Swelling phenomena and effective stress in compacted expansive clays

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Abstract

The central aim of the paper is to discuss the applicability of the effective stress principle as defined by Terzaghi (total stress minus pore water pressure) to predict the behaviour of expansive clay aggregates. Phenomena occurring between individual clay minerals are reviewed first at the molecular level obtained in the colloid science research. In particular, it is noted that, for inter-particle distances higher than approx. 1.5 nm, the pore water pressure in bulk equilibrium solution forms an additive component of the inter-particle disjoining pressure. It is concluded that for these distances the Terzaghi's effective stress principle should be adequate to describe the clay behaviour. To support the developments, an extensive experimental database of nine different sodium and calcium bentonites available in publishhed literature was analysed. With the aid of double structure constitutive modelling, procedures were developed to extract the information on the behaviour of clay aggregates from the experimental measurements. It was then shown that unconfined water retention curves, swelling pressure tests, swelling under constant load tests and mechanical unloading tests are all uniquely related in terms of the dependency of dry density (or void ratio) of clay aggregate vs. mean effective stress. By considering reversibility of aggregate behaviour and full saturation of aggregates, this implies that the effective stress principle is valid in predicting expansive clay aggregate volumetric deformation.

Keywords: expansive clay; bentonite; effectives stress; swelling pressure.

Introduction

Compacted bentonites are clays with a high smectite content and they have received considerable research attention in the past, particularly due to their application in engineered barrier systems for high-level nuclear waste repositories. Various models have been developed to reproduce their swelling behaviour. The empirical models relate the swelling pressure developed under confined swelling (Nayak and Christensen 1971, Low 1979) or swelling strain under constant load (Yevnin and Zaslavsky 1970, Buzzi et al. 2007, Buzzi and Giacomini 2010, Buzzi 2010) to the clay dry density or to the inter-particle distance under the assumption of homogeneous and parallel particle distribution. Another group of models is based on the diffuse double layer theory, attributed to Gouy (1910) and Chapman (1913) in its basic form. The dependency of swelling pressure on inter-particle distance is calculated through quantification of the repulsive and attractive forces caused by physico-chemical effects at the particle scale level. The basic form of the diffuse double layer theory is only applicable for smectite particles suspended in monovalent electrolyte solutions at low concentrations

(Warkentin and Schofield 1958, Yong et al. 1963, Sridharan and Jayadeva 1982, Zhang et al. 1993, Yong 1999a, Yong 1999b, Komine and Ogata 2004, Mitchell and Soga 2005, Phillips and Tripathy 2011, Liu 2013). For different cation types and higher densities, various modifications to this theory have been proposed (Yong and Mohamed 1992, Komine and Ogata 1999, Sridharan and Choudhury 2002, Tripathy et al. 2004), including numerical studies using the finite element method (Smith et al. 2009) and the discrete element method (Katti et al. 2009). An essential component of these models is the evaluation of the mid-point electrical potential between two parallel clay particles. This potential, which primarilly controls the inter-particle repulsion, is controlled by the ionic strength of pore water (and thus depends on osmotic suction in equilibrium solution). It can be balanced either by the matric potential or by the external load, which is then denoted as a swelling pressure. In principle, it is possible to by-pass the description of the inter-layer phenomena and directly relate the swelling pressure to the soil water potential. Such an approach has been adopted in the continuum thermodynamic models of Low and Anderson (1958), Sposito (1972), Oliphan and Low (1982), Dueck and Börgesson (2007) and Lempinen (2011).

The above models relate the swelling pressure/water potential to inter-particle distance and assume that the particle distribution is homogeneous, in which case the inter-particle distance can be calculated from the specific surface area and global dry density of the material. The actual structure of a compacted clay is, however, more complex. It is formed by clay particle clusters (hereafter referred to as aggregates), which form the primary component of a rather complex structure (Fig. 1). The structure schematically depicted in Fig. 1 has been justified by various experimental and numerical studies, see Mašín (2013) for a detailed review. In order to predict the mechanical response of such a material, models that explicitly consider two structural levels have been developed by several authors (Gens and Alonso 1992, Yang et al. 1998, Alonso et al. 1999, Thomas and Cleall 1999, Sánchez et al. 2005, Mašín 2013). The double structure models often assume a simple representation of the aggregate response; that is (1) the Terzaghi's effective stress is assumed to be valid, (2) the behaviour of the aggregate is assumed to be reversible elastic and (3) the aggregate is assumed to be fully saturated. The focus of this paper is the issue of the applicability of the Terzaghi's effective stress.

Throughout the paper, the following terminology is adopted, which is also identified in Fig. 1. The notion of a *clay layer* represents a 2:1 TOT (tetrahedral-octahedral-tetrahedral) sandwich layer of clay mineral crystals. The notion of a *clay particle* represents a stack of clay layers inter-connected by up to four layers of *crystaline water*. An assembly of clay particles, denoted as the *clay aggregate*, forms the unit of the compacted clay double structure.
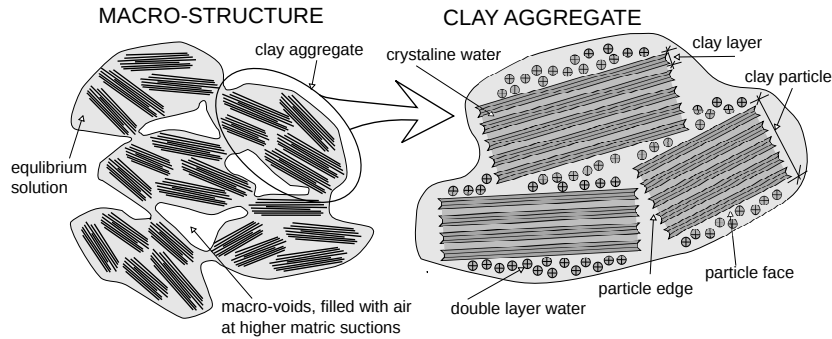
Figure 1: Schematic representation of the structure of a compacted clay and of the adopted terminology.

An assembly of aggregates is denoted as the *macro-structure*, while the voids between the aggregates are denoted as *macro-voids*. The part of the clay particle surface parallel to the clay layers is denoted as the *particle face*. The portion of the clay particle surface normal to the particle face is denoted as the *particle edge*. Diffuse double layers form around particle faces, with the bound water denoted as *double layer water*. The water beyond the diffuse double layers is denoted as the *equilibrium solution*. Note that there is no strictly defined boundary between the double layer water and equilibrium solution. Instead, ion concentration gradually decreases with distance from the clay particle, finally reaching a concentration of the equilibrium solution.

Effective stress in compacted clays – review of current understanding

The concept of effective stress, introduced by Terzaghi (1936), transforms a multiphase porous medium into a mechanically equivalent single-phase continuum. It therefore provides a powerful unifying reference for the analysis of soils as multiphase porous media. For saturated soils, Terzaghi (1936) introduced the effective stress in the form

$$\sigma' = \sigma^{tot} - u_w \tag{1}$$

where σ' is the effective stress, σ^{tot} is the total external stress and u_w is the pore water pressure[1].

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[1]Note that tensorial formulation of Eq. (1) reads $\boldsymbol{\sigma}' = \boldsymbol{\sigma}^{tot} - \mathbf{1}u_w$, where $\mathbf{1}$ is a second-order identity tensor and $\boldsymbol{\sigma}'$ and $\boldsymbol{\sigma}^{tot}$ are effective and total stress tensors respectively.

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In the more general case of partially saturated soils with variable fluid chemistry, pore water pressure is only one component of the total energy status of the pore water. Matric potential, measured by matric suction $s_m = u_a - u_w$ (where u_a is the pore air pressure), represents the potential of the pore water with respect to water of the same chemistry at ambient atmospheric conditions; this is related to the air-water interfacial tension phenomena in unsaturated soils. Osmotic potential, measured by osmotic suction s_π, represents the potential of the pore water at ambient atmospheric conditions with respect to pure water at ambient atmospheric conditions. Osmotic suction is proportional to the molar concentration of chemical species present in the solution. Total suction s_t is the sum of the osmotic and matric components (among other components, which can however be neglected in the present analyses). Matric suction can also be adopted in the description of saturated soil with negative pore water pressure, in which case the matric suction is equal to minus the pore water pressure.

Central to the present work is the effective stress concept, whose very deffinition is interpreted in different ways by various authors. Terzaghi (1936) stated that "all measurable effects of a change of stress, such as compression, distortion, and a change in shearing resistance of a soil are due exclusively to changes in effective stress". It was soon understood that most soils, in fact, do not follow this definition. By example, Leonards (1962) pointed out that during creep (deforming soil under constant effective stress), the soil does not obey the effective stress principle in a strict sense. There is a number of other similar examples in soil mechanics. Apart from the secondary compression (1), we may list wetting-induced compaction of partially saturated soils (2); thermally-induced soil deformation (3); whethering-induced soil compaction (4) (Nova et al. 2003); deformation of clayey soil induced by variation of osmotic suction under constant matric suction (5).

Regardless of the fact that Leonards (1962) is correct that the Trezaghi's principle looses its validity in a strict sense for predicting the secondary compression phenomena, the equation (1) can still be used with advantage for constructing constitutive models (see, by example, Niemunis et al. 2009). Similarly, the concept of effective stress can be used for predicting the other phenomena listed above: the additional deformation may be treated either by incorporating additional elastic or plastic mechanism into the model, or (in some cases) by modifying the effective stress equation itself. Relevant for this paper is mechanism (6) (deformation of clayey soil induced by variation of osmotic suction). Here, we follow the first of the two approaches, while we point out that, in principle, incorporating the osmotic suction into the effective stress equation would also be possible.

Water in fine pores of clayey soils

The effective stress expression (1) has generally been accepted as representing the behaviour of saturated soils with free pore water, provided that the compressibility of the solid particles is negligible (Biot 1941, Skempton 1960, Nur and Byerlee 1971). A more complicated situation arises in the case of fine-grained soils, where a significant amount of water is tightly bound to the solid particles by physico-chemical phenomena at the molecular scale. Water in clayey soils exists in three basic states (Madsen and Müller-Vonmoos 1989): (1) Crystalline water, which is tightly bound within the structure of the clay particle in the inter-layer space; (2) double layer water, present between individual clay particles as part of the diffuse double layers and (3) equilibrium solution. For soils with larger inter-particle distances, the double layer water gradually becomes equilibrium solution with increasing distance from the particle surface due to gradually decreasing concentration of ions.

The amount of crystalline water depends on the matric forces (Yong 1999b) and is insensitive to the pore water chemical properties (Herbert and Moog 2000). Hydration with the crystalline water occurs in steps corresponding to the molecular layers of water (maximum of 3 to 4 layers) and is dependent on the major exchangeable cation type (Farmer and Russell 1971, Ferrage et al. 2005, Zheng et al. 2010). For example, Na-montmorillonite hydrates step-wise within a high relative humidity range (approx. 20%-100%, Devineau et al. 2006), whereas Ca-montmorillonite becomes hydrated at low relative humidities (high total suctions) and remains essentially intact with further wetting (Villar 2007, Morodome and Kawamura 2009). The step-wise hydration was explained by Israelachvili and Pashley (1983), who attributed it to deviations from the diffuse double layer theory at distances very close (< 1.5 nm) to the clay particle surfaces. The dependency of inter-particle forces on distance becomes oscillatory at distances below 1.5 nm and their minima correspond to stable states with one, two or three layers of crystalline water.

The diffuse double layer water is bound to the cation cloud that forms around negatively charged clay particles where hydration due to suction decrease depends on the clay mineralogical composition. During hydration Na-montmorillonite breaks apart into clay particles with variable number of layers due to diffuse double layer repulsion. For example, Saiyouri et al. (2004) observed that the particle size decreased from 350 layers to 10 layers when suction was reduced from 100 MPa to 1 kPa. On the other hand, Ca-montmorillonite remains more stable after saturation with crystalline water (Madsen and Müller-Vonmoos 1989, Mitchell and Soga 2005). The diffuse double layer size depends on the ion concentration in the equilibrium solution; it is thus directly related to osmotic suction (Callaghan and Ottewill 1974, Yong 1999b, Pusch 2006). An increase in the osmotic suction decreases the diffuse dou-

ble layer size, and consequently, for the given inter-particle distance, it decreases the particle repulsion when the layers overlap.

Existing effective stress models

The stress state in highly compacted clays is often considered to be composed of two parts: "mineral stress" (σ^m) which is transferred through the crystal structure of the clay mineral by means of edge-to-face contacts, and "inter-particle stress" (σ^{R-A}), transferred through diffuse double layers and crystalline water layers acting at face-to-face contacts. The inter-particle stress represents the electrostatic component of the disjoining pressure, and is equal to the difference between attractive (London van der Waals) and repulsive (in particular diffuse double layer) forces between the clay particles. Both these stresses contribute to the so-called structural stress σ^s, overall stress transferred by the clay structure.

Various authors have suggested different simplified models of the clay structure with the aim of isolating the σ^{R-A} and σ^m stress components from the structural stress σ^s. These models were classified by Hueckel (1992a). In the *parallel* models, the strains in the inter-particle spaces and in the mineral skeleton are assumed to be equal. Thus, the structural stress may be considered as the sum of the mineral and inter-particle components (Fig. 2a). The models by Lambe (1958, 1960) and Lu and Likos (2006) consider σ^s to represent the effective stress, whereas the models by Balasubramonian (1972), Sridharan and Venkatappa Rao (1973) and Morgernstern and Balasubramonian (1980) consider the effective stress to be represented by the mineral stress $\sigma^m = \sigma^s - \sigma^{R-A}$. This form of effective stress has been supported by Murad and Cushman (1996), Bennethum et al. (1997) and Moyne and Murad (2006) using the thermodynamic hybrid mixture theory of (Hassanizadeh and Gray 1979). Sridharan and Venkatappa Rao (1979) evaluated the parallel model of the latter type using shear strength data obtained by varying the dielectric constant of the pore medium. The cohesion intercept varied with the pore fluid type used, indicating that the structural stress, which was independent of the pore fluid chemistry, could not correctly predict the variation of shear strength under variable osmotic suction. This model was also adopted by Fam and Santamarina (1996).

Contrary to the parallel formulation, the *series* models (Bolt 1956, Hueckel 1992a) consider the structural stress, mineral stress and the inter-particle stress to be equal, while deformations in each of the components may be different (see Fig. 2b). In this case the structural stress is considered to represent the effective stress. A similar explanation for the effective stress within dense water-saturated Na-bentonite has been provided by Pusch (1982). He observed that under a given pore water chemistry the shear strength, as well as the elastic

response, could be interpreted uniquely in the σ^s space. Hueckel (1992a) argued that the parallel model of the $\sigma = \sigma^m$ type is suitable for prevailing edge-to-face particle contacts. On the other hand, the series model is, in his view, more suitable for representing face-to-face particle arrangements, which is relevant for highly compacted clays (Emerson 1962, Pusch 2006).

Sridharan and Venkatappa Rao (1973) argued that the shear resistance of the soil is controlled by the mineral stress σ^m, which, in their view, should be considered as the effective stress. However, they did not consider the shear resistance of the inter-layer and inter-particle spaces. Studies of the shear forces within molecular thin films indicate that the shear strength of the inter-particle space is dependent on the inter-particle distance. The inter-particle water does not transfer shear stresses for inter-particle distances larger than 7-10 molecular layers of water (1 molecular layer represents an approximate distance of 0.3 nm, Saiyouri et al. 2000). With decreasing distance, water progressively changes its properties from a Newtonian viscous fluid to solid-like behaviour (Israelachvili et al. 1988, Schoen et al. 1989, Homola et al. 1989, Cushman 1990, Gee et al. 1990). Water then has a yield point and the shear stress no longer depends on the shear rate. If the shear viscosity of the molecular film is back-calculated using its shear strength properties (although the viscosity concept actually breaks down), it is 5-7 orders of magnitude higher than its bulk value (Gee et al. 1990). Such an increase in viscosity is also much higher than indicated by Allam and Sridharan (1984) in their arguments against the use of face-to-face contact-based shearing resistance models for clays.
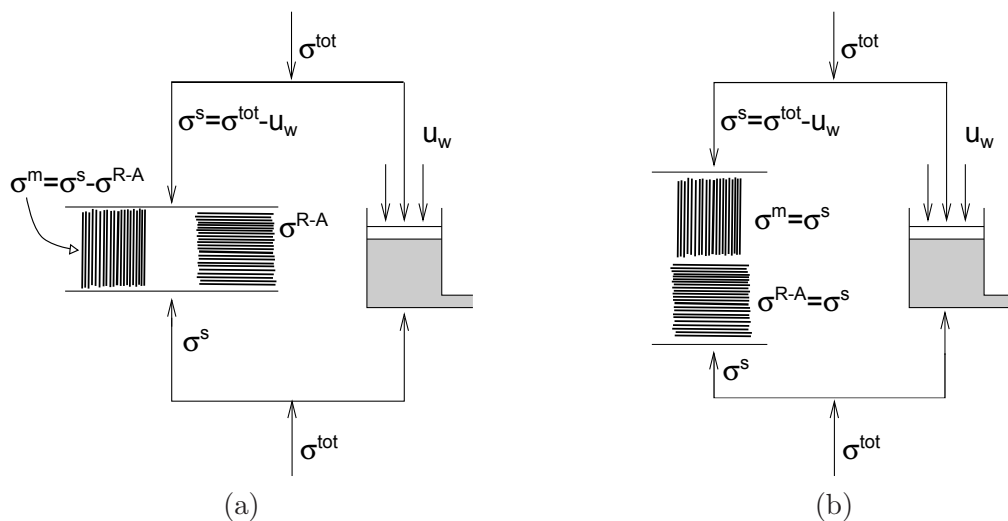

Figure 2: Different models for the stress state within the soil element. (a) parallel connection model, (b) series connection model.

Proposed interpretation of the effective stress in clay aggregates

In order to decide whether the Terzaghi's effective stress principle is applicable to the behaviour of clayey soils, it is necessary to observe in greater detail the pressure field in thin water films between clay particles. This problem has been studied by interface scientists working on colloids. Derjaugin et al. (1987) presented a solution for the mechanical volumetric force balance equation within a film of water between two charged surfaces embedded in an equilibrium solution. They have shown that the pressure normal to the clay particle can be expressed as

$$p_N^p = u_w + p^{R-A} \tag{2}$$

where p_N^p is the inter-particle pressure, u_w is the hydrostatic pressure in the equilibrium solution and p^{R-A} is the disjoining pressure containing, additively, the electrostatic diffuse double layer repulsive forces, the structural repulsive component (active at short distances only) and the attractive London van der Walls forces. For larger particle distances, p^{R-A} diminishes and the inter-particle pressure is equivalent to that in the equilibrium solution, i.e. to u_w. Similar reasoning regarding the influence of pore pressure in shale has been presented by Gonçalvès et al. (2010).

Eq. (2) reveals that u_w forms one component of the overall stress balance in a parallel assembly of particles loaded in the normal direction *irrespective* of the distance between the individual particles. To ensure that the system of particles is in static equilibrium, the inter-particle pressure must be balanced by the total external stress p_N^t, that is $p_N^t = p_N^p$. Therefore,

$$p_N^t = u_w + p^{R-A} \tag{3}$$

which can be rearranged and written in the rate form as

$$\dot{p}^{R-A} = \dot{p}_N^t - \dot{u}_w \tag{4}$$

where the dot is a rate operator ($\dot{x} = \partial x / \partial t$, t being time). Eq. (4) implies that if the rate of total stress $\dot{p}_N^t$ is the same as the rate of pore water pressure $\dot{u}_w$, then $\dot{p}^{R-A} = 0$. Since the p^{R-A} magnitude is, for constant pore water chemistry, dependent only on the inter-particle distance, $\dot{p}^{R-A} = 0$ implies that there is no change in the inter-particle distance and thus no volumetric deformation of the soil.

The above analysis implies that an assembly of clay particles in face-to-face contact loaded in the direction normal to the particle surface satisfies Terzaghi's effective stress principle.

The molecular-scale forces causing a disjoining pressure p^{R-A} then affect the compressibility of the system. By assuming that the system of particles with prevailing edge-to-face contact also satisfies Terzaghi's effective stress principle, it is possible to generate the general effective stress model depicted in Fig. 3. As in the series connection model shown in Fig. 2b, the effective stress in the clay grain assembly is equal to the structural stress calculated by

$$\sigma^s = \sigma^{tot} - u_w \tag{5}$$

Unlike in the series model, however, it is not assume here that the mineral stress σ^m and the inter-particle stress σ^{R-A} are necessarily equal. While the relative distribution of different contact types within the specimen affects mechanical properties of the soil (edge-to-face contacts and face-to-face contacts have different compressibilities and shearing resistances), it is not relevant for the effective stress quantification.
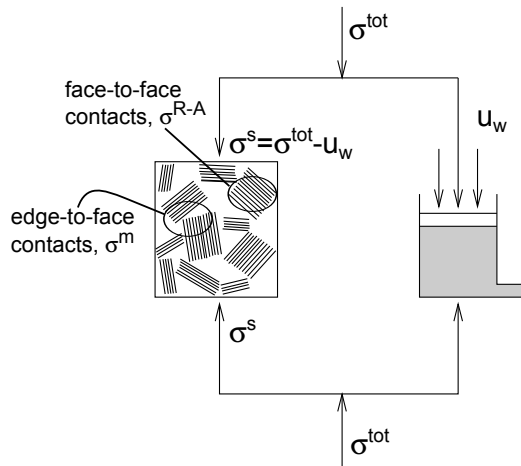

Figure 3: Proposed global description of the stress state within the soil element.

From a constitutive modelling perspective, such an approach to the effective stress allows us to use standard procedures adopted in structured clay modelling. Constitutive models may be defined within the Terzaghi's effective stress space. The relative proportions of the edge-to-face and face-to-face contacts affecting the overall mechanical properties of the assembly, can then be considered by means of *state variables* describing the soil fabric. It is outside the scope of the present paper to list all the possible variables, but the void ratio is the most important one (but not the only one). While void ratio is insufficient to describe the structural arrangement of particles, it is important to characterise the overall density of particles, which affects the relative proportion of edge-to-face and face-to-face contacts. The other suitable variable is the specific surface area, representing the size of the individual particles. As

pointed out by Aylmore et al. (1970), surface area (when measured by nitrogen and carbon dioxide sorption) does not involve the inter-layer space and it is thus representative of the particle size.

When variable fluid chemistry is considered, another state variable that has to be taken into account is *osmotic suction* in equilibrium solution. This does not affect the validity of Eq. (3), but it does control the dependency of the disjoining pressure p^{R-A} on the inter-particle distance, thus controlling the soil compressibility. In addition, *osmotic suction* variation can induce soil deformation under constant effective stress (in the same way as, for example, temperature). With osmotic suction being a state variable in a constitutive model (rather than being part of the effective stress formulation), one can represent the data of Sridharan and Venkatappa Rao (1979) discussed above (i.e., cohesion is dependent on fluid chemistry). Similar reasoning to that outlined in the present paper was proposed by Lu and Likos (2006), but they resorted back to the parallel type model formulation. The interpretation suggested in this work supports the modelling approach adopted by Hueckel (1992b), Kaczmarek and Hueckel (1998) and Loret et al. (2002), who used Terzaghi's's effective stress based constitutive model with additional chemically-induced strain added to the mechanical strain. This work, however, is not restricted to the series model interpretation, which assumes $\sigma' = \sigma^m = \sigma^{R-A}$. Any stress distribution bounded by the limit cases of parallel and series models is admitted , but point out that an understanding of the internal mechanisms is not needed to interpret the soil response in global terms.

It is noted that the diffuse double layer theory leading to Eq. (2) is inaccurate when distances between the clay particle surfaces is very small (< 1.5 nm), as pointed out by Israelachvili and Pashley (1983). This minimum inter-particle distance is thus considered as a possible limit to the validity of the proposed approach. Dry densities corresponding to this limit may be approximately quantified using equation due to Bolt (1956). The void ratio can be calculated using

$$\rho_d = \frac{\rho_s}{1 + \rho_s A_s d_{1/2}} \tag{6}$$

where ρ_s is a soil particle density [kg/m^3], A_s is a specific surface area [m^2/kg] and $d_{1/2}$ is one-half of the separation distance between particles [m]. As indicated by Mitchell and Soga (2005), specific surface area of smectites exclusive of interlayer zones ranges from 50 to 120 m^2/g (the secondary specific surface area that is exposed by expanding the lattice so that polar molecules can penetrate between layers can be up to 840 m^2/g). For our purposes, the A_s exclusive of interlayer zones is relevant. For the upper bound $A_s = 120$ m^2/g, for $\rho_s = 2.7$ g/cm^3 and $d_{1/2} = 0.75$ nm, Eq. (6) yields the dry density of 2.2 g/cm^3. This dry density is higher than the dry densities of clay embedding canisters in the planned nuclear waste

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disposal applications, which are expected in the range between 2 and 2.1 g/cm^3 (Pusch 1982, Pusch 2006). It must however be also pointed out that the assumed parameters (and the Eq. (6) itself) leading to the limitting value of 2.2 g/cm^3 were only approximate and depend on the soil type.

A possible manifestation of this limitation is the experimental data set presented by Bucher and Müller-Vonmoos (1989), who studied compacted bentonites under the condition of zero deformation (and thus a constant value of p^{R-A}) and imposed a pore water pressure increase measuring the total stress response in terms of the ratio $\Delta\sigma^{tot}/\Delta u_w$. The Terzaghi's effective stress implies the ratio $\Delta\sigma^{tot}/\Delta u_w$ to be equal to one. However, Bucher and Müller-Vonmoos (1989) observed that the ratio $\Delta\sigma^{tot}/\Delta u_w$ attained values lower than one, even for dry densities below 2 g/cm^3.

Investigation of the experimental data from available literature

Soils studied

Experimental data on a number of different compacted clays, mostly Na- or Ca-montmorillonitic bentonites, have been used in the investigations. These investigations include five primarily sodium montmorillonitic bentonites, three primarily calcium montmorillonitic bentonites, and one bentonite composed of an interstratified calcium beidellite and kaolinite. Experimental data was obtained from the following sources:

– Avonseal bentonite: Graham et al. (1989).
– FEBEX bentonite: Villar (2002).
– FoCa7 bentonite: Cui et al. (2002), Delage et al. (1998) and Van Geet et al. (2009).
– Bavaria bentonite: Baille et al. (2010).
– Kunigel V1 bentonite: JNC (2000), Komine (2004).
– Volclay bentonite: Komine (2004).
– Kunibond bentonite: Komine (2004).
– Neokunibond bentonite: Komine (2004).
– MX-80 bentonite: Johannesson et al. (1999).

It should be pointed out that different soils were tested in different soil mechanics laboratories, which may cause inconsistencies in the relative comparison of different experimental data sets. Evaluation of the relative measurement error is outside the scope of the present paper. It is to be pointed out, however, that different experiments within a single experimental set (for the given soil) were performed in the same soil mechanics laboratory, different data on

particular soil should thus be directly comparable.

The primary chemical and mineralogical compositions of the soils tested are summarised in Table 1. It is noted that, in some cases, the literature used to extract the experimental data did not contain sufficient information on the soil mineralogical composition. In these cases, composition data was obtained from other sources that described the same or a similar soil. The references used for the soil composition data are cited in the footnote of Table 1.

Table 1: *Mineralogical and chemical composition of the soils studied. M – montmorillonite, B – beidellite, K – kaolinite, CEC – cation exchange capacity.*

bentonite	smectite content [%]	dominant smect. min.	dominant ex. cation	CEC $\left[\frac{meq.}{100g}\right]$	Na $\left[\frac{meq.}{100g}\right]$	Ca $\left[\frac{meq.}{100g}\right]$	Mg $\left[\frac{meq.}{100g}\right]$
Avonseal[1]	79	M	Na	82	41	35	6
Bavaria[2]	60	M	Ca	74		65	
FEBEX[3]	92	M	Ca	111	25	47	36
FoCa7[4]	80	B+K[5]	Ca	69	3	63	
Kunibond[6]	80	M	Ca	80	12	59	7
Kunigel V1[6]	48	M	Na	73	41	29	3
MX-80[7]	83	M	Na	88	67	8	5
Neokunibond[6]	76	M	Na	104	62	33	6
Volclay[6]	69	M	Na	101	57	29	13

[1]JNC (2000); [2]Baille, Tripathy, and Schanz (2010); [3]Villar (2002); [4]Saiyouri et al. (2000); [5]FoCa7 clay composed of kaolinite interstratified with the beidellite in equal proportion (Van Geet et al. 2009); [6]Komine (2004); [7]Karnland et al. (2006).

Reasoning behind experimental data choice and underlying assumptions

The presented investigation only applies to the behaviour of clay aggregates (see Fig. 1), while the behaviour of most compacted clays is influenced by its macrostructure and is thus more complex. For this reason, the choice of experimental data used in the evaluation was restricted.

First of all, the focus was given to the volumetric behaviour. The effective stress framework must also be capable of representing the response in shear; however, the shear response of compacted clay is predominantly controlled by the behaviour of macrostructure (as predicted by the double structure modelling), which is outside the scope of the present paper.

In the investigation, two main assumptions were adopted: the clay aggregate was considered to be fully saturated up to high suction levels and the aggregate response was considered to be elastic. This choice has been supported by investigation of various authors in the past.

Full-saturation of aggregates even at high suction has been suggested by Martin (1960), Gens and Alonso (1992), Alonso et al. (1999), Yong (1999b) and Fityus and Buzzi (2008). For example, Fityus and Buzzi (2008) have shown water retention curves of the natural clay samples indicating ir entry value of suction approximately 10 MPa, while they argue that the microstructural units (aggregates) could theoretically resist air entry up to suctions as great as 80 to 100 MPa. The upper limit of 100 MPa corresponds to the tensile strength of water at 20°C calculated using the van der Waals equation (Marinho et al. 2008). Another argument supporting full aggregate saturation is given in Delage et al. (1998), who observed that the total volume change of the soil specimen during unconfined swelling is exactly equal to the water volume sucked into the sample. If the aggregates were in the unsaturated state, then decrease of matric suction would imply increase of their degree of saturation (following their water retention curve). The pores which were air-filled initially would saturate with water, and thus the total aggregate volume change would be smaller than the volume of water sucked into the sample.

Referring to the reversibility of the mechanical response, Madsen and Müller-Vonmoos (1989) and Pusch (2006) noted that the contraction-expansion process of smectite-rich clays is an almost reversible process since it depends primarily on the hydration potential. Reversibility of the aggregate behaviour has also been suggested by Callaghan and Ottewill (1974), Delage et al. (1998), Tripathy et al. (2002), Cui et al. (2002) and Rao et al. (2004). This conclusion is not general and depends on the particular clay mineralogy. While montmorillonitic aggregates generally show a reversible response, the deformation of illitic clays is not solely governed by diffuse double layer phenomena (Pusch 2006) and the response is to some degree irreversible.

Original experimental data

Unconfined water retention curve The water retention curve (WRC), in unconfined conditions, shows the relationship between the water content and suction at zero net stress. Experimental water retention curves are available for FEBEX, Kunigel V1, MX-80 and FoCa7 bentonites; see Fig. 4. Wetting (that is, "unloading") branches of the water retention curves were evaluated, for consistency with the mechanical unloading tests.

In the data evaluation, the findings by Villar and Lloret (2004), Jacinto et al. (2009) and Romero et al. (2011) were adopted. They observed that the water retention curve is in the higher suction range ($s_t > 2$ MPa for Boom clay by Romero et al. 2011) independent of the global dry density ρ_d^t (solid mass over total soil volume). Della Vecchia et al. (2013) interpreted this finding in the following way: In this suction range the macrostructure is
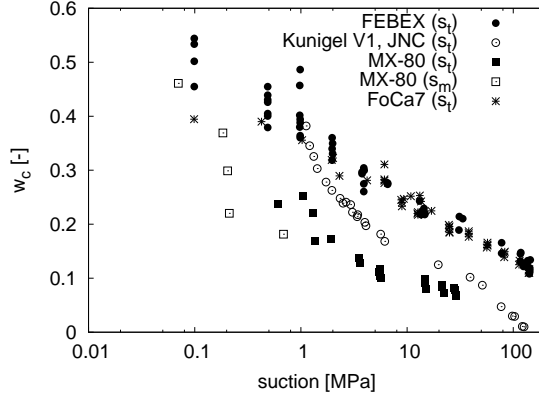
Figure 4: Wetting branches of water retention curves of soils from original experimental data. w_c is the gravimetric water content.

dry, and all water contained within the specimen is present within the clay aggregates. The fact that the water retention curve is (in the higher suction range) independent of ρ_d^t means that the dry density of aggregates is independent of the compaction effort adopted in sample preparation (compaction affects the macropores; recall also that the aggregate response is primarily reversible).

With regard to the saturation of aggregates, an approach adopted by Mašín (2013) was followed. He assumed that the aggregates are saturated, and attributed any water content changes in the WRC measurements to aggregate dry density changes (void ratio), rather than to changes in their degree of saturation. With this assumption, together with the assumption of dry macrostructure, the global water content w_c can be used to calculate the dry density of aggregates ρ_d (solid mass over volume of aggregates) and the void ratio e (volume of pores within aggregates over volume of aggregates) using the following relationships:

$$\rho_d = \frac{G_s}{G_s w_c + 1}\rho_w \qquad e = G_s w_c \qquad (7)$$

where $G_s = \rho_s/\rho_w$ is solid specific gravity, $w_c = m_w/m_s$ is the gravimetric water content, and m_s, m_w, ρ_s and ρ_w are solid and water masses and densities respectively. As noted above, Eq. (7) could theoretically be valid up to the suctions of 80 to 100 MPa. In the evaluation of the experimental data, $G_s = 2.7$ (Mitchell and Soga 2005) and $\rho_w = 1000$ kg/m³ has been adopted throughout. A simplification has thus been introduced here, since the intercrystalline water may have a higher density than the equilibrium solution and the double layer water (Martin 1960, Hueckel 1992a, Yan et al. 1996, Mitchell and Soga 2005, Jacinto et al. 2012). For example, Jacinto et al. (2012) estimated the average water density for MX-80 bentonite

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to vary between $\rho_w = 1080 \text{ kg/m}^3$ (for suction of 1 MPa) and $\rho_w = 1160 \text{ kg/m}^3$ (for suction of 100 MPa). Any uncertainty in G_s was also inglected. It should be noted that an error in the ρ_d evaluation due to G_s uncertainty and ρ_w variation, is in the order of 10% to 20%, which is less than the data scatter typically observed in the experiments (see the data evaluation in Figs. 9 and 10).

Under the assumption of full aggregate saturation, matric suction is equal to the negative pore water pressure, and the Terzaghi's effective mean stress p' is equal to the matric suction s_m. In the higher suction range, suction in the experiments is imposed by controlling the relative humidity of the system, which represents the total suction s_t. In the following, matric suction has been estimated from the experimental data using an approximation $s_m = s_t - s_{\pi 0}$. A single value of $s_{\pi 0} = 0.41$ MPa has been adopted in evaluating all the available data. This value was obtained by Romero (1999) as the osmotic suction of pore water of compacted Boom clay prepared by mixing it with demineralised water. As the osmotic suction is reported to increase with increasing matric suction due to the higher proportion of double layer and equilibrium solution volumes (Krahn and Fredlund 1972, Mata et al. 2002, Miller and Nelson 2006, Stenke et al. 2006 and Thyagaraj and Rao 2010), the values for the effective stresses obtained represent an upper bound for the exact values.

Confined wetting (swelling pressure) tests   The confined wetting test (swelling pressure test) is an important method for characterising the swelling properties of compacted soils, as it resembles the situation encountered in engineered barriers, where the bentonite hydrates under confined conditions. The experiments are typically performed in the as-compacted state at total suctions corresponding to the relative humidity in the laboratory. The soil is soaked, and the resultant total stresses due to confined wetting are measured. Swelling pressure test results are available for FEBEX, MX-80, Bavaria, Avonseal, FoCa7, Kunigel V1, Volclay, Kunibond and Neokunibond bentonites (Fig. 5).

Figure 5 shows the swelling pressure test results plotted in the plane of the initial global dry density ρ_{d0}^t (solid mass over total soil volume at the beginning of the test) with respect to the final swelling pressure at zero matric suction. In the initial as-compacted state, the global dry density does not represent the dry density of aggregates due to the presence of macropores. After saturation, however, experimental observations by different authors suggest that more uniform pore size distribution is attained. For example, Monroy et al. (2010) studied the development of pore size distribution with wetting of a statically dry-of-optimum compacted London clay using mercury intrusion porosimetry. Initially, the porosity distribution was bi-modal, with clearly distinguishable macropores (inter-aggregate pores) and inter-particle
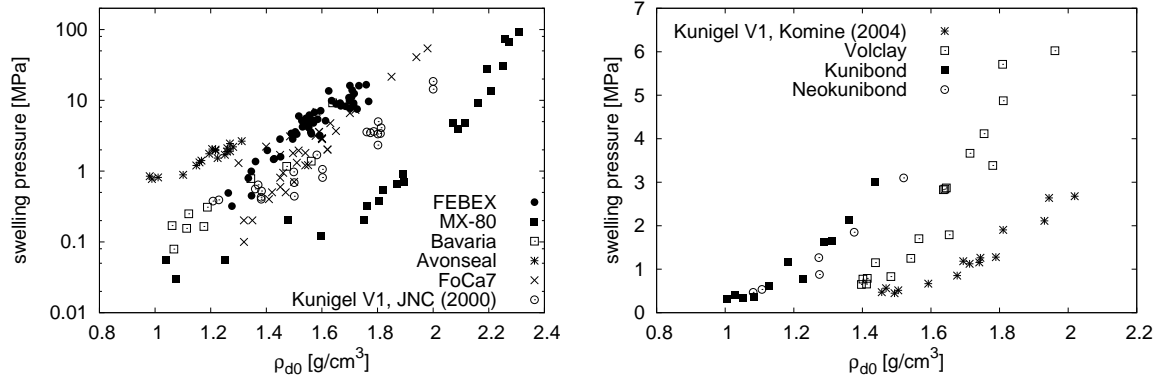
Figure 5: Swelling pressure tests from original experimental data. (a) various sources, (b) data by Komine (2004).

pores (intra-aggregate pores). As the suction decreased, the inter-particle porosity increased, implying swelling of the aggregates. The macroporosity, however, remained largely unaltered. In the last wetting step (wetting from a suction of 40 kPa to 0 kPa), however, the porosity became mono-modal, and the aggregated structure was no longer clearly observable. Therefore at full saturation, the global dry density can be assumed to represent the dry density of the "aggregate". Because the global dry density is constant during the swelling pressure test, the initial global dry density represents the "aggregate" dry density at full saturation. Similar results to Monroy et al. (2010) indicating mono-modal porosity distribution at saturation were reported by Simms and Yanful (2001) (clayey till), Lloret and Villar (2007) (FEBEX bentonite) and Romero et al. (2011) (Boom clay).

From the test specification, matric suction is nil at the end of the swelling pressure test, and the swelling pressure thus represents the effective stress p' within soil specimen.

Mechanical unloading tests on saturated soil The next experiments that were evaluated were mechanical unloading tests on saturated soils or those at a constant suction. Only those experiments starting from high stress levels (of the order of tens of MPa) are relevant for the data evaluation. In this case, the macroporosity is very low and the global dry density (global void ratio) represents the dry density (void ratio) of the aggregates.

The behaviour of aggregated soil during subsequent unloading can be studied with the aid of double-structure models. The model proposed by Mašín (2013) assumes the following function to represent the double structure coupling for the effective stress decrease:

$$f_m = 1 - (r_{em})^{m_c} \tag{8}$$

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In Eq. (8), f_m is the so-called double structure coupling function. $f_m \to 1$ implies that no macrostructural changes occur during unloading (aggregate deformation is equal to the total sample deformation), whereas $f_m \to 0$ implies that the aggregates occlude freely into macropores with no deformation of the macrostructure. r_{em} in Eq. (8) represents the relative void ratio ($r_{em} = 0$ for maximum density state and $r_{em} = 1$ for the loosest packing). m_c in Eq. (8) is a positive valued parameter.

For mechanical unloading from the dense initial state, the model (Eq. (8)) implies $f_m \to 1$, that is, the aggregate deformation is equal to the total sample deformation. Approximate validity of Eq. (8) was assumed in the evaluation of the experimental data. Oedometric unloading test data at saturated conditions are available for FEBEX and Bavaria bentonites (Fig. 6a). Isotropic loading-unloading tests at extremely high cell pressures and different suctions are available for FoCa7 bentonite (Fig. 6b).

Isotropic compression results on reconstituted Avonseal bentonite were also included (Fig. 6a). Comparisons with the Avonseal data should be considered with caution, however; swelling pressure measurements were performed using samples compacted at total suctions corresponding to the relative humidity in the laboratory, whereas bentonite reconstituted in a saturated state was used in the isotropic compression tests. Reconstitution is likely to introduce changes into soil structure and its mechanical response (Fearon and Coop 2000).
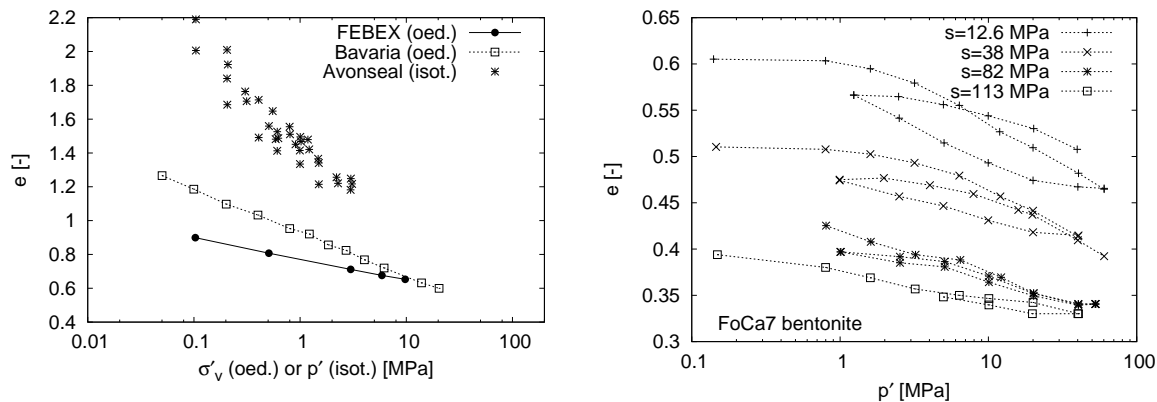

Figure 6: Mechanical unloading and compression tests; original experimental data. (a) different sources, $s = 0$; (b) isotropic tests on FoCa7 bentonite at different suctions.

Evaluation of oedometric unloading tests (that is, tests on FEBEX and Bavaria bentonites) is complicated by the fact that the measured vertical stress does not represent the mean effective stress. For the purpose of the evaluation, the value of K_0 (ratio of horizontal and vertical effective stresses) has been estimated using the Mayne and Kulhawy (1982) empirical

relationship

$$K_0 = (1 - \sin \varphi_c) \text{OCR}^{\sin \varphi_c} \tag{9}$$

where OCR denotes the overconsolidation ratio, calculated as the ratio of initial vertical stress of the unloading experiment (equal to the maximum vertical stress to which the soil was subjected) and current vertical stress σ'_v. $\varphi_c = 25°$, as reported for FEBEX bentonite by Gens et al. (2009), was used in the K_0 estimation using Eq. (9). The value of φ_c was not available for Bavaria bentonite. In the K_0 estimation, FEBEX bentonite value for Bavaria bentonite was used; this should not introduce substantial errors due to the similar composition of the two bentonite types. The effective mean stress can be calculated from the known vertical stress and K_0 estimated using Eq. (9) by

$$p' = \sigma'_v \frac{1 + 2K_0}{3} \tag{10}$$

Test data on Avonseal and FoCa7 clays are at isotropic conditions, where the mean stress is measured directly and thus an estimation of K_0 is not required.

Wetting under constant load tests The last type of experiments evaluated are wetting-induced swelling tests at a constant vertical load. The tests were performed under oedometric conditions on unsaturated soil samples compacted to different known initial global dry densities. The soil was then soaked and left to swell under constant vertical load. The experimental data relate the swelling strain ϵ_s to the sample initial global dry density. Data are available for FEBEX, Kunigel V1, Volclay, Kunibond and Neokunibond bentonites (Fig. 7).

The experimental data were evaluated in the following way: Firstly, it was assumed that at saturation the pore space distribution is mono-modal by the end of the test, similar to the confined swelling pressure tests. The final soil dry density ρ_d thus represents the aggregate dry density at full saturation. The experimental sources, however, are normally presented in terms of the initial global dry density vs. vertical swelling strain. The final soil dry density can be calculated from the initial global dry density ρ_{d0} and swelling strain ϵ_s using a simple relationship $\rho_d = \rho_{d0}/(1 + \epsilon_s)$ (note that under oedometric conditions the measured vertical strain equals the volume strain). As in the case of the oedometric unloading tests, the oedometric conditions imply development of horizontal stresses, which are not measured. However, in this case since the initial stress state does not correspond to the oedometric normally consolidated conditions, K_0 cannot be estimated using Eq. (9). In the evaluation, a fixed value of $K_0 = 1.5$ was adopted and the mean stress was calculated using Eq. (10), and also investigated the sensitivity of the results on the selected K_0 value. The relative
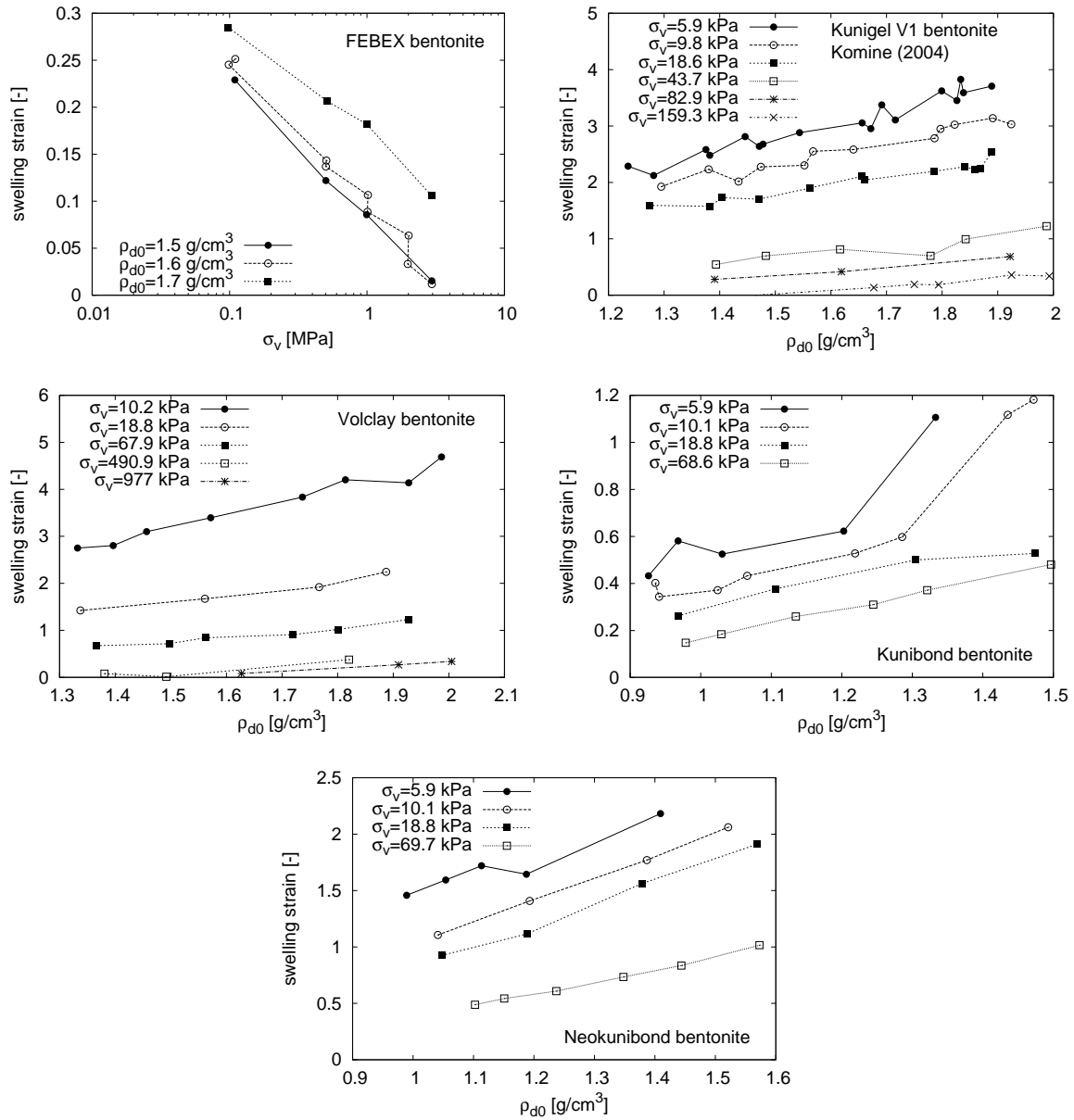
Figure 7: Swelling under constant load, original experimental data.

insensitivity of the results to the K_0 value assumed in evaluation of the swelling during constant load tests is shown in Fig. 8.

Unified interpretation of the experimental results

All the results of the above described experiments have been plotted in the space of mean effective stress p' vs. aggregate dry density ρ_d. Results for all soils and test types are given in Figs. 9 and 10. It can be seen that the data form distinct bands in the p' vs. ρ_d space, although it should be pointed out that the data themselves are relatively scattered. These results are in agreement with findings by Johannesson et al. (1999) and Dueck et al. (2010), who observed equivalence between the swelling pressure and water retention behaviour evaluated under the assumption of full saturation of the aggregate. It has also been discussed in terms of the continuum thermodynamic model by Dueck and Börgesson (2007).

The fact that the data reveal a unique relationship between the mean effective stress p' and aggregate dry density ρ_d has the following consequence. First of all, assumptions supported by investigations of various authors need to be recalled: the aggregate response is primarily reversible, and the aggregates may be considered as saturated up to very high values of suction. The evaluated experiments involve both tests with suction variability at constant total stress and constant suction tests with variable total stress. The unique representation in the p' vs. ρ_d plane means that the soil deformation depends solely on the mean effective stress, which is consistent with Terzaghi's effective stress definition.
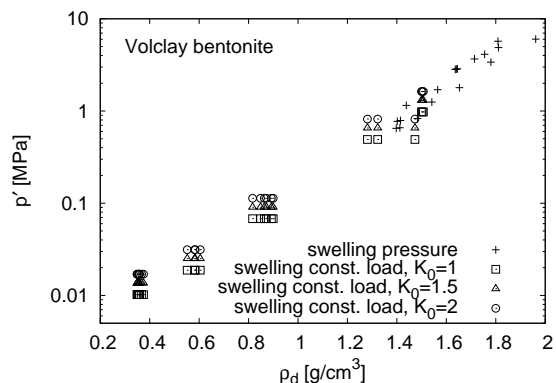

Figure 8: Experimental data on Volclay re-plotted in the ρ_d vs. p' space; sensitivity of results to the assumed K_0 value in swelling under constant load tests.

To combine such a large experimental database into a single-purpose work required obviously a number of assumptions, which were however in the author's opinion reasonably justified by recalling investigations by various authors. Specific assumptions and their possible effect on conclusions may be studied in a more detailed work later. The most important ones were full saturation of aggregates and aggregate response reversibilty. In addition, we assumed that:
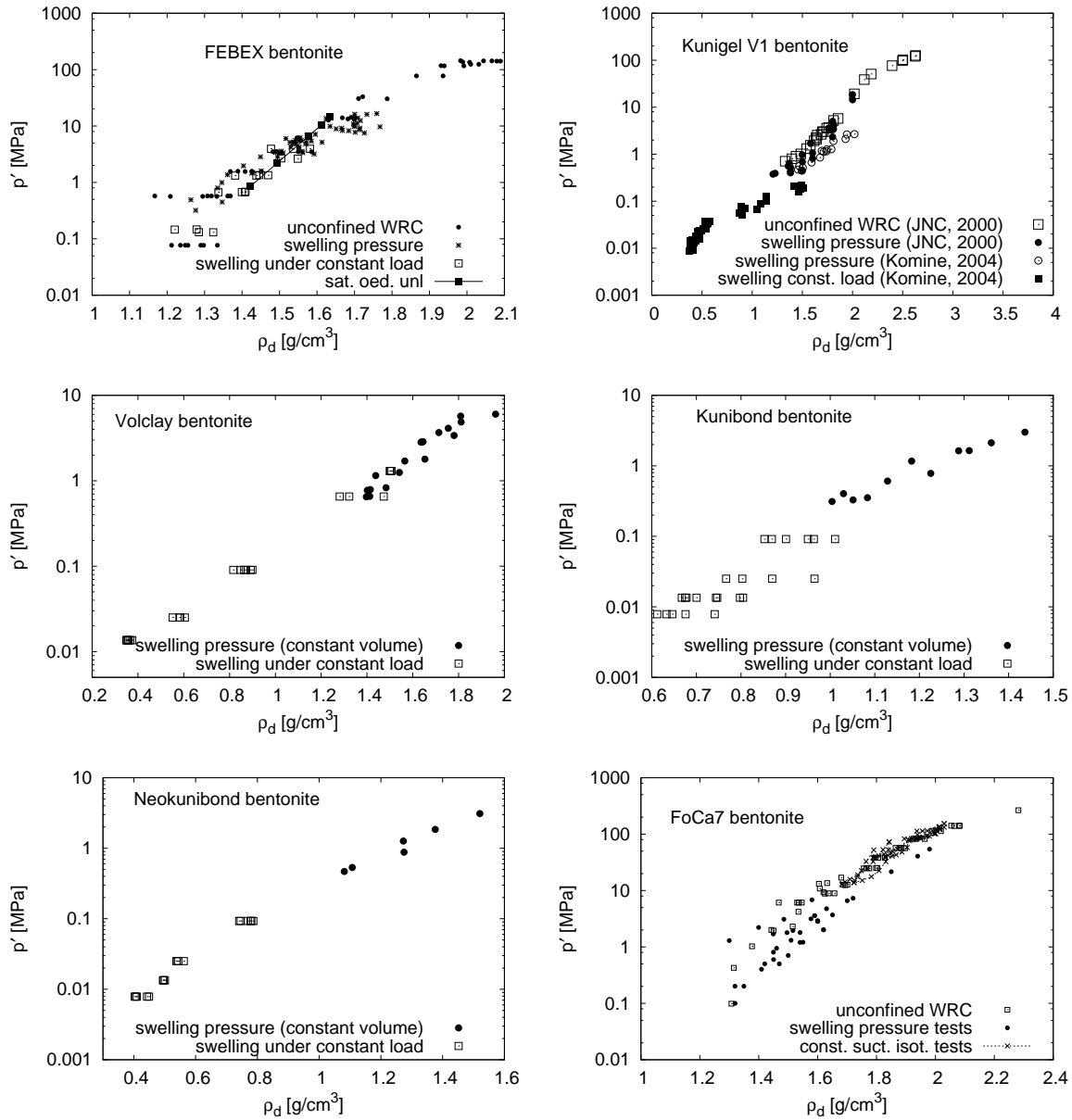
Figure 9: Experimental data re-plotted in the ρ_d vs. p' space, part 1.

different experiment types performed in a single soil mechanics laboratory were consistent with respect to each other; macrovoids were dry in water retention tests at higher matric suctions (approximately $s_t > 2$ MPa); density of water was assumed to be constant ($\rho_w = 1000$ kg/m^3); osmotic suction was assumed to be constant throughout the water retention experiment ($s_\pi = 0.41$ MPa); void distribution was monomodal at the end of swelling pressure
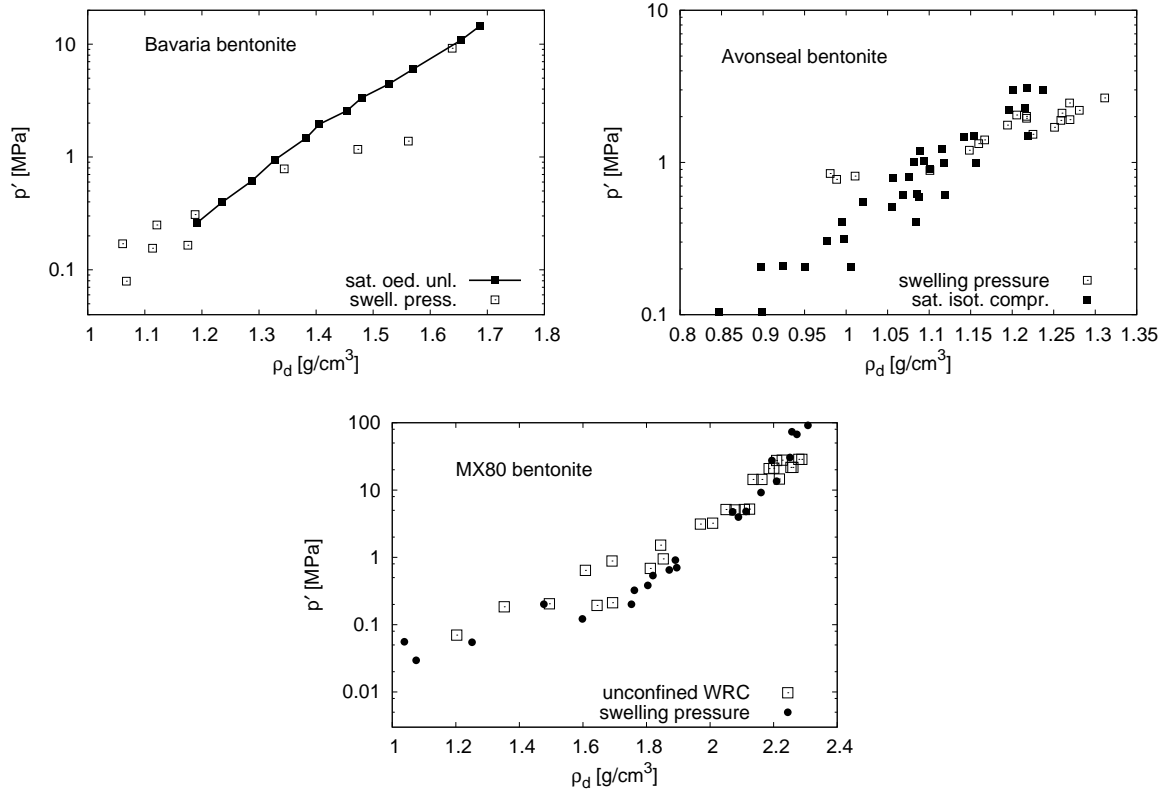
Figure 10: Experimental data re-plotted in the ρ_d vs. p' space, part 2.

tests and wetting under constant load tests; samples for mechanical unloading tests loaded to tens of MPa had initially closed macrovoids; macrovoids in these samples did not open up in unloading; K_0 in oedometric unloading tests could be represented using Mayne and Kulhawy (1982) formula; K_0 in wetting under constant load tests was assumed to be equal to 1.5 (the relatively minor effect of this relatively strict assumption was investigated in a parametric study).

Concluding remarks

In this paper, phenomena occurring between individual clay particles and layers in bentonite soils at the molecular scale were analysed. The work by Derjaugin et al. (1987) implies that the pressure normal to the clay particle predicted by the diffuse double layer theory is equal to the sum of disjoining pressure and hydrostatic pressure in equilibrium solution. This indicates that the Terzaghi effective stress principle is adequate to describe clay behaviour at

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the aggregate level, with the limits coinciding with the limits of validity of the diffuse double layer theory.

To support these conclusions, data were analysed from an extensive experimental database on compacted smectitic clays from the literature. The results are scattered, but there appears to be an interestingly unique relationship between the aggregate dry density (void ratio) and the mean Terzaghi's effective stress (quantified under the assumption of aggregate saturation), for both variable suction and variable total stress experiments. In combination with the assumption of aggregate response reversibility, the experiments support the validity of using the Terzaghi's effective stress to predict the aggregate volumetric deformation. It was also pointed out that the proposed approach was limitted to inter-particle distances larger than approx. 1.5 nm (dry densities up to 2.2 g/cm^3). At smaller distances, the Eq. (2), upon which the developments proposed here are based, becomes inaccurate. The experimental data set given in Bucher and Müller-Vonmoos (1989) is a possible manifestation of this limitation.

Acknowledgment

Notation

A_s specific surface area

$d_{1/2}$ half the distance between clay layers

e void ratio

f_m double structure coupling function of the Mašín (2013) model

G_s specific gravity of mineral grains

K_0 coefficient of Earth pressure at rest; ratio of horizontal and vertical stress

m_c double structure coupling function parameter of the Mašín (2013) model

m_s solid mass

m_w water mass

OCR   overconsolidation ratio

p' mean effective stress

p^{tot} mean total stress

p_N^p total inter-particle pressure

p^{R-A} disjoining pressure

p_N^t external pressure acting normal to the clay particle

r_{em} relative void ratio of the Mašín (2013) model

s_m matric suction

s_t total suction

s_π osmotic suction

u_w pore water pressure

w_c gravimetric water content

ϵ_s swelling strain

φ_c critical state friction angle

ρ_d^t	soil total dry density
ρ_{d0}	soil total dry density at the beginning of the experiment
ρ_d	dry density of the aggregate
ρ_s	density of mineral grains
ρ_w	density of water
σ'	effective stress
σ^m	"mineral stress"
σ^s	"structural stress"
σ^{R-A}	"inter-particle stress"
σ^{tot}	total stress
σ_v'	vertical effective stress
σ_v	vertical total stress

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