Contents lists available at ScienceDirect

Geoderma

journal homepage: www.elsevier.com/locate/geoderma

Towards predicting the soil-specific threshold electrolyte concentration of soil as a reduction in saturated hydraulic conductivity: The role of clay net negative charge

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ARTICLEINFO	A B S T R A C T
A R T I C L E I N F O Handling Editor: Morgan Cristine L.S. <i>Keywords:</i> Dispersion Ionicity Soil stability Net dispersive charge Disaggregation Flocculation	A B S T R A C T The use of marginal quality water with saline and dispersive properties is set to become more prevalent in agricultural production given demands on freshwater resources. The threshold electrolyte concentration (C_{TH}) is often used to define the suitability of such water, with irrigation practitioners seeking a general predictive model based on soil order. Recent work supports that the C_{TH} absolute value is soil-specific, although there remains a requirement to identify the contribution of predictive factors in an effort to create a predictive function for the C_{TH} . This work used 58 soils to explore the variability of the C_{TH} within and between soil orders, subsequently comparing saturated hydraulic conductivity to leachate turbidity as a means to investigate C_{TH} existing prior to the point of spontaneous dispersion. The role of clay net negative charge was investigated in terms of its relation to the C_{TH} in an effort to move towards a predictive capacity for this soil-specific characteristic. Subsequently, it was demonstrated that the C_{TH} was apparently described by net negative charge of clay narticles, suggesting that
	surface interactions between clay particles and the bulk solution are important in governing soil stability from a soil-specific point of view The C_{TH} occurred prior to the aggregate–dispersion threshold and was clearly soil-specific, even within a soil order. Future work is required to incorporate the ionisation potential and clay domain pressure models of soil stability to move towards a C_{TH} predictive function based on quantified differences in soil

mineralogy and charge characteristics, which is discussed.

1. Introduction

Irrigation is vital to the success of modern day society, given demands placed on food and fibre production systems. However, industries will continue to compete for freshwater resources, meaning that groundwater and industrial by-product water will become more important in agricultural production. The interaction between water and soil is complex, due to the fact that water is a polar fluid, and that clay colloids are generally negatively charged via isomorphic substitution and pH dependent protonation-deprotonation processes. Solutes within water sources, irrespective of their origin, affect soilwater interactions, both through a contribution to electrolyte concentration (directly proportional to electrical conductivity; EC) as well as via the suite, charge and hydrated radius of the cations contained in the solute. The former affects the osmotic pressure, while the latter describes electrostatic interaction characteristics; these are not mutually exclusive in their operation and both are responsible for governing soil dispersive processes. Dispersion is considered an irreversible process, in terms of the time it takes soil to regenerate structure (Stockmann et al., 2014), making soil structure a non-renewable attribute of the soil-resource. Hence, the development and choice of soil stability indices guiding practical irrigation using groundwater and industrial by-product water must consider this, and plan accordingly.

Increasing the salinity of the soil-solution has a positive effect on soil structure, via increasing osmotic pressure of a soil solution as the concentration, or salinity, of that solution increases (Cambell et al., 1948). The net result is that for greater soil solution salinity the diffuse double layer DDL thickness decreases, allowing attractive forces between clay particles to dominate and result in a stable soil (Frenkel et al., 1977; Quirk and Schofield, 1955; Scotter, 1985). It is important to note that expansion and compression of the DDL without disassociation of the particles is a form of swelling, albeit within the order of angstrom separation. Therefore, osmotic pressure, and its dynamic, is a key governing factor describing soil structure, and change in the soil pore geometry responsible for hydraulic conductivity. The pioneering work of Quirk and Schofield (1955) demonstrated this whereby the soil

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https://doi.org/10.1016/j.geoderma.2018.08.030

Received 3 April 2018; Received in revised form 20 July 2018; Accepted 31 August 2018 0016-7061/ © 2018 Elsevier B.V. All rights reserved.





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solution electrolyte concentration (EC) reduced dispersivity at high EC, as the DDL development was constrained from inducing dispersion by the osmotic pressure. This is so even in the presence of high sodium concentrations known to induce dispersion at lower EC, and vice versa, which is the basis of the threshold electrolyte concentration (C_{TH}) concept.

The C_{TH} , described by the pioneering work of Quirk and Schofield (1955), lies between absolute stability, which they describe as a potential minima or well, and the threshold turbidity concentration (C_{TU}). The C_{TU} differs substantially from the C_{TH} , and is defined as the point at which spontaneous dispersion is observed in a soil system (Ouirk, 2001), or the aggregate-dispersion boundary condition (Dang et al., 2018a). The classical definition of the C_{TH} is a 10% reduction in hydraulic conductivity from absolute stability, as a measureable departure from the potential well occurring at this stability state. This reduction is considered somewhat arbitrary, in that it is a measureable departure from the stable condition, however, it is should not be considered as a completely arbitrary point. Quirk and Schofield (1955) observed that osmotic pressure reduction and increase within a Ca dominated system did not result in soil structural stability changes; i.e. the spacing between clay platelets did not expand from the minimum spacing condition. Initial changes in the system energy do not result in hydraulic conductivity reduction for a Ca saturated soil. However, adding Na into the system resulted in sufficient change in energy to overcome the potential well, which was subsequently affected by the variations in osmotic pressure and characterised by swelling - intra-crystalline and inter-crystalline swelling - in the clay domain (swelling beyond the potential minima) (Quirk, 2001). Departure from the potential minima is an extremely difficult discrete point to measure, leading Quirk and Schofield (1955) to utilise reduction in saturated hydraulic conductivity (rK_{sat}) as the proxy determinant, and selecting a 10% reduction as the indicator for the CTH position. Latter work by McNeal and Coleman (1966), Ezlit et al. (2013) and Dang et al. (2018a), utilising replicated soil cores, suggested that the C_{TH} was better defined as a 25% (McNeal and Coleman, 1966) and 20% (Ezlit et al., 2013; Dang et al., 2018b) rKsat on the basis of measurement residual error in the replicated samples (Raine et al., 2007).

Importantly, the C_{TH} is distinctly different to the C_{TU} , or aggregation-dispersion boundary condition (Quirk, 2001; Dang et al., 2018a). Fig. 1 demonstrates the initial Sawyers' soil (Rothamsted) from Quirk and Schofield (1955) progressively decreasing in permeability as the EC in the permeate solution is decreased for the given exchangeable sodium percentage (ESP). Numerous literature has confirmed this phenomenon (e.g. Bennett and Warren, 2015; Ezlit et al., 2013; McNeal and

Coleman, 1966; Menezes et al., 2014; Rengasamy et al., 1984; Rowell et al., 1969). Of note, an $rK_{sat} \approx 56\%$ is observed prior to the point that dispersion was observed and the C_{TU} is breached. This is explained by Quirk (2001) as a function of the osmotic pressure and the net available charge of the clay system. Similarly, Dang et al. (2018b) demonstrated rK_{sat} > 80% for Vertisols that had not undergone spontaneous dispersion (i.e. C_{TU} not breached) and further showed that the net negative charge associated with this reduction suggested a stable system. On this basis, and as conceptually demonstrated in Fig. 1, the soil structural condition undergoes three conditions: 1) absolute stability defined by the potential well condition in a Ca dominant system; 2) disaggregation, due to departure from the potential well threshold (C_{TH}) as 2:1 clav lattice intra-crystalline swelling occurs (depending on 2:1 smectitic clay presence) and/or DDL development inter-crystalline swelling occurs; and, 3) clay dispersion, where repulsive pressure overcomes attractive pressure (C_{TU}) allowing clay platelets to be dislocated by Brownian motion. Other factors that physically bond the internal structures of soil aggregates, and the constituents that make up aggregates, affect the point along the line in Fig. 1 — and more generally for any soil — where the aggregates disperse and the C_{TU} is breached.

Practitioners and industry use Government C_{TH} guidelines (e.g. ANZECC, 2000) to make practical irrigation implementation decisions. In essence, this is a form of controlled environmental degradation, where the reduction in saturated hydraulic conductivity is a calculated tolerable risk. The C_{TH} is used as it presents a safety factor for soil structural management. Quirk and Schofield (1955) demonstrated that the C_{TH} occurred at approximately 3–4 times the solution EC of the C_{TU} . Shainberg and Letey (1984) suggest that an $rK_{sat} = 50\%$ would be suitable as a safe irrigation threshold given their assumptions about field versus laboratory measurement, and Menezes et al. (2014) discuss that $rK_{sat} > 80\%$ for a soil with very high initial absolute K_{sat} may still be practically manageable, while Rengasamy et al. (2016) assert, based on their net dispersive charge concept, that the point of zero net dispersive charge (equivalent to C_{TU}) is suitable as an irrigation management threshold. The choice of threshold has very important implications for practical implementation of irrigation regimes (Dang et al., 2018a). If the boundary between an aggregated system and spontaneous dispersion is utilised then small errors may have irreversible dispersive effects on soil structure. On the other hand, the use of an rK_{sat} between the C_{TH} and C_{TU} may be possible with adequate management, assuming some safety factor based on C_{TH} variability. Furthermore, industry C_{TH} guidelines, such as ANZECC (2000), are typically based on a generalised equation for the C_{TH} , which is contrary to the fact the C_{TH} is soil-specific. This is presumably due to the fact there is a dearth of



Fig. 1. Hydraulic reduction curve at ESP 21 with the turbidity concentration (C_{TU}) and threshold electrolyte concentration as defined by Quirk and Schofield (1955) (CTH; 10% reduction in permeability) identified, along with the absolute stability (S_A) and C_{TH} as defined by Ezlit et al. (2013) (20% reduction in permeability). The right hand side depicts the 3-slit pore, clay domain and changes in this as C_{TU} is approached. P_R is the repulsive pressure equivalent to the diffuse double layer, and P_A is the attractive pressures principally governed by London van der Waals forces. The observed reduction in permeability from S_A to C_{TU} (the point of dispersion) is explained as expansion between the overlapped clay platelet sections of the clay domain, resulting in an increase in the volume of the clay domain. Where the repulsive pressures overcome the attractive pressures the

clay domain becomes unstable to the point thermal motion agitates clay platelets out of the domain resulting in dispersion; these dispersed clay platelets form a stable suspension of dispersed clay that subsequently can move with the permeate solution, giving rise to greater and increasing permeability reduction. Of note, point B represents a reduction in permeability of \approx 56%.

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