 – Dependency of the void ratio at liquid limit e_{l} on NaCl molarity of the pore fluid for

different clays

194 Table 1 – Mineralogy and index properties with distilled water as saturating fluid for different clay

195 soils

Soil	Reference	Main minerals	Liquid limit	Void ratio at liquid
			w∟ (%)	limit with distilled
				water
				e ^{L0} (-)
Ponza Bentonite	Di Maio (1996)	montmorillonite 80%	390	10.80
		kaolinite 20%		
75 Illite – 25 Bentonite	This study	illite 75 %	161	4.43
		bentonite 25 %		
Ariake clay	Ohtsubo et al.	beidellite type, non-	89	2.54
	(1985)	expansive smectite 46%		
		Illite		
llite	This study	illite > 95 %	83	2.26
Kaolin	Di Maio et al	kaolinite 75-80%	50	1.37
	(2004)	illite 8-10%		
Marino clay	Di Maio et al.	kaolinite 30%	50	1.52
	(2004)	illite 10 %		
		mixed illite – expansive		
		smectite 10%		
Spigno Monferrato	This study	nontronite,	41	1.13
		non expansive smectite		
		illite,		

	chlorite		
Torrance (1974)	Illite Chlorite	32	0.88
Bjerrum and Rosenqvist (1956)	Illitic clay	29	0.79
	Bjerrum and	Torrance (1974) Illite Chlorite Bjerrum and Illitic clay	Torrance (1974)Illite32ChloriteBjerrumandIllitic clay29

197 The results for the expansive soils agree with the observations of Sridharan et al. (1986), 198 according to whom their liquid limit is mostly ruled by effect of the reduction with salinity of the 199 thickness of the double layer on the particles face. This appears not to be the case with the other 200 materials. The increase of e_L with salinity for the quick clays is consistent with the flocculated 201 fabric exhibited by these soils in saline water, which is lost upon exposure to fresh water 202 (Rosenqvist, 1966, Torrance & Ohtsubo, 1995). The flocculated fabric implies larger pores than 203 the aggregated fabric, explaining the increase of $e_{\rm L}$ with salinity. A similar phenomenon is 204 expected for the pure illite. While the liquid limit of kaolinite is controlled mostly by the pH while 205 it is not much affected by salinity alone (Sridharan et al., 1988), the kaolin considered in Figure 1, proceeding from the work of Di Maio et al. (2004) contains a 10 % fraction of illite, which might 206 207 justify the increase of eL with salinity, while the different behaviour of different minerals might 208 explain the trend of e_{L} for Spigno Monferrato clay, where e_{L} decreases slightly at lower salinities 209 and increases at higher salinities.

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- 211

212 3. Compression behaviour of non expansive 213 clays along chemo-mechanical paths

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216

215 3.1 Elastic and elasto-plastic compliance along constant salinity paths

As salinity can impact on the fabric and liquid limit of clays, it is expected to affect also their behaviour along compression. The dependency of oedometer compressibility on the salinity (NaCl concentration) of the pore water for the soils listed in Table 1 and a few other materials from the literature is shown in Figure 2. All the samples considered were loaded while saturated with the water of preparation (i.e. no replacement of the pore fluid have been performed). Compressibility is here expressed in terms of elastoplastic and elastic logarithmic compliance, λ and κ respectively:

224

$$\lambda = -\frac{\Delta e}{\Delta \ln \sigma'} \tag{1}$$

$$\kappa = -\frac{\Delta e}{\Delta l n \sigma'} \tag{2}$$

227

228 λ was evaluated along the virgin loading branch in the stress range between 150 kPa and 300 229 kPa, while κ was determined along the unloading branch. The relationship between λ and salinity 230 is similar to the one between e_{L} and salinity: λ decreases dramatically with concentration for soils 231 containing expansive minerals, it has a moderate increment with concentration for the illite and 232 for the quick clays, and it remains about constant for kaolin and Spigno Monferrato clay. The 233 influence of salinity on κ is generally very small. While most data refer to reconstituted 234 conditions, Figure 2 also reports data of an intact sample of Drammen clay (Torrance, 1974) and

235	of a compacted low-activity illitic clay (Witteveen et al., 2013). The compliance of the intact
236	Drammen clay is higher than the one of the reconstituted Drammen clay, but the role of pore
237	fluid chemistry is similar, as λ moderately increases with the concentration in both cases. No
238	measurable effects of the pore fluid concentration on compliance were found for the illitic clay
239	in Witteveen et al. (2013). Ù
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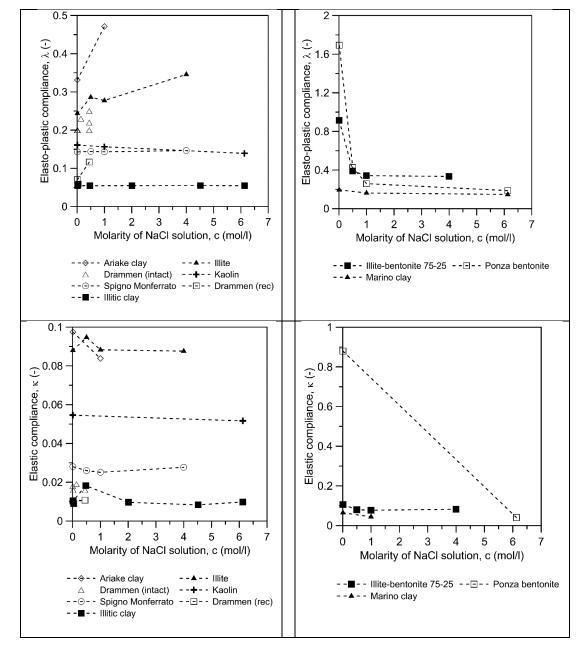




Figure 2 – Dependency of soil compressibility on NaCl molarity for different clays. (a) elasto plastic compliance of non-expansive clays, (b) elasto-plastic compliance of expansive clays, (c)
 elastic compliance of non-expansive clays, (d) elastic compliance of expansive clays

247 3.2 Compression curves of non-active clays and relationship with fabric

248

249 The salinity of the pore water of preparation might affect the compression behaviour also by 250 impacting on the position of the Normal Compression Line (NCL), i.e. on the void ratio associated 251 to first loading under a given effective stress. Restricting the analysis to the soils tested in the 252 present work, Figure 3 provides the experimental results for the illite and Spigno Monferrato 253 samples. All the specimens were reconstituted and they were prepared by mixing the dry soil 254 powder with the mass fluid required to impose an initial void ratio $e \cong 1.2 e_{L}$. Distilled water and 255 NaCl solutions with different molarities (0.5 M, 1 M, 4M) were used. For both soils, salinity has a 256 small effect on λ , but its increase translates the NCL to higher void ratios. The opposite effect 257 occurs for expansive soils, whose NCL moves to lower void ratios as salinity increases (see e.g. 258 Di Maio, 1996).

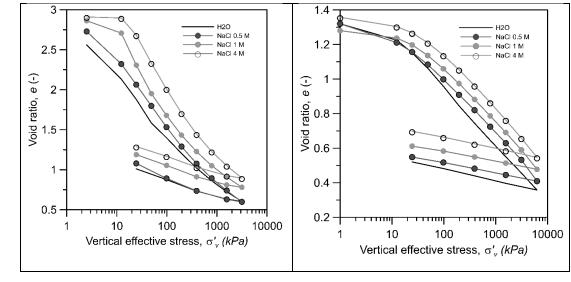


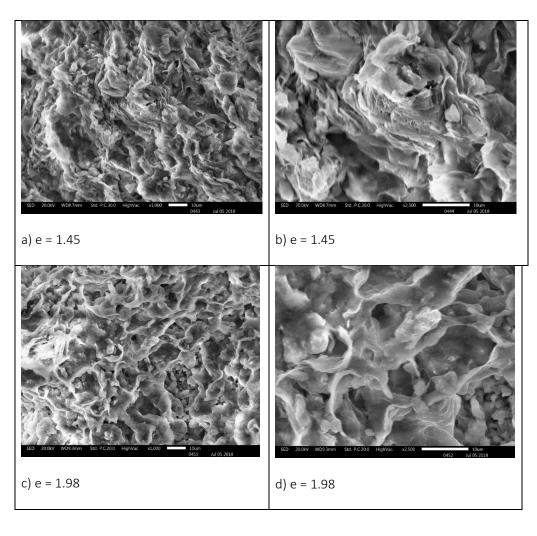


Figure 3 – Oedometer compression curves of illite (left) and Spigno Monferrato clay (right) saturated with NaCl solutions of different molarity

The effects of salinity on the compression behaviour descend from the effects of salinity on fabric. Different microstructural analyses were conducted on specimens normally consolidated at a vertical stress $\sigma'_v = 98$ kPa. All specimens were freeze dried before microscopic analysis, to avoid modifications of the pore network due to water evaporation in natural conditions. Scanning Electron Microscope (SEM) images of the illite specimens prepared with distilled water

267 and with the 4 M NaCl solution are presented in Figure 4. The differences between the two fabrics 268 is very clear. The distilled water specimen (Fig. 4a and 4b) has a void ratio e = 1.45 and its particles 269 are mostly aggregated parallel one to the other in a face to face arrangement, with elongated 270 pores whose main apertures have size of the order of about 1 μ m. The 4 M NaCl specimen (Fig. 271 4c and 4d) has a larger void ratio e = 1.98. A cardhouse fabric emerges, with particles disposed in 272 an edge to edge arrangement, forming larger pores with diameters of the order of 5 – 10 μ m. 273 Notwithstanding the load that was applied, the flocculated fabric imparted to the specimen 274 mixing the soil with highly saline water is still evident, which explains the higher void ratio of this 275 specimen with respect to those prepared at lower salinities.

276



278Fig. 4 SEM images of reconstituted illite specimens prepared at $e = 1.2 e_l$ and loaded to 98 kPa.279Pore fluid is: (i) distilled water for images a) and b); (ii) 4 M NaCl solution for images c) and d).280Bar length is 10 μ m in all pictures281

The effects of salinity on the fabric of Spigno Monferrato clay were investigated by means of Mercury Intrusion Porosimetry (MIP) tests, performed on the specimens prepared with distilled water and with the 4 M NaCl solution. The Pore Size Density function (PSD) curves of these specimens are presented in Figure 5: they show a peak in correspondence of a pore radius slightly smaller than 1 µm and they almost overlap at smaller radii. However, the saline specimen has a significant fraction of larger pores with radii ranging from 1 to 10 μ m, which is not present in the distilled water specimen. This is consistent with the interpretation provided for the fabric of Spigno Monferrati clay, that is characterized by larger voids in the presence of saline pore fluid.

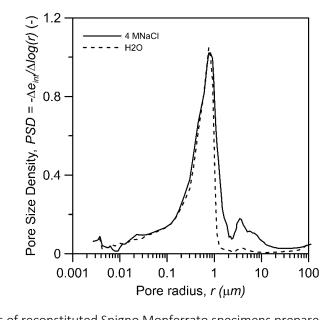


Fig. 5 PSD curves of reconstituted Spigno Monferrato specimens prepared at their liquid limitand loaded to 98 kPa.

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3.3 Chemo mechanical loading paths: evidences on reconstituted SpignoMonferrato clay
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299 To investigate the combined effects of mechanical and chemical solicitations, a chemo-
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300 mechanical loading path was imposed on a reconstituted specimen of Spigno Monferrato clay,

301 according to the sequence shown in Figure 6, while the experimental results in the compression

- 302 plane are provided in Figure 7. A slurry specimen was prepared using distilled water to obtain an
- initial void ratio $e_0 = 1.2 e_L$ and then placed in an oedometer.

304 Distilled water was poured in the oedometer cell and mechanical loading was applied by doubling 305 the vertical stress every 24 hours up to σ'_{v} = 196 kPa (point A in Figures 6 and 7). This vertical 306 stress was kept for 24 hours and then the fluid in the cell was replaced with a 4 M NaCl solution, 307 renewed daily to ensure constant salt concentration of the pore fluid at the boundary of the 308 specimen. The slow process of cation diffusion from the oedometer cell to the interior of the 309 specimen caused volume change to occur along time (i.e. osmotic consolidation, according to 310 Barbour & Fredlund, 1989). The corresponding volumetric strain took place in about 1 week. As 311 it can be appreciated in Figure 7, a decrease in void ratio occurred (Point A' in Figures 6 and 7). 312 To remove the effects that imparted by creep (as suggested by Torrance, 1974), in Figure 7 the 313 void ratios under the mechanical loads refer to the end of mechanical consolidation, while the 314 changes in void ratio measured during the chemical loads were corrected removing the effects 315 of secondary compression, expected to grow linearly with the logarithm of time.

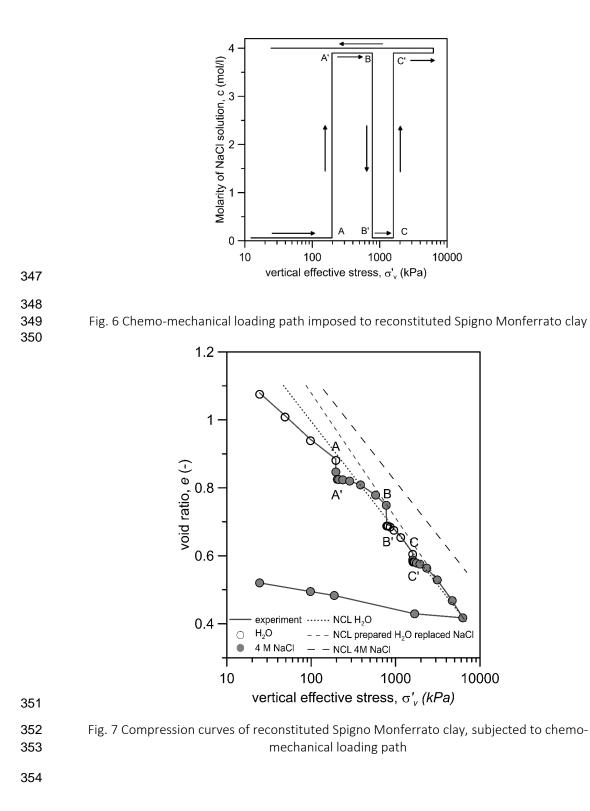
316 A mechanical loading sequence started again with the saline solution as pore and cell fluid. Small 317 stress increments were initially applied to appreciate possible effects of the chemical history on 318 the mechanical behaviour, i.e. to detect as precisely as possible any change in preconsolidation 319 pressure. Under a vertical effective stress σ'_v = 780 kPa (point B), the procedure was inverted and 320 the cell fluid was replaced with distilled water, renewed daily to ensure constant fluid 321 composition at the specimen boundary. As long as the NaCl ions diffused outside of the specimen, 322 the specimen progressively shrank and the void ratio reduced (point B' in Figures 6 and 7). Load 323 increments were then imposed at constant pore fluid composition, again paying attention to the 324 identification of preconsolidation pressure evolution. Another salinisation step was performed 325 under a vertical effective stress σ'_{v} = 1600 kPa with a small decrease in the void ratio from

e = 0.59 to e = 0.58 (points C and C'). The following loading and unloading were performed at
 constant salinity.

328 As evident from Figure 7, the stress increments which immediately followed the first salinisation 329 were characterised by an increase in stiffness (i.e. smaller compliance) with respect to the 330 previous ones, so the experimental points do not align with the projection of the compression 331 line deduced for the previous steps with distilled water as saturating fluid. A larger compliance 332 was recorded again for the stress increment between 600 and 780 kPa. Salinisation appears thus 333 to provide a sort of preconsolidation, causing an apparent OCR of the order of 2.5 - 3. A small 334 preconsolidation effect also occurred with the salinization step imposed at σ'_{v} = 1600 kPa. On 335 the other hand, after desalinisation (e.g. between points B' and C) the compliance that was 336 registered immediately is comparable to one of the virgin material, and no preconsolidation 337 effects were observed.

338 It is interesting to observe that, on the compression plane, the points which correspond to 339 saturation with distilled water align reasonably well along a certain Normal Compression Line. 340 The 'normally consolidated' points (i.e. for which the apparent OCR is 1) of the 4 M NaCl condition 341 align instead along another NCL. The position of the NCL of the specimen that was prepared and 342 loaded with the 4 M NaCl solution is also drawn in Figure 7. It can be appreciated that the NCL 343 for the condition of preparation with distilled water and loading with 4 M NaCl lays between the 344 NCL of preparation and loading with distilled water and the NCL of preparation and loading with 345 brine.

346



4 An elasto-plastic model for the chemo mechanical behaviour of non-expansive clays

357

358 The experimental evidences collected in the previous sections provide the basis for the 359 formulation of a simple phenomenological constitutive model capable of predicting the 360 mechanical response of non - expansive clays subject to salinity changes. The model is developed 361 the framework of elasto-plasticity with generalized hardening in (see, e.g., 362 Della Vecchia et al, 2013, Tamagnini & Ciantia, 2016). The role of pore fluid chemistry on the 363 mechanical response is assumed to directly influence both the constitutive stress and the evolution of the internal state variable. 364

365

366 4.1 Stress variables 367

368 The model is based on the definition of a mechanical constitutive stress variable, to reproduce 369 mechanical solicitations, and one environmental process variable to account for changes in the 370 chemistry of the pore fluid. The mechanical variable is the Terzaghi effective stress tensor, with 371 components σ'_{ij} :

372

373

 $\sigma'_{ij} = \sigma_{ij} - u\delta_{ij} \tag{3}$

374

where σ_{ij} is the total stress, *u* is the pore fluid pressure and δ_{ij} is the Kronecker's delta. The role of cation exchange is here neglected and the osmotic suction π is adopted as chemical process variable. The general expression for π is :

(5)

378

$$\pi = -\frac{RT}{v_w} ln(a_w) \tag{4}$$

380

381 where *R* is the universal gas constant (8.31 J mol⁻¹ K⁻¹), *T* is the absolute temperature, v_w is molar 382 volume of water and a_w is the activity of water, which depends on the concentration of dissolved 383 salts. At low concentrations the van't Hoff equation can be used:

384

385

386

where *i* is the number of species in which the salt dissolves (e.g. 2 for NaCl and 3 for CaCl₂) and *c* is the molar concentration of the electrolyte. For molar concentrations below 1 mole/liter, the error introduced by van't Hoff equation is smaller than 5 % (Mitchell & Soga, 2005).

 $\pi = icRT$

For the sake of simplicity, the usual volumetric and deviatoric decomposition of the stress and
 strain tensor in axis-symmetric conditions is introduced in the following.

392

393 4.2 Elastic behaviour

394

Elastic volumetric strain increments are split into two contributions, one due to mechanical loading, $\dot{\varepsilon}^{e}_{v\,mec}$ and the other due to osmotic suction changes, $\dot{\varepsilon}^{e}_{v\,ch}$. The mechanical contribution is defined evaluated through the logarithmic compliance κ adopted in critical state soil mechanics. A similar parameter, the chemical elastic logarithmic compliance κ_{π} , is introduced to evaluate the chemical component. The whole elastic volumetric strain increment $\dot{\varepsilon}^{e}_{v}$ then reads:

401
$$\dot{\varepsilon}_{v}^{e} = \dot{\varepsilon}_{v \ mec}^{e} + \dot{\varepsilon}_{v \ ch}^{e} = \frac{\kappa \dot{p}'}{v \ p'} + \frac{\kappa \pi}{v} \frac{\dot{\pi}}{\pi + \pi_{0}}$$
(6)

403 where *v* is the specific volume, π_0 is a reference osmotic suction (e.g. 1 kPa) introduced to avoid 404 infinite values of v when π goes to zero and p' is the mean effective stress. The contribution of 405 osmotic suction to elastic shear strain is neglected, so that increment of elastic deviatoric strain 406 $\dot{\varepsilon}_s^e$ reads:

 $\dot{\varepsilon}_s^e = \frac{1}{3G}\dot{q} \tag{7}$

408 where G is the (constant) shear modulus and q the deviator stress.

409

410 4.3 Elastic-plastic behavior

411

The yield surface in the (p',q, π) space is defined on basis of a simple mathematical interpretation
of the chemo-mechanical response in the compression plane. The approach followed is inspired
by the proposal of Alonso et al. (1990) to account for the role of matric suction on the behaviour
of unsaturated non-expansive soils.
As shown in the previous sections, the position and the slope of the NCL depend on the fabric
imparted by the history of mechanical and chemical loads. Both the parameters that identify the

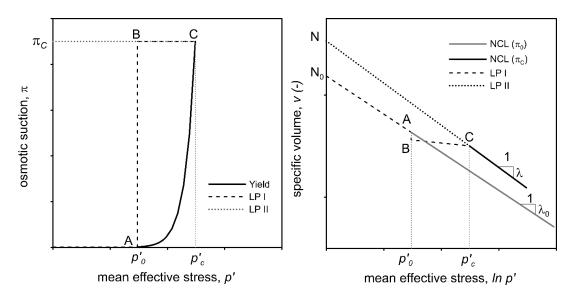
418 NCL in the compression plane (the specific volume N for a reference mean effective stress p'_r and

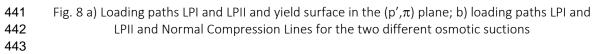
- 419 the elasto-plastic logarithmic compliance λ) may change with π . In the following, N_0 and λ_0 refer
- 420 to the soil saturated with distilled water, whereas $N(\pi)$ and $\lambda(\pi)$ refer to a generic saline solution
- 421 of osmotic suction π .

The preconsolidation pressure is expressed as p'_0 when the soil is saturated with distilled water and it is expressed as $p'_c(\pi)$ when the soil is saturated by the generic saline solution. The mathematical link between p'_0 and p'_c can be obtained considering two different loading paths, LPI and LPII in the stress plane (p', π) of Figure 8a and in the compression plane (v, ln p') of Figure 8b.

427 Along loading path LPI a sample 'prepared' and saturated distilled water is loaded in virgin 428 conditions up to p'_0 (point A): during this stage it moves along the NCL corresponding to $\pi = 0$. 429 Osmotic suction is then increased to $\pi = \pi_c$ while the mean effective stress is kept constant: this 430 causes compressive elastic volumetric strains and the specific volume reduces (path AB in 431 Figure 8). From B, the mechanical stress is further increased while the osmotic suction is kept 432 constant. Elasto-plastic volume strains take place only when the specific volume lays on the NCL, 433 thus volume contraction will initially be elastic as the NCL (π c) lays above the NCL (π = 0). Yielding 434 occurs when the elastic reloading line meets the NCL (π _c), i.e. at point C in Figure 8, where the 435 mean effective stress is p'_{c} and the specific volume is v_{c} . By further increasing the mechanical 436 stress, the behaviour will be elastoplastic and the sample will move along the NCL corresponding 437 to $\pi = \pi_c$.

Point C can also be reached through the loading path LPII, corresponding to mechanical loading at constant osmotic suction $\pi = \pi_c$ of a specimen that has the same fabric of C. In the compression plane, LPII marks the NCL(π_c) between $p' = p'_r$ and $p' = p'_c$.





444 By evaluating v_c along LPI:

445
$$v_{C} = N_{0} - \lambda_{o} ln \left(\frac{p'_{0}}{p'_{r}}\right) + \Delta v^{ch} - \kappa ln \left(\frac{p'_{c}}{p'_{0}}\right)$$
(8)

446 where Δv^{ch} is the elastic change in specific volume when the osmotic suction increases from 0 447 to π , evaluated with eq. (5):

$$\Delta v^{ch} = -\kappa_{\pi} ln \left(\frac{\pi + \pi_0}{\pi_0} \right) \tag{9}$$

449 By evaluating v_c along LPII:

448

450
$$v_C = N - \lambda \ln\left(\frac{p'_c}{p'_r}\right)$$
(10)

451 By introducing (8) in (7) and equating with (9), it follows:

452
$$\frac{p_{c}}{p_{0}'} = \left(\frac{p_{0}'}{p_{r}'}\right)^{\frac{\lambda_{0}-\lambda}{\lambda-\kappa}} \cdot e^{\frac{N-N_{0}}{\lambda-\kappa}} \cdot \left(\frac{\pi+\pi_{0}}{\pi_{0}}\right)^{\frac{\kappa_{\pi}}{(\lambda-\kappa)}}$$
(11)

453 Equation (10) provides the evolution of the yield mean stress p'_{c} with osmotic suction in isotropic

455 surface (*f* =0) of the Modified Cam Clay model:

$$f(p',q,p'_{c}) = \frac{q^{2}}{M^{2}} + p'(p'-p'_{c})$$
(12)

457 where *M* is the slope of the Critical State Line in the (p', q). The isotropic hardening law of the

458 Modified Cam Clay is used, with p'_0 as the internal variable:

459
$$\dot{\varepsilon}_{v}^{pl} = \frac{\lambda_{0} - \kappa}{v_{0}} \frac{\dot{p}_{0}'}{p_{0}'}$$
 (13)

460 where v_0 is the specific volume associated to p'_0 . An associated flow rule is finally adopted.

Suitable expressions relating N and λ to osmotic suction are also introduced. In analogy with what observed for the liquid limits, whose evolution with concentration follows a logarithmic trend (Figure 1), the following expressions are proposed:

464
$$N(\pi) = N_0 + \beta \ln\left(\frac{\pi + \pi_0}{\pi_0}\right)$$
 (14)

465

469

456

466
$$\lambda(\pi) = \lambda_0 + \eta \ln\left(\frac{\pi + \pi_0}{\pi_0}\right)$$
(15)

467 where β and η are model parameters.

468 4.4 Shape of the yield surface in the
$$(p',\pi)$$
 space

470 The shape of the yield function of eq. (11) in the (p', π) is controlled by:

471 (i) the dependency of
$$\lambda$$
 on osmotic suction $\left(\frac{p_{0}}{p_{r}'}\right)^{\frac{\lambda_{0}-\lambda}{\lambda-\kappa}}$,

472 (ii) the dependency *N* on osmotic suction
$$e^{\frac{N-N_0}{\lambda-\kappa}}$$
;

473 (iii) the value of osmotic suction itself
$$\cdot \left(\frac{\pi + \pi_0}{\pi_0}\right)^{\frac{\kappa_{\pi}}{(\lambda - \kappa)}}$$

These three factors depend on the mineralogy and initial fabric of the clay, so they might have different relevance for different clays or clay "preparations". For modelling purposes, two simplified hypotheses might be of interest:

a) osmotic suction does not affect the position and the slope of the NCL. In this case eq. (10)
reduces to:

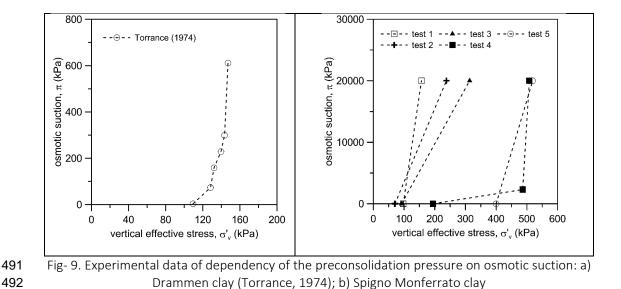
479
$$\frac{p'_c}{p'_0} = \left(\frac{\pi + \pi_0}{\pi_0}\right)^{\frac{\kappa_\pi}{(\lambda_0 - \kappa)}}$$
(16)

which points out the effects of the elastic chemical strains on the preconsolidation pressure;
b) osmotic suction affects the position of the NCL but not its slope. According to Section 3,
this seems to be the case for many non-expansive clays. Eq. (10) reduces then to:

483
$$\frac{p_{\prime c}}{p_{\prime 0}} = e^{\frac{N-N_0}{\lambda_0 - \kappa}} \cdot \left(\frac{\pi + \pi_0}{\pi_0}\right)^{\frac{\kappa_{\pi}}{(\lambda_0 - \kappa)}},\tag{17}$$

484

The shape of the yield surface is consistent with the experimental evidences provided by Torrance (1974) for Drammen clay and the ones collected in this study for Spigno Monferrato clay, shown in Figure 9. Both cases refer to reconstituted samples prepared mixing the dry soil powder with distilled water at a content higher than the liquid limit, and then loaded in the oedometer according to the sequence LPI from Figure 8a.



494 5 Model validation for reconstituted, 495 compacted and undisturbed soils

Since the mechanical behaviour of a given soil depends on the fabric imparted at its formation, the capabilities of the model were checked simulating oedometer tests performed on samples of reconstituted, compacted and undisturbed clays. To this extent, the model was implemented in a driver for the integration of the constitutive equations in rate form (see e.g. Cattaneo et al., 2011, 2014). The driver allows imposing histories of prescribed chemomechanical loading, which were assigned according with those adopted during the simulated experiments. In all the simulations, the reference osmotic suction was imposed as $\pi_0 = 1$ kPa.

504

505 5.1 Reconstituted Spigno Monferrato clay

507The simulations of two oedometer tests run on reconstituted Spigno Monferrato clay were508performed. The values of the parameter used in the simulation are reported in Table 2.

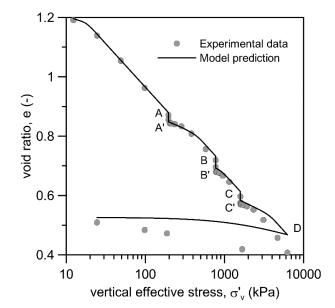
509 Table 2 Parameters and initial value of p'_0 used for the simulation of Spigno Monferrato clay

κ	$\lambda_0 = \lambda$	κ_{π}	ν	М	β	p'_0
(-)	(-)	(-)	(-)	(-)	(-)	(kPa)
0.04	0.127	0.006	0.3	0.98	0.003	18

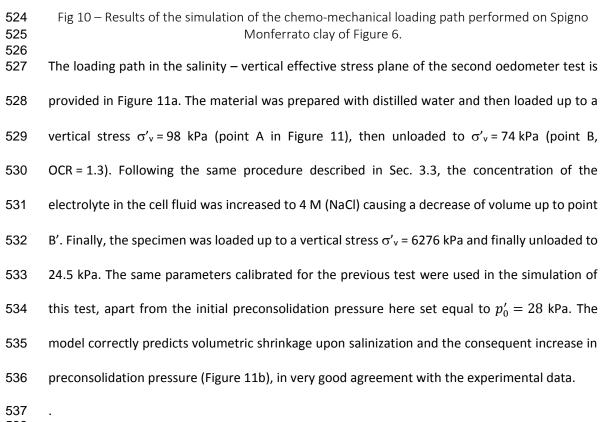
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511 κ and λ were calibrated on the compression response of a sample prepared and mechanically 512 loaded with distilled water as cell fluid, while κ_{π} was calibrated on the volumetric strains due to 513 salinization of the first test described below. The dependence of N on pore fluid salinity was 514 simulated with β = 0.003. The slope of the critical state line M^* was set equal to 0.98 and as a 515 first approximation it was set independent from the chemical concentration of the pore fluid. 516 The two tests are characterized by different chemo-mechanical loading paths. The first one was 517 provided in Figure 6 (experimental results in Figure 7). The predictions of the model, shown in 518 Figure 10, are in good agreement with the experimental data. The model is capable of predicting 519 volumetric shrinkage not only upon salinization (AA' and CC'), but also an irreversible void ratio 520 reduction upon desalinization (BB'). In accordance with experimental data, the convex shape of 521 the yield domain in the $(p' - \pi)$ plane implies an elastic response due to mechanical loading after 522 salinization and an elasto-plastic response due to mechanical loading after desalinization.







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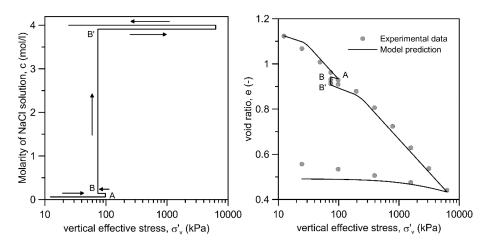


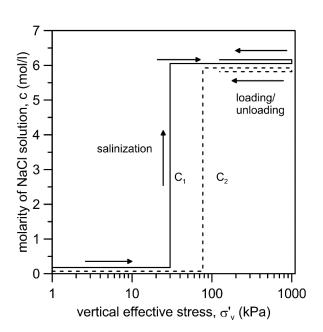
Fig 11 – (a) Loading path and (b) model predictions of the second chemo-mechanical test performed on Spigno Monferrato clay

546 5.2 Compacted illitic clay (Witteveen et al., 2013)

548 The capability of the model to reproduce the behaviour of compacted clays was checked 549 simulating two oedometer tests performed by Witteveen et al. (2013) on a compacted illitic soil 550 (liquid limit $w_L = 54\%$ and plasticity index PI = 24 %). Specimens have been prepared by mixing 551 the powder of the soil with distilled water at a water content below the liquid limit (w = 39 %) 552 and statically compacted in oedometer up to different vertical stresses (σ'_v = 30 kPa for test C1 553 and σ'_{v} = 77 kPa for test C2). According to the authors, water saturation was reached during static 554 compaction. Under these vertical stresses the specimens have been exposed to a saturated NaCl 555 saline solution: after suction equalization they have been loaded at constant suction to 556 σ'_v =1000 kPa and finally unloaded. The stress paths in the (σ'_v - M) plane is shown in Figure 12. 557 The parameters of the model were calibrated on the results of test C2 and then used to predicted 558 the response for test C1: their values are provided in Table 3 together with the initial value of p'_{0} . 559 The dependence of elasto-plastic compliance λ on pore fluid concentration was neglected, while

a small increase of *N* with osmotic suction was imposed ($\beta = 0.0013$). The comparisons between model predictions and experimental data are shown in Figure 13. The model proves able to reproduce with a very limited number of parameters the main features of the chemo-mechanical response on non-active clays, also if prepared by compaction. Remarkably, the obtained shape of the yield function in the (p'- π) plane allows a correct prediction of the elastic reloading stage if the material is loaded after salinization.

566



568 Fig 12. Chemo-mechanical loading path for compacted illitic soil (Witteveen et al, 2013) 569

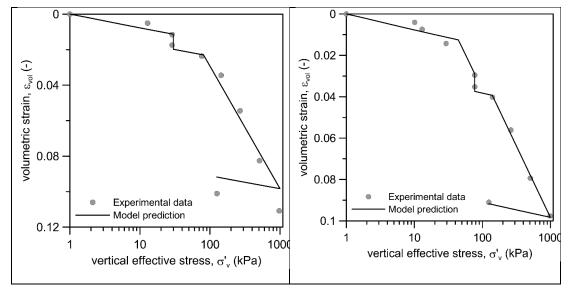


Fig- 13. Comparison between experimental data and model predictions for compacted illitic soil
in Witteveen et al, 2013. Specimens C1 (left) and C2 (right).

573 Table 3 Parameters and initial value of p'_0 used for the simulation of compacted illitic soil in

574 Witteveen et al. (2013)

κ	λ_0	κ_{π}	G	Μ	β	p_0'
(-)	(-)	(-)	(kPa)	(-)	(-)	(kPa)
0.006	0.058	0.0016	67	0.98	1.3 e-3	43

575

577

576 5.3 Intact and reconstituted quick clays

578	Torrance (1974) performed oedometer tests on undisturbed samples of intact Drammen clay, a
579	natural a low activity (w_L = 54 %, IP = 23 %, Activity = 0.46) Norwegian quick clay, whose pore
580	water has an initial (natural) salt concentration of 26 g/l (Torrance 1974). Undisturbed specimens
581	have been loaded up to 150 kPa, a value larger than the in situ preconsolidation pressure, and
582	then unloaded to 15 kPa, inducing an OCR equal to 10. Afterwards, the specimens have been
583	subjected to leaching (i.e. the exposure to a fluid with a smaller saline concentration than the
584	initial one) at constant vertical stress. The fluids used for the leaching process have different NaCl

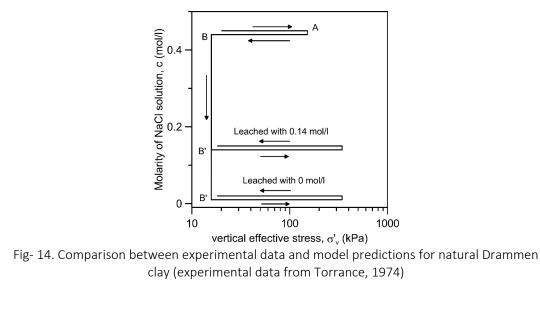
585 concentrations, namely 0, 1, 2, 3, 4 and 8 g/l. After leaching, specimens have been loaded again. 586 The loading path is shown in Figure 14 for leaching with 0 g/l and 8 g/l of NaCl (c = 0 mol/l and 587 c = 0.14 mol/l respectively). Experimental results show desalinization-induced swelling and, upon 588 reloading, a reduction in the preconsolidation pressure with respect to the one induced by 589 previous loading. The entity of the reduction is larger the lower the salinity of the leaching fluid. 590 The results of the simulations referred to the specimens leached with 0 g/l and 8 g/l are reported 591 in Figure 15. The complete set of parameter values and the initial value of the p'_0 used in the 592 simulation are reported in Table 4. Again, the dependence of λ on pore fluid concentration was 593 neglected, without compromising the quality of the numerical predictions. The model correctly 594 reproduces not only the elastic and elasto-plastic response upon mechanical loading, but also the 595 magnitude of swelling upon leaching.

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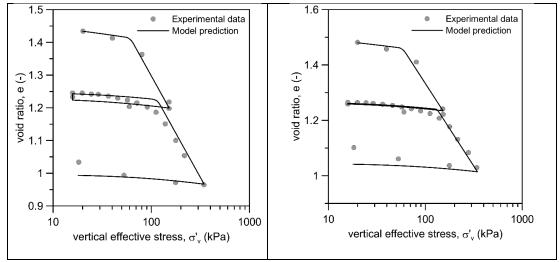


Fig- 15. Comparison between experimental data and model predictions for natural Drammen clay
(experimental data from Torrance, 1974). Leaching with distilled water (left) and with NaCl
solution with salt concentration 8 g/l (right)

606 Table 4 Parameters and initial value of p'_0 used for the simulation of natural Drammen clay

κ	λ_0	κ_{π}	ν	М	β	p'_0
(-)	(-)	(-)	(-)	(-)	(-)	(kPa)
0.022	0.235	0.0031	0.3	0.98	4.8 e-3	39

608 A further validation of the model was performed against the results of oedometer tests on 609 Vaterland clay (w_L= 40 %, IP=16%, Activity=0.40), another Norwegian quick clay (Torrance, 1974). 610 Four specimens of Vaterland marine clay had been remoulded with a NaCl solution of 611 concentration 26 g/L (c = 0.44 mol/l) as saturating fluid, loaded in oedometer to a vertical 612 effective stress $\sigma'_v = 18$ kPa (point A in Fig. 16 a), and then unloaded to $\sigma'_v = 6$ kPa (point B in 613 Fig. 16 a), inducing an OCR = 3. The specimens were then leached, i.e. exposed to distilled water 614 keeping the vertical stress constant (point B'): as consequence, the specimens experienced a 615 volume reduction. After leaching, three specimens had been exposed to potassium chloride at 616 different concentrations (namely K-low, K-medium and K-high, corresponding to 0.014 mol/l,

617 0.045 mol/l and 0.097 mol/l). One specimen, referred to as "leached specimen", had only been

subjected to distilled water, without further changes in the pore fluid.

Afterwards, all the specimens had been reloaded to a vertical stress of 168 kPa. The loading paths

are shown in Figure 16 a, while the experimental data, as well as the model predictions, referring

621 to the leached specimen and to the specimen exposed to a medium concentration of potassium,

622 are shown in Figure 16 b.



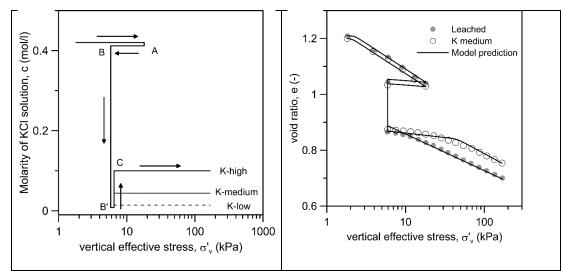
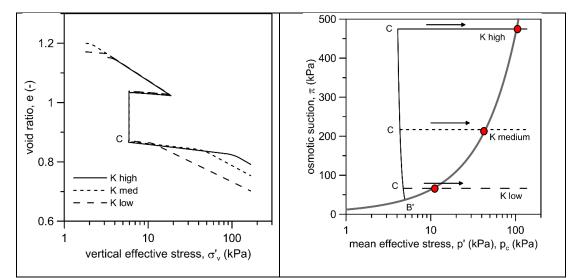
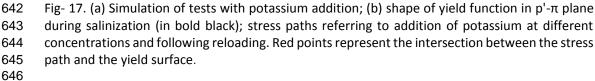


Fig- 16. Loading paths for Vaterland specimens leached with distilled water (BB') and then exposed
to potassium chloride (B'C) (left); comparison between experimental data and model predictions
for the leached specimen and for the specimen exposed to a medium concentration of potassium
chloride (right) (experimental data from Torrance, 1974)

The experimental data confirm that the normal compression lines interpolating the virgin compression branches tend to shift upwards in the compression plane at increasing electrolyte concentrations. The model proved able to reproduce the volumetric collapse caused by leaching of moderately overconsolidated specimens and the simulations are in good agreement with the experimental results. The dependence of λ on π (see eq. 15) was considered and the parameters adopted in the simulations are reported in Table 5. The proposed framework consistently
predicts that the preconsolidation pressure depends on the history of both mechanical and
chemical loadings: indeed, after volumetric collapse the leached specimen behaved as a normally
consolidated material, while the K-medium specimen behaved as an overconsolidated material.
Figure 17a presents model predictions for the three specimens exposed to potassium chloride.
Due to the shape of the yield function which expands with osmotic suction (Figure 17b), the yield
stresses increase at increasing salt concentration.

641





647

Table 5 Parameters and initial value of p'_0 used for the simulation of Vaterland clay

κ	λ_0	κ_{π}	ν	Μ	β	η	p_0'
(-)	(-)	(-)	(-)	(-)	(-)	(-)	(kPa)
0.015	0.04	0.003	0.3	0.98	0.045	0.002	1.0

649 6 Conclusions

650 The physico-chemical interaction between clay particles depends on the type of pore fluid and 651 the mineralogy of the clay. The fabric of natural and reconstituted non-expansive illitic and 652 smectitic clays in distilled water is mostly related to particle aggregation without flocculation, 653 whereas it shows flocculation of particles or aggregates of particles in presence of dissolved salts. 654 The flocculated fabric persists also when the stress is increased, whereas it is at least partially 655 destroyed upon desalinization. The flocculated fabric is associated to larger pores with respect 656 to the aggregated one, which helps explaining the experimental evidences that show the NCL of 657 these soils moving to higher void ratios as the salt content increases. Changes in pore chemistry 658 occurring under constant stress cause similar movements of the NCL, although to a limited 659 extent. Even if the fabric of compacted soils is dominated by the effects of the mechanical load 660 imparted in unsaturated conditions during compaction, the same effects seems to occur also for 661 these soils. While changes in the thickness of the DDL alone would suggest an elastic type of 662 behaviour, with volume contraction upon salinity increase and expansion upon salinity decrease, 663 the experimental evidence shows that in normally consolidated non-expansive clays 664 desalinitisation leads to inelastic compressive volume strains (collapse). These evidences at the 665 phenomenological level resemble those observed for non-active unsaturated soils exposed to a 666 decrease in matric suction. Their critical analysis lead to the proposal of an elasto-plastic model, 667 which was formulated following the same procedure adopted by Alonso et al. (1990) for the 668 Barcelona Basic Model (BBM). A yield function which expands with osmotic suction, governed by 669 the elastic chemical compressibility and the variations in the position and slope of the NCL, 670 follows naturally from this procedure. The role of such a yield function with respect to chemo-

671	mechanical plastic effects is analogous to the one played by the Loading Collapse (LC) in the BBM.
672	However, it is worth noting that the LC for unsaturated soils and the present yield function (eq.
673	10) account for different physical processes (LC for capillarity and the present one for salinity,
674	mediated through changes in fabric arrangements discussed previously).
675	

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681

682 Notation

a _w	activity of water
С	molar concentration
eL	void ratio at liquid limit with saline solution as saturating fluid
e_L^0	void ratio at liquid limit with distilled water as saturating fluid
i	number of species of the solute
М	slope of the Critical State Line in the (p',q) plane
Ν	intercept of the Normal Compression Line
No	Intercept of the Normal Compression Line with distilled water

p'	mean effective stress
p'c	preconsolidation pressure
p'r	reference mean stress
p'_	preconsolidation pressure with distilled water
q	deviatoric stress
R	universal gas constant
Τ	absolute temperature
и	pore pressure
V	specific volume
Vw	molar volume of H ₂ O
β	model parameter describing the dependency of N on osmotic suction
δ_{ij}	Kronecker delta
ės ^e	increment of deviatoric strain
έ ^e ν ch	increment of elastic volume strain due to osmotic suction change
Ė ^e v mec	increment of elastic volume strain due to stress change
έ ^e _v	increment of elastic volume strain
η	model parameter describing the dependency of $\boldsymbol{\lambda}$ on osmotic suction
К	elastic logarithmic mechanical compliance
κπ	chemical elastic compliance
λ	elasto-plastic logarithmic mechanical compliance
λ_0	elasto-plastic logarithmic compliance at reference osmotic suction
π	osmotic suction

π_0	reference osmotic suction
σij	component of total stress
σ'_{ij}	component of Terzaghi effective stress

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