

1 **Reduction of hydraulic conductivity and loss of organic carbon in non-dispersive soils of**
2 **different clay mineralogy is related to magnesium induced disaggregation**

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7 **Abstract**

8 Irrigation water containing appreciable amount of magnesium (Mg) is increasingly used in
9 farming. However, studies on the effect of Mg on soil permeability and structural stability are
10 limited and the results are contradicting. In this study, four soils of different dominant clay
11 mineralogy, specifically kaolinite, illite and montmorillonite, were used to investigate the
12 effect of Mg on saturated hydraulic conductivity (K_{sat}), interaction with organic carbon and soil
13 clay dispersion and to compare to the effect of Ca. The soils were packed in columns and
14 leached with four groups of either Ca-Ca, Ca-Mg, Mg-Mg or Mg-Ca solutions at the successive
15 concentrations of 0.5, 0.25, 0.1, 0.05, 0.025, 0.01, 0.005, 0.001 and 0.0001 M, and finally with
16 deionised water. K_{sat} was measured and leachates were assessed for dissolved organic carbon
17 (DOC) and dispersion as turbidity. The leached soils were tested for aggregate stability,
18 mechanical dispersion and zeta potential. This study showed that Mg had a disaggregation
19 effect on soil structural stability. As compared to Ca, K_{sat} on Mg treated cores were lower over
20 a wide range of concentrations, desorption of DOC and turbidity from Mg clay surface was
21 higher, although the dispersion in leachates was insignificant under both Mg and Ca treatments.
22 The effect of Mg treatment was more pronounced on smectic and illitic soils. The decrease in
23 K_{sat} was related to Mg induced disaggregation, due to intra-crystalline swelling and/or the inter-
24 crystalline swelling in Mg treated soil clays.

25 **Keywords**

26 Magnesium; calcium; saturated hydraulic conductivity; dissolved organic carbon; turbidity.

27 **1. Introduction**

28 Pore shape, size distribution and pore continuity are key factors affecting the function of a soil,
29 which directly influence the movement of water, expressed as permeability. It is well known
30 that the concentration and the composition of the percolating solution has a great influence on
31 the structural stability and the hydraulic conductivity (K) of soil (Ali et al., 2018; Aringhieri
32 and Giachetti, 2001; Dang et al., 2018a; Dang et al., 2018b; Levy and Van Der Watt, 1990;
33 Pupisky and Shainberg, 1979; Quirk and Schofield, 1955). In most soils, the deterioration of
34 structure and hydraulic conductivity occurs when electrolyte concentration decreases (de
35 Menezes et al., 2014; Ezlit et al., 2013; Jayawardane, 1979; McNeal and Coleman, 1966)
36 and/or monovalent ions increase (Bennett et al., 2016b; Jayawardane et al., 2011; Rengasamy
37 and Marchuk, 2011). There are emerging examples that irrigation waters containing
38 appreciable amount of magnesium (Mg) are used in farming, due to the scarcity of good quality
39 irrigation water (Karimov et al., 2009; Qadir et al., 2018; Skilton et al., 2000; Vyshpolsky et
40 al., 2008). However, to date, there are only a few studies on the effect of Mg on soil structural
41 stability and saturated hydraulic conductivity (K_{sat}), as relative to calcium (Ca) and sodium
42 (Na) effects (Curtin et al., 1994; Dontsova and Norton, 1999; Emerson and Chi, 1977; Keren,
43 1991; Reading et al., 2012; Rengasamy et al., 1986). The results of these studies suggested Mg
44 itself can decrease soil structural stability, which is the definition of “Mg specific effect”
45 referred to in the literature (Curtin et al., 1994). However, decrease in structural stability is
46 somewhat ambiguous, as slaking, dispersion and disaggregation (expansion of clay domain
47 without dispersion) all contribute to structural instability. Disaggregation is a state in between
48 absolute stability and dispersion, where soil aggregate does not spontaneously disperse. When
49 the ionic strength of the equilibrium solution is decreased, the repulsive pressure increases
50 while the attractive pressure decreases due to the development of diffuse double layers.

51 Consequently, the distance between the clay particles increases. Soil disaggregation can also
52 occur due to the intra-crystalline swelling in the presence of 2:1 smectitic clay.

53 The specific effect of Mg has been observed for the K_{sat} of illite and “occasionally”
54 montmorillonite soils, but is suggested to rarely affect kaolin soils (Alperovitch et al., 1981;
55 Keren, 1991; Quirk and Schofield, 1955; Rahman and Rowell, 1979; Zhang and Norton, 2002).

56 The lower effect on K_{sat} in Kaolinitic soil, than for other soils, may be related to the common
57 existence of sesquioxides in kaolinite soils or soils being acidic (Arias et al., 1995; Frenkel et
58 al., 1978; McNeal and Coleman, 1966; McNeal et al., 1968), although any definitive reasoning
59 is unclear. Rahman and Rowell (1979) reported that Mg had no specific effect on
60 montmorillonite soils, arguing that K_{sat} is simply controlled by exchangeable sodium
61 percentage (ESP) and solution concentration, and clay swelling is the dominant process causing
62 structural deterioration. In contrast, exchangeable Mg (in the presence of Na) has been shown
63 to have a specific effect on K_{sat} of a non-calcareous montmorillonite soil, but not a calcareous
64 one (Alperovitch et al., 1981). This observation does not refute the work of Rahman and Rowell
65 (1979), but tends to suggest that Mg exacerbates the effect of Na. Later, Keren (1991) reported
66 that Mg had a significant effect on erosion and infiltration rate when montmorillonite soils
67 were exposed to rainfall regardless of CaCO_3 content in soils, while Zhang and Norton (2002)
68 reported a specific Mg effect, compared with Ca, on the K_{sat} and clay dispersion of two humid
69 soils containing smectite, kaolinite, vermiculite and illite. It is clear that the specific effect of
70 Mg is not well established in terms of its mechanism, its operation, or the scale on which it
71 might act. We suggested that the specific effect of Mg is mainly due to its induced
72 disaggregation. It means that physical/chemical bonds are potentially able to overcome the
73 effect, which could explain the observation with CaCO_3 containing soils. Surface erosion does
74 not negate this reasoning, as montmorillonitic soils in an unconfined environment, such as the
75 surface where erosion occurs, are prone to instability when subject to kinetic energy and the

76 energy of wetting. Hence, further investigation is required to elucidate the extent and
77 mechanism of Mg specific effect on the K_{sat} of soils with the consideration of their mineralogy.
78 There is a paucity of literature concerning the interaction between Mg and organic matter in
79 terms of soil structural stability. In soil aggregates, both organic matter and clay contribute to
80 the cation exchange capacity (CEC) of soils, and their interactions have a major influence on
81 soil structural stability (Nelson and Oades, 1998). Organic matter may enhance or prevent soil
82 dispersion depending on its nature. Goldberg et al. (1990) found that adding dissolved organic
83 matter enhances clay dispersion, and claimed that the positive effect of organic matter on soil
84 structure probably occurs through binding of soil particles by roots and hyphae at the aggregate
85 level, but at the clay-particle level the negative charge of organic anions enhances clay
86 dispersion. Other researches indicated that when organic matter is high in soils, water
87 interaction leading to clay dispersion is minimal because charge on clays is reduced by clay-
88 organic bonds, which are mostly covalent, or soil aggregates are enveloped by organic
89 materials formed by covalent bonding (Marchuk and Rengasamy, 2012; Marchuk et al.,
90 2013a). Marchuk et al. (2013a) observed that NaCl caused higher dissolution of organic carbon
91 than KCl in soils containing smectite and illite-kaolinite, which was in line with the finding
92 that exchangeable sodium led to higher clay dispersion than potassium. However, there is
93 limited literature existing on this same phenomenon for Mg, as far as the authors are reasonably
94 aware. Therefore, investigation into the interaction of Mg with soil organic matter is prudent
95 in driving our understanding of soil structural dynamics.

96 This study aimed to investigate the impact of Mg effect on the K_{sat} of soils, as compared to Ca
97 treatment at the equivalent condition, whereby soil clay mineralogy was contrasting, *i.e.*
98 montmorillonite, illite and kaolinite. Secondly, the investigation aimed to explore the
99 interaction between permeate Mg and soil organic carbon by measuring the dissolved organic
100 carbon (DOC) from the collected leachate. The hypotheses of this study are that: 1) Mg reduces

101 K_{sat} , as compared to Ca, induced by disaggregation, rather than dispersion; 2) the difference
102 between the K_{sat} of Mg and Ca varies dependent on clay mineralogy and soil texture; and, 3)
103 DOC concentrations from Mg leached samples are greater than those from Ca leached samples,
104 with this related to reduction in K_{sat} , but not dispersion of soil primary particles.

105

106 **2. Materials and methods**

107 2.1 Soil used

108 To control for the effect of pH on soil clay dispersion and hydraulic conductivity, four soils
109 with neutral pH were selected. All four soils were collected from 0–10 cm depth at each site.
110 Soil samples were air-dried, carefully ground and sieved to pass a 2 mm threshold. The physical
111 and chemical properties for the soil samples are presented in Table 1. Soil pH and electrical
112 conductivity (EC) were measured in a 1:5 soil/water suspension. The exchangeable cations
113 were determined using method 15A2 as per Rayment and Lyons (2011). The effective cation
114 exchange capacity (CEC_{eff}) was calculated as a sum of exchangeable Na, K, Mg and Ca.
115 Sodium absorption ratio (SAR), cation ratio of soil structure stability (CROSS), exchangeable
116 sodium percentage (ESP) and exchangeable dispersive percentage (EDP) were calculated
117 according to Rengasamy and Marchuk (2011) and Bennett et al. (2016a). Particle size
118 distribution was determined using the method of Gee and Bauder (1986). Water-dispersible
119 clay was measured in the same procedure as particle size analysis without any pre-treatment or
120 chemical addition (FAO, 2014). Clay mineralogy was examined using X-ray diffraction: Soil
121 M4 was classified as a Black Vertisol and was montmorillonite dominant; Soil D6 was
122 classified as a Black Vertisol and was montmorillonite and kaolinite dominant; the Urrbrae
123 soil was a Red Chromosol that was illite dominant, with traces of kaolinite and quartz; while
124 the Timberlea soil was classified as a grey Chromosol and the dominant mineral was kaolinite
125 with traces of illite.

126

Table 1 Selected soil physical and chemical properties and locations

<i>Soil</i>	<i>units</i>	<i>M4</i>	<i>D6</i>	<i>Urrbrae</i>	<i>Timberlea</i>
Depth	<i>cm</i>	0–10	0–10	0–10	0–10
pH		7.3	6.7	6.7	6.3
EC	<i>dS/m</i>	0.18	0.19	0.15	0.05
Exchangeable Na	<i>cmol/kg</i>	1.21	0.43	0.12	0.11
Exchangeable K	<i>cmol/kg</i>	0.94	0.42	0.67	0.37
Exchangeable Mg	<i>cmol/kg</i>	16.61	7.26	0.91	1.07
Exchangeable Ca	<i>cmol/kg</i>	26.33	13.17	6.41	2.00
CEC _{eff}	<i>cmol/kg</i>	45.08	21.29	8.10	3.56
ESP	<i>%</i>	2.68	1.2	1.44	3.16
EDP	<i>%</i>	5.21	2.9	6.46	10.13
Organic carbon	<i>%</i>	1.02	1.01	1.50	1.54
Dominant clay mineral ^a		M	M, K	I	K
Taxonomic class ^b		Black Vertosol	Black Vertosol	Red Chromosol	Grey Chromosol
Texture		Clay	Sandy clay	Sandy clay loam	Sandy loam
Clay content	<i>%</i>	56	36	29	15
Silt content	<i>%</i>	14	8	17	17
Sand content	<i>%</i>	30	56	54	68
Water-dispersible clay	<i>%</i>	35	20	13	8
Location		Dalby, QLD	Dalby, QLD	Adelaide, SA	Gore, QLD

127

^a I, illite; K, kaolinite; M, montmorillonite

128

^b Soil classification according to Australian soil classification (Isbell, 1997; 2016)

129

130 2.2 Hydraulic conductivity

131 Soil columns were prepared by packing 85 g of sieved soil (air dried, <2 mm) into a plastic
132 cylinder (10 cm length, 4.25 cm diameter). Whatman No. 1 filter papers were placed beneath
133 and above the soil surface of each soil column. To investigate the effect of Mg on K_{sat} as
134 compared to Ca, four groups of solutions, labelled as G1, G2, G3 and G4, were applied. The
135 columns were wetted from the bottom using the baseline solutions, *i.e.* G1 and G2, 0.5 M of
136 CaCl_2 ; G3 and G4, 0.5 M of MgCl_2 , and kept saturated. The bulk density (wet) for soil column
137 M4, D6, Urrbrae and Timberlea was 0.93, 1.11, 1.12, 1.33 g/cm^3 , respectively. The columns
138 were then leached with 1.0 litre (appro. 25 pore volumes) of 0.5 M of MgCl_2 or CaCl_2 for better
139 equilibrium of Mg or Ca with soils and to eliminate the effect of exchangeable Na and K on
140 K_{sat} . The solution was applied with a constant hydraulic head of 30 mm for each column.
141 During leaching, montmorillonite clays underwent initial intra-crystalline swelling, with the
142 change in boundary condition measured, and the head height readjusted. Any deviation from
143 this head height was recorded in order to ensure that hydraulic calculations were sufficiently

144 reflective of the changed conditions. Subsequently, the soil columns were leached with a series
 145 of percolating solutions with decreasing electrolyte concentration: 0.25 to 0.0001 M CaCl₂ for
 146 G1, 0.25 to 0.0001 M MgCl₂ for G2, 0.25 to 0.001 M MgCl₂ for G3 and 0.25 to 0.0001 M
 147 CaCl₂ for G4, as shown in Table 2. Each treatment solution was of 1.0 litre. As a final solution
 148 designed to result in rapid dilution, 1.0 litre of deionised water was used to leach each soil. The
 149 volume of 1 litre was sufficient to ensure chemical breakthrough and steady state conditions
 150 has been reached. The K_{sat} was recorded and calculated using Darcy's equation. Twelve soil
 151 columns were prepared for each group, including three columns for K_{sat} measurement and nine
 152 further columns for destructive measurement of exchangeable cations after each leaching
 153 solution.

154 Table 2 The ionic composition and concentration of baseline treatment solutions and subsequent leaching
 155 solutions used to treat four soils of different clay mineralogy

Group No.	Baseline treatment	Subsequent leaching solutions								Final solution
G1	0.5 M CaCl ₂	CaCl ₂ concentrations in leaching solutions (M)								DI water
G4	0.5 M MgCl ₂	0.25	0.1	0.05	0.025	0.01	0.005	0.001	0.0001	
G2	0.5 M CaCl ₂	MgCl ₂ concentrations in leaching solutions (M)								
G3	0.5 M MgCl ₂	0.25	0.1	0.05	0.025	0.01	0.005	0.001	0.0001	

156

157 2.3 Leachate characterisation

158 The leachate of each treatment solution was collected periodically and the turbidity of the
 159 leachate was measured using a Hach Turbidimeter 2100N. Then the leachates were filtered
 160 through 0.45 µm filter paper and DOC concentration was measured using a TOC-L Shimadzu
 161 Total Organic Carbon Analyzer.

162 2.4 Exchangeable cations of treated soils

163 After completion of each percolating solution for each treatment, the soil was removed from
 164 the column, air-dried, crushed, and passed through a 2-mm sieve. The EC of the treated soils
 165 was measured at soil water ratio of 1:5. The exchangeable cations were extracted according to
 166 the method 15A2 as described in Rayment and Lyons (2011) and measured on ICP-MS.

167 2.5 Critical flocculation concentration of soil clays

168 Clay suspensions (<2 microns), separated from each soil without chemical treatment (Jackson,
169 2005), were equilibrated in 1 M MgCl₂ or CaCl₂ solutions. The excess salts were removed
170 using the dialysis method until EC<0.05 dS/m. 30 ml of clay suspension (1.0 g/L) was pipetted
171 into a 50 ml falcon tube. The initial turbidity of each sample was measured. A series of
172 concentrations of MgCl₂ or CaCl₂ solutions were then added into Mg or Ca clay suspension
173 respectively. The critical flocculation concentration (CFC) was recorded as the sum of the
174 addition of MgCl₂ or CaCl₂ at the point the turbidity of clay suspension was less than 5% of
175 the initial turbidity at a depth of 5 cm in 4 h.

176 2.6 Soil dispersion assessment after leaching with DI water

177 The soils were collected and air-dried after DI water leaching. Aggregate stability test was
178 conducted in order to assess soil aggregate dispersion (Loveday and Pyle, 1973). The soil
179 aggregates were collected from the middle section of soil core, and then carefully immersed
180 into DI water. A visual judgment (scores) was made according to the degree of dispersion. In
181 addition, mechanical dispersion test was performed to examine soil dispersive behaviour under
182 external energy input (Rengasamy, 2002). The turbidity of the colloidal sample was measured.
183 The zeta potential was measured on dispersed clays using a Malvern Zetasizer Nano-ZS
184 analyser.

185 2.7 Statistical analysis

186 The average value of K_{sat} for each soils at each leaching solution was plotted with standard
187 deviation as error bar. The significant differences between Mg and Ca treatments were
188 investigated using ANOVA single factor test, in terms of DOC values, turbidity and zeta
189 potentials. Statistical significance was established as $\alpha<0.05$. Statistical tests were performed
190 with Minitab software.

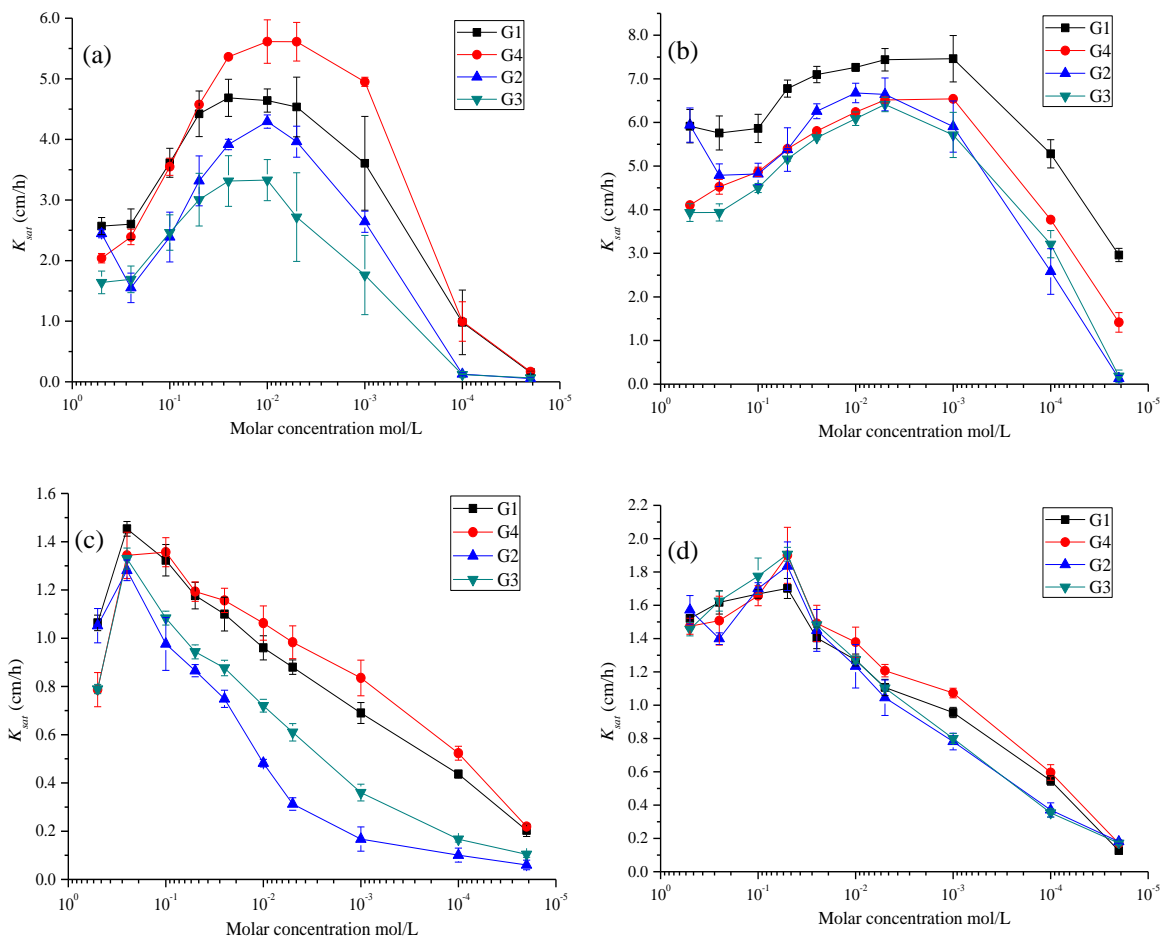
191 **3. Results and discussion**

192 3.1 Exchangeable cation percentage after each treatment

193 The results of exchangeable cation percentage after each treatment of the four soils are shown
194 in Appendix A.1. The exchangeable calcium percentage (ECP) or exchangeable magnesium
195 percentage (EMP) were greater than 91% after the baseline solution treatment, depending on
196 the cation type of the treatment solution. The ECP values of G1 and the EMP values of G3
197 remained largely unchanged during following treatments even after leaching with deionised
198 water. For G2 (Ca-Mg), after leaching with 0.25 M MgCl_2 solution, the ECP dropped from
199 >91% to <4% while EMP surged >91%. In contrast, for G4 (Mg-Ca), after percolating with
200 0.25 M CaCl_2 , EMP decreased to <6%, while ECP increased to >91% and maintained at this
201 level for the remaining leaching at lower concentrations. In general, the amount of
202 exchangeable Na and K was lower than the untreated soils after baseline leaching and
203 throughout the following leaching.

204 3.2 Evaluation of hydraulic conductivity

205 The K_{sat} of the four soils after each leaching treatment is shown in Fig. 1. After baseline
206 treatment (0.5 M), the K_{sat} values of all Mg treated soils were lower than that of Ca treated soils,
207 regardless of clay mineralogy. An initial increase of K_{sat} was observed for all soils when
208 leached with successive solutions of MgCl_2 (G3, Mg-Mg) or CaCl_2 (G1, Ca-Ca). The greatest
209 K_{sat} of the Mg treated M4 soil column 3.33 cm/hr was found when 0.01 M MgCl_2 was used,
210 which was twice the baseline K_{sat} value of 1.64 cm/hr. The greatest K_{sat} for the Ca treated M4
211 soil was found at 0.005 M CaCl_2 , which increased by 80% compared to its Ca baseline K_{sat} .
212 The D6 soil, like the M4 soil, was classified as a Vertosol, having the greatest K_{sat} value at
213 0.005 M in the Mg treatment and 0.001 M in the Ca treatment, increasing by 63% and 27%
214 compared to the equivalent baseline K_{sat} value, respectively. For the Urrbrae soil, the initial
215 increase of K_{sat} ceased after leaching with 0.25 M solutions and was 25% (Mg) and 37% (Ca)
216 greater than baseline values. An initial increase was also found in the Timberlea soil with the
217 greatest K_{sat} reached at 0.05 M, for both the Ca (12% greater) or Mg (25% greater) treatment.



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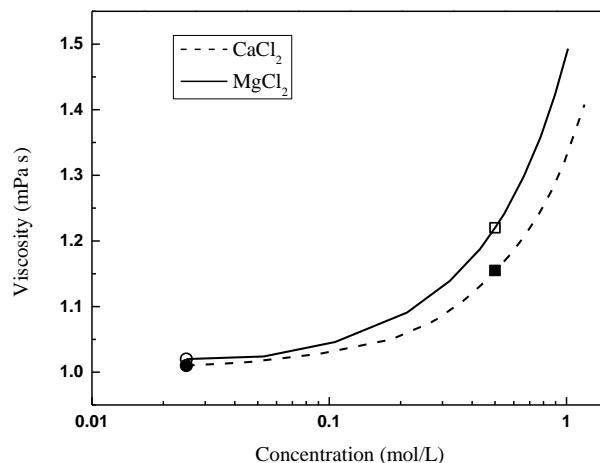
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221 Fig. 1. The K_{sat} (cm/h) against molar concentration of percolating solutions (log scale) of four soils leaching with
 222 four groups of solutions (a) M4 soil, (b) D6 soil (c) Urrbrae soil (d) Timberlea soil

223

224 The initial increase of K_{sat} can be related to the decrease of viscosity of $MgCl_2$ and/or $CaCl_2$
 225 leaching solutions. The K_{sat} is inversely proportional to the viscosity of the leaching solution
 226 and is proportional to the intrinsic permeability of the porous media (k) (Artiola et al., 2004;
 227 Jarsjö et al., 1997). According to Haynes (2014), when the concentration of $MgCl_2$ and $CaCl_2$
 228 solutions decreases from 0.5 to 0.025 M, their respective viscosity decreases from 1.22 to 1.02
 229 mPa·s and 1.16 to 1.01 mPa·s. At even lower concentration of $MgCl_2$ and $CaCl_2$, the viscosity
 230 of solutions remained largely unchanged and close to that of pure water, *i.e.* 1.00 mPa·s, as
 231 shown in Fig. 2. Therefore, when the viscosity of percolating solution was decreased by 12%

232 (e.g. MgCl_2 from 0.5 to 0.05 M), K_{sat} was expected to increase to a similar degree, if the k value
 233 of soil was stable. The results showed that the K_{sat} values of all soils under either Mg or Ca
 234 treatment increased by more than 12%, with the magnitude of K_{sat} increase, and the
 235 concentration where the greatest increase occurred, varying with the soil and the cation type
 236 used in the leaching solution. It is noted that the viscosity decrease magnitude was less than
 237 the K_{sat} increase magnitude, suggesting there is a further mechanism acting. That is, Mg and
 238 Ca leaching may improve soil structure, due to greater covalent bonding formed between the
 239 divalent cation Mg (or Ca) and clay particles by replacing exchangeable monovalent cations
 240 Na and K (Marchuk and Rengasamy, 2011; Marchuk et al., 2013b). There were negligible
 241 amounts of exchangeable Na and K left on exchange sites after Mg and Ca treatments.
 242 Consequently, the k and K_{sat} values were increased.



243
 244 Fig. 2. The viscosity (mPa·s) of MgCl_2 and CaCl_2 solution of different concentrations at 20 °C. □ 0.5M MgCl_2 ; ■
 245 0.5M CaCl_2 ; ○ 0.05M MgCl_2 ; ● 0.05M CaCl_2 . The viscosity of pure water at 20 °C is 1.002 mPa·s. Data extracted
 246 from (Haynes, 2014).

247
 248 However, in Urrbrae soil, the K_{sat} started to decrease when the ionic strength of the leaching
 249 solutions was <0.25 M. The K_{sat} value of Urrbrae soil was still maintained above or equal to
 250 the initial baseline (0.5M), even with 0.025 M solution. A rapid reduction in K_{sat} was observed
 251 after leaching with 0.001 M solution, which was in good agreement with the measured CFC of

252 Urrbrae soil (Table 3). In addition, rapid reduction of K_{sat} was found in M4 and D6 soils when
 253 leached with 0.0001 M solution, and 0.001 M for Timberlea soils, which was in good
 254 agreement with the measured CFC for each of the soils.

255 Table 3 The critical flocculation concentration (mmol) of homoionic Mg and Ca soil clays

	M4	D6	Urrbrae	Timberlea
Mg	0.66	1.08	2.73	2.73
Ca	0.57	0.85	1.62	1.89

256

257 When DI water was used, the K_{sat} of the M4 soil was reduced to less than 2% of the baseline
 258 K_{sat} . Reduction in K_{sat} of the D6 soil was evident in both G1 and G3 treatments after leaching
 259 with 1.0 L of DI water. When the electrolyte concentration decreased, divalent montmorillonite
 260 crystals had limited crystalline swelling due to the hydration of ions and surfaces of the clay
 261 platelets (<1.9 nm), which was supported by Norrish and Quirk (1954). However, the work
 262 from Posner and Quirk (1964) showed that for Ca and Mg saturated montmorillonite, the
 263 change in basal spacing from 1 to 1.55 nm is not affected by the electrolyte concentration,
 264 whereas the transition from 1.5 to 1.9 nm can be regarded as an osmotic effect depending on
 265 the charge density and origin in montmorillonite. The osmotic repulsive pressure was much
 266 less for divalent cations than monovalent cations, hence divalent montmorillonite particles
 267 separated by large distances (>2.5 nm) can collapse together if this force is insufficient to
 268 counterbalance the van der Waals attractive forces (Norrish, 1954). Therefore, the volume
 269 change of the clay beyond the crystal swelling is attributed to the osmotic phenomenon and is
 270 associated with the interactions of the diffuse double layers (Schanz and Tripathy, 2009; Van
 271 Olphen, 1977). Unlike montmorillonite, illite and kaolinite are non-expanding clays. Hence,
 272 the osmotic swelling due to the ionic concentration gradient between the clay surface and the
 273 pore water was the main mechanism in explaining the reduction of K_{sat} . Montmorillonite soils
 274 (both M4 and D6 soils) had the greatest reduction in absolute K_{sat} value, compared to the illite
 275 and kaolinite soils, which was related to the great affinity to adsorb water (swelling capacity)

276 of montmorillonitic soil clay, due to its expanding nature, larger surface area and CEC than
277 other clay minerals.

278 The difference in structure and hydraulic conductivity between the M4 and D6 soils could be
279 related to the clay texture and the abundance of montmorillonite clay. The D6 soil contained
280 20% less clay and 26% more sand than the M4 soil. Furthermore, the clay minerals in D6 are
281 a mixture of montmorillonite and kaolinite, rather than montmorillonite only in M4. Upon
282 soaking of CaCl_2 or MgCl_2 baseline solutions, the soil surface rose approximately 13 mm in
283 height for the M4 soil, while this increase was only 4 mm for the D6 soil. This difference in
284 expansion is in general agreement with the relative greater abundance of montmorillonite clay
285 in the M4 soil.

286 For the other two treating groups (G2 Ca-Mg and G4 Mg-Ca), the K_{sat} values are also plotted
287 in Fig. 1. When Ca-saturated soil was leached with 0.25 M MgCl_2 solution (G2, Ca-Mg), an
288 evident reduction in K_{sat} was found as compared to that leached with 0.25 M CaCl_2 (G1, Ca-
289 Ca). To the contrary, when Mg-treated soils were leached with CaCl_2 solution (G4, Mg-Ca),
290 clear increase in K_{sat} was found as compared to that leached with MgCl_2 solution (G3, Mg-Mg).
291 During successive leaching, the G4 treatment reached the greatest K_{sat} value at the same
292 concentration as the G1 treatment for each soil, while the G2 treatment reached the greatest
293 K_{sat} at the same concentration as the G3 treatment at equivalent condition. It is important to
294 note the fact both Mg and Ca resulted in an increased K_{sat} , which supports a reversible process.
295 Such reversibility cannot occur for a dispersed soil. However, in most cases, soils leached with
296 Ca solutions had greater K_{sat} than those leached with Mg solutions for each of the four soils,
297 particularly at low EC, which demonstrated the effect of Mg on hydraulic conductivity.

298 3.3 The interaction between cations and dissolved organic carbon

299 The free movement of DOC is mainly controlled by its adsorption-desorption to soil clay
300 surfaces (Ussiri and Johnson, 2004). In Figure 3, four different zones were determined for

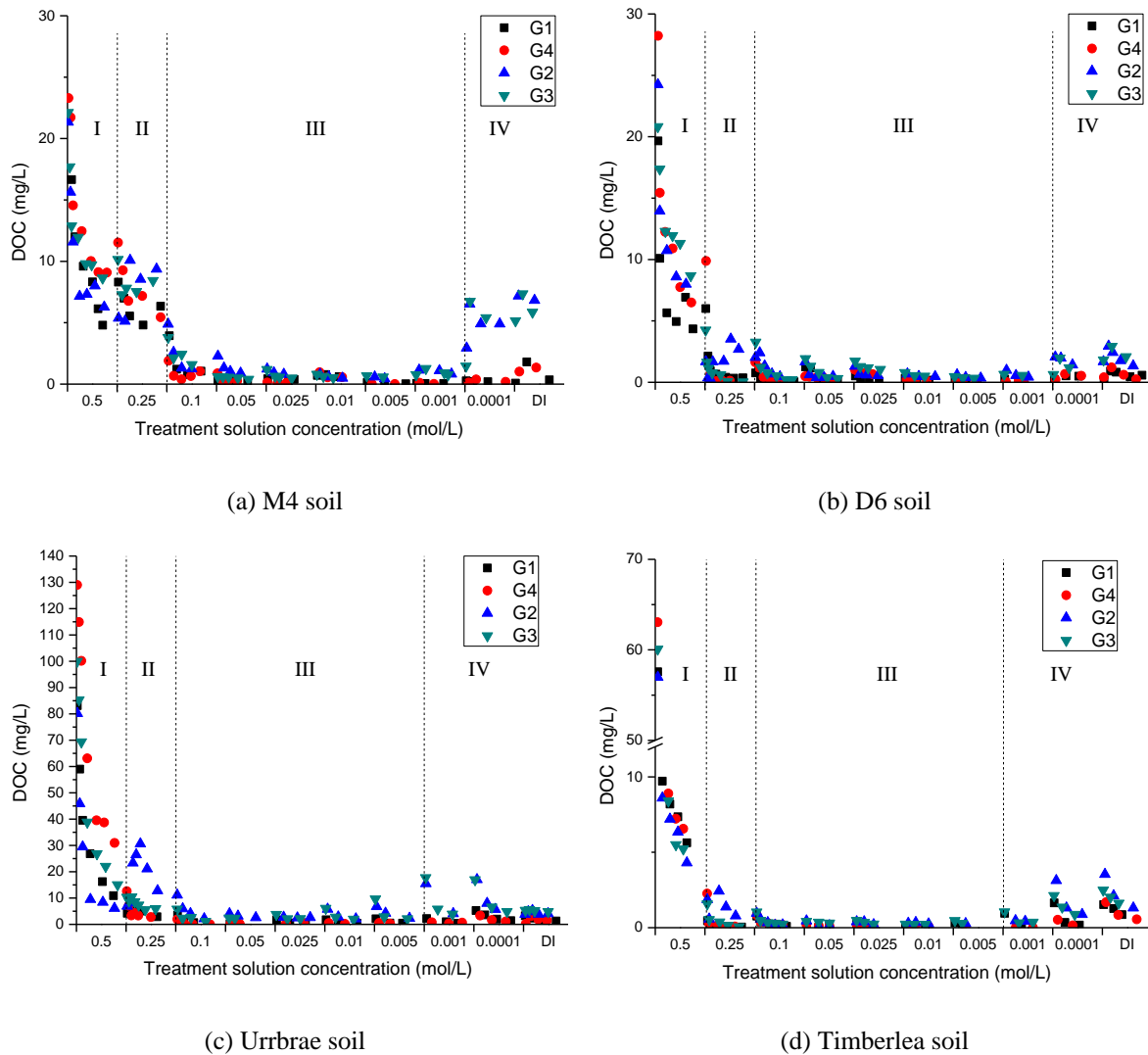
301 hydraulic network changes in relation to the changes of DOC from the leachate. In Zone I,
302 initial high DOC values were observed in each soil and then DOC dropped quickly as 1.0 L of
303 baseline solution percolated over, irrespective of the cation dominating the solution. The
304 initially high DOC values in the leachate were due to flushing of soluble organic matter
305 dissolved during wetting, which can be attributed to the response of the faster-cycling carbon
306 pools that contribute most of the decomposition flux (Wong et al., 2010). There was no
307 significant difference ($P < 0.05$) in DOC values between Mg and Ca treatment in Zone I for each
308 soil. However, it was evident that during baseline solution leaching, loss of DOC from Mg-
309 treated soils was greater than for Ca-treated soils under equivalent conditions.

310 In Zone II, a clear increase ($>50\%$) of DOC was observed in each soil with the G2 treatment
311 (Ca in Zone I, changing to Mg in Zone II); *i.e.* 0.25 M MgCl_2 solution leached through Ca
312 treated soil samples. The DOC values did not increase immediately upon leaching with MgCl_2
313 solution, but increased beyond the Ca system DOC leaching levels after approximately 150 ml
314 (≈ 4 pore volumes). This indicated that the increase in DOC was due to replacement of
315 exchangeable Ca with Mg. In contrast, for the G4 treatment (Mg in Zone I, changing to Ca in
316 Zone II), where Mg treated soil was leached with 0.25 M CaCl_2 , the DOC values decreased
317 significantly. This demonstrates that Mg has a distinct effect on the adsorption-desorption
318 behaviour of organic carbon to clay surfaces.

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322 Fig.3 The DOC values collected from the leachates for four groups (G1, G2, G3 and G4) of four soils (a) M4 soil
 323 (b) D6 soil (c) Urrbrae soil (d) Timberlea soils; (zones I, II, III and IV are explained in the text).

324

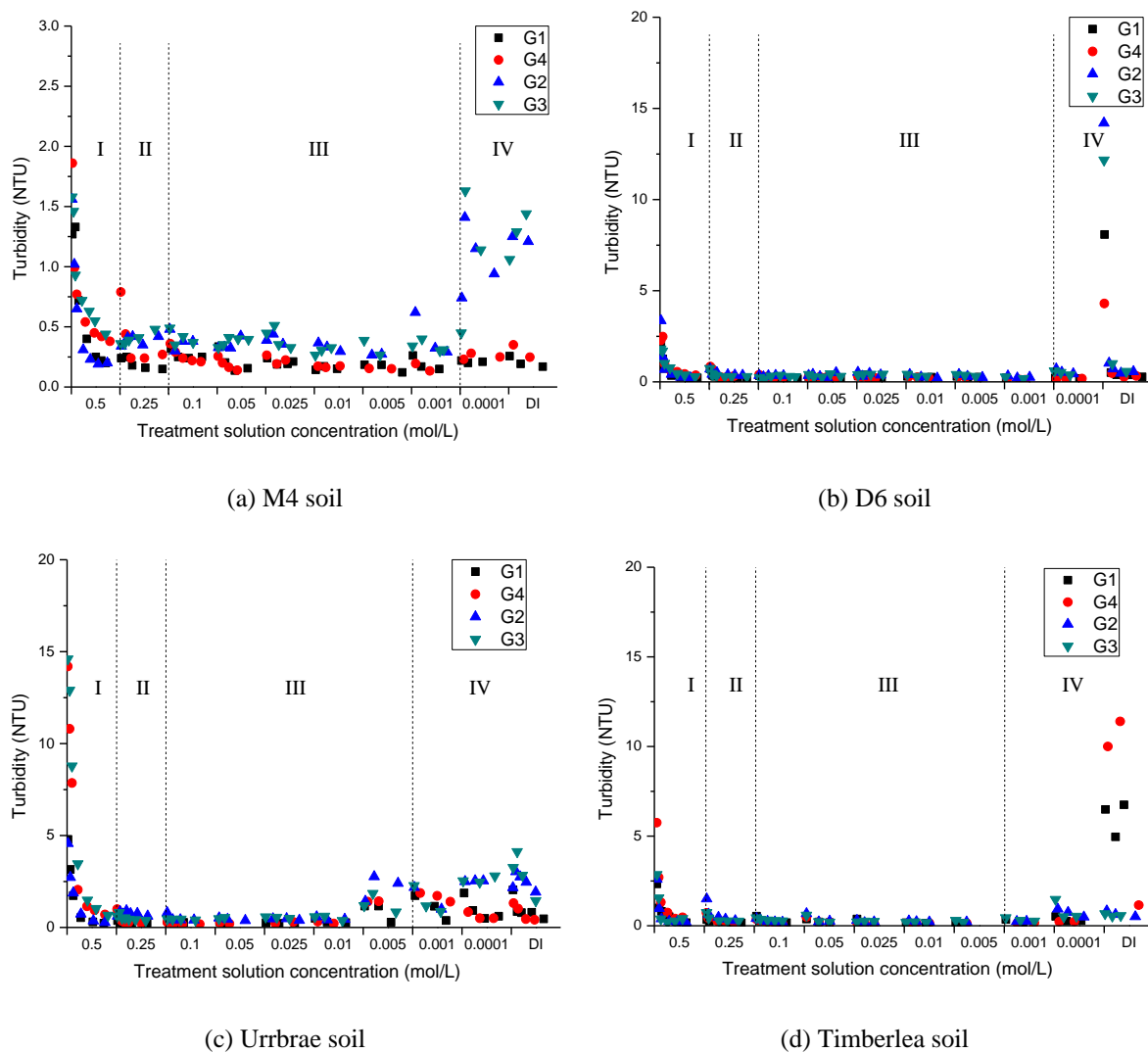
325 In Zone III, where the concentrations of the leaching solution were below 0.25 M, the DOC
 326 values of both Ca treated and Mg treated remained very low. This is mainly because the easily
 327 soluble organic matter has been flushed during the previous leaching. Within this range of EC,
 328 there was no evident effect on desorption of DOC from clay minerals. Once the leaching
 329 concentration was below the CFC (*i.e.*, M4 0.0001 M, D6 0.0001 M, Urrbrae 0.001 M, and
 330 Timberlea 0.001 M), where the K_{sat} value of each soil began to reduce significantly, it is noted
 331 the DOC values of the Mg treated soils started to increase correspondingly (Zone IV). This can
 332 be related to the primary particles in soils leached with low EC solutions separating, allowing

333 clay swelling/disaggregation to occur, which can increase the accessibility and availability of
334 covered SOM and accelerate carbon loss (Oades, 1984). It was noted that DOC of Ca treated
335 soils was 60% less than Mg treated soils in Zone IV. This suggests Mg induced disaggregation
336 results in removal of carbon from the system through desorption processes.

337 3.4 Evaluation of leachate turbidity

338 As shown in Fig. 4, the turbidity of the leachate was maintained at extremely low levels (<20
339 NTU; 0.016 % dispersed clay from soil) throughout the entire leaching process (Zhu et al.,
340 2016). The same divisional Zones for each soil used in DOC figure were applied here. In Zone
341 I, turbidity values decreased quickly as the baseline solution percolated through. The Urrbrae
342 soil had the highest initial turbidity compared to the other three soils; *i.e* the Mg treated Urrbrae
343 soils had initial turbidity of 15 NTU while Ca treated Urrbrae soils had 5 NTU. In Zone II,
344 there was no significant difference ($P<0.05$) between G2 (Ca-Mg) and G1 (Ca-Ca) treatments,
345 or G4 (Mg-Ca) and G3 (Mg-Mg) treatments, which indicates that the replacement between Ca
346 and Mg was unlikely to have caused dispersion at high EC. In Zone III, the turbidity value
347 remained extremely low, regardless of being Mg-treated or Ca-treated. In Zone IV, a slight
348 increase of turbidity value in each soil was observed when low EC solution was leached.
349 According to Quirk (2001), the solution concentration where dispersed clay particles appeared
350 in the percolate is defined as turbidity concentration, and is usually in the order of 1/8 of the
351 flocculation concentration. When the concentration of percolating solution was several times
352 below the CFC (*i.e.* M4 and D6 soil <0.0001 M, Urrbrae and Timberlea <0.001 M), soil clay
353 dispersion is expected to occur. However, the turbidity values of the leachate subsampled at
354 concentration much less than the CFC (Zone IV), even leached with 1.0 L of DI water, were
355 extremely low (<15 NTU), irrespective of the treatment or soil. This can be partially explained
356 by the fact that the flocculation of a clay suspension is not directly equivalent to a soil
357 aggregation system (Quirk, 2001), whereby in comparison to extracted soil clay, soil

358 aggregates are built up of structural units of various sizes held together by organic matter and
 359 other binding agents (Tisdall and Oades, 1982). However, even for an aggregated system we
 360 expect to find dispersed clay in the leachate when the aggregation-dispersion boundary is
 361 breached (Dang et al., 2018b; Quirk and Schofield, 1955), which could reasonably be expected
 362 at the low EC of the experimental DI treatment. The extremely low turbidity of leachate
 363 suggested that Mg had negligible effect on causing soil clay dispersion, which was very similar
 364 to the effect of Ca.



365 Fig.4 The change of turbidity values of the leachates collected from four groups (G1, G2, G3 and G4) of four soils
 366 (a) M4 soil (b) D6 soil (c) Urrbrae soil (d) Timberlea soils; (zones I, II, III and IV are explained in the text).

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369 3.5 Soil dispersion assessment after leaching with DI water

370 To further assess the potential for Mg to cause soil clay dispersion, the aggregate stability test
371 was conducted (Table 4). Instant slaking was observed in all soil aggregates upon immersion
372 with DI water, while no dispersion was observed, irrespective of Ca or Mg treatment for any
373 of the soils.

374 The turbidity and zeta potential measured from mechanical dispersion test are shown in Table
375 4. For the Timberlea soil (kaolinite dominant), minimal difference in turbidity values between
376 Mg and Ca treatments was observed in mechanical dispersion tests. This observation was in
377 line with limited difference in K_{sat} between Mg and Ca treatment, due to the low CEC of
378 kaolinite and low clay content of Timberlea soil. For the other three soils, Mg-treated soil
379 demonstrated greater turbidity than the Ca treatment. In particular, mechanical dispersion of
380 Mg treated M4 soils had 2800 NTU, while Ca treated M4 soil had 2300 NTU at similar
381 condition. Zhang and Norton (2002) found that Mg had a more deleterious effect on high
382 montmorillonite containing soil. However, the zeta potential of each soil clay suspension
383 showed that the two montmorillonite soils (M4 and D6) had lower zeta potential than illite and
384 kaolinite soils. Therefore, it was reasonable to assume that the relatively high turbidity shown
385 in M4 and D6 soils were partially related to their high clay content. It was further noted that
386 the zeta potential of Mg and Ca treated soil were >-30 mV, meaning soil clays were expected
387 to conform to flocculation conditions (Molina, 2014). In addition, the difference of zeta
388 potential between Mg and Ca treatments was minimal (<1.2 mV), *i.e.* no significant difference
389 ($P<0.05$), suggesting Mg behaved similarly to Ca, causing minimal dispersion. In summary,
390 the results from the aggregate stability test and zeta potential support the fact dispersion has
391 not occurred in Mg treated soils without external mechanical force, rather increased
392 disaggregation presumably due to enhanced inter-crystalline swelling and/or the diffuse double
393 layer development inter-crystalline swelling, meaning the clay domains remain non-dispersive.

394 Table 4 The turbidity and zeta potential measured from mechanical dispersion test

Soil	Treated group	Aggregate stability test score	Turbidity NTU	Zeta potential mV
M4	G1	0	2347	-20.8
	G2	0	2896	-21.3
	G3	0	2723	-21.6
	G4	0	2260	-20.9
D6	G1	0	798	-22.4
	G2	0	1120	-23.6
	G3	0	953	-23.5
	G4	0	689	-23.0
Urrbrae	G1	0	1421	-26.5
	G2	0	1685	-27.7
	G3	0	1669	-27.1
	G4	0	1321	-26.8
Timberlea	G1	0	587	-29.5
	G2	0	663	-30.1
	G3	0	634	-29.9
	G4	0	595	-29.1

395

396 3.6 The disaggregation effect of Mg on soil structural stability

397 The results of this work allow us to accept the three hypotheses that: 1) Mg reduces K_{sat} , as
 398 compared to Ca, induced by disaggregation, rather than dispersion; 2) the difference between
 399 the K_{sat} of Mg and Ca varies dependent on clay mineralogy and soil texture; and, 3) DOC
 400 concentrations from Mg leached samples are greater than those from Ca leached samples, with
 401 this related to reduction in K_{sat} , but not dispersion of soil primary particles.

402 This study demonstrated that over a wide range of concentrations, Mg had an effect on soil
 403 structural stability by reducing K_{sat} as compared to Ca. This was evidenced by:

404 1) In most cases, greater K_{sat} values for soil leached with CaCl_2 solutions (G1; Ca-Ca) than for
 405 those leached with MgCl_2 solutions at equivalent conditions (G3; Mg-Mg), irrespective of soil
 406 clay mineralogy;

407 2) An abrupt reduction in K_{sat} was observed when Ca treated soils were leached with MgCl_2
 408 solutions (G2; Ca-Mg), as compared to G1 (Ca-Ca) for all of the four soils; and,

409 3) An obvious increase in K_{sat} found for Mg treated soils subsequently leached with Ca (G4;
 410 Mg-Ca), as compared to a maintained Mg equivalent condition (G3; Mg-Mg).

411 According to Rengasamy et al. (1986), when the percolating solution exceeded the flocculation
412 values, hydraulic conductivity was not affected by either SAR or Ca/Mg ratio. However, the
413 results in this study showed that Mg had a reducing effect on K_{sat} , and thereby soil structural
414 stability, even when the concentration of the percolating solution was well above the CFC. This
415 can be related to Mg induced disaggregation, due to the occurrence of lattice intra-crystalline
416 swelling in montmorillonite clays and/or diffuse double layer development, inter-crystalline
417 swelling when electrolyte concentration decreases (Bennett et al., 2019). Furthermore, the
418 disaggregation effect of Mg is also related to its degree of covalent bonding between clay and
419 Mg cations. Despite both Ca and Mg having similar charge, the two cations have different ionic
420 potential (Huheey et al., 1993) and Misono's softness parameter (Sposito, 2008). Consequently,
421 as compared to Ca, the Mg treated soils had a lower degree of covalency in clay-cation bonding
422 (Marchuk and Rengasamy, 2011), which caused smaller mean particle size (aggregation size),
423 lower porosity and lower hydraulic conductivity (Marchuk et al., 2013b), on this basis. The
424 greater turbidity values of Mg treated soils in the mechanical dispersion test, than for those of
425 Ca treated soil, occurred with minimal difference in zeta potential, which can also be partially
426 attributed to the small mean particle size of Mg soil clays. Thus, Mg clay suspension would
427 have required more time to achieve full flocculation (Stokes law). This is a very important
428 result, as it changes the way that we expect Mg to interact within the soil system; *i.e.* Mg is not
429 directly responsible for dispersion, but could be expected to exacerbate the effects of
430 monovalent ions on soil dispersion.

431 The results also demonstrated that Mg had a disaggregation effect on soil structural stability
432 through increasing desorption of DOC from clay surfaces, as compared to Ca, whereby
433 replacing Ca with Mg solution, even at high EC, resulted in more DOC being removed from
434 the soil system. Furthermore, leachate from the Mg treatments always contained more DOC
435 than the Ca treated soils. These suggested that the loss of organic carbon is related to Mg

436 induced disaggregation, which can increase the accessibility and availability of covered SOM
437 and accelerate carbon loss. Moreover, Ca improves soil structure stability through cationic
438 bridging with clay particles and soil organic matter, as it can effectively link clays with
439 permanent negative charges to anionic functional groups on organic matter (Bronick and Lal,
440 2005; Churchman et al., 1993; Oades, 1984; Singh et al., 2016). From this study, Mg has shown
441 a greater capacity than Ca for desorption of DOC, which suggests Mg is less effective than Ca
442 in forming strong bonds between negatively charged clay minerals and DOC through cation
443 bridging. The Mg bond is less covalent and more ionic in nature than Ca (Marchuk and
444 Rengasamy, 2011), meaning water molecules can break the covalent Mg bonds easier than that
445 of Ca.

446 The results also indicated that the degree of disaggregation effect of Mg on K_{sat} and DOC varied
447 depending on clay mineralogy and texture. The difference in K_{sat} between Mg and Ca baseline
448 treatments was as large as 35% in the two Vertosols, while it was 25% for the Urrbrae soil
449 (illitic) and 4% for Timberlea soil (kaolinitic). Similar results were reported by Zhang and
450 Norton (2002) that the K_{sat} of a smectitic soil leached with 0.01 M Mg solution was 35% less
451 than that leached with 0.01 M Ca solution, while on the other hand, 13% difference was
452 reported for an illite dominant soil. The low initial K_{sat} values and high turbidity values in
453 mechanical dispersion tests of Mg montmorillonite soils, as compared to Ca montmorillonite
454 soils, were partly due to the difference in covalency degree between Mg and Ca ions, and partly
455 due to the high CEC of montmorillonite clay and high clay content. To the contrary, there was
456 less difference between Mg and Ca on the K_{sat} , DOC and mechanical dispersion of the
457 Timberlea soil, as compared to the other three soils. This is mostly likely because of the low
458 clay percentage in the Timberlea soil, as well as kaolin being the dominant clay. With lower
459 surface area and CEC, the exchangeable cations had less impact on kaolin than illite and
460 montmorillonite clays. In this work, three contrasting mineralogies were selected in order to

461 explore the effect of clay mineralogy on the obtained results. However, future work should
462 seek to consider this aspect of the work in order to incorporate such relationships into predictive
463 models.

464 **Conclusions**

465 This study demonstrated that Mg had a disaggregation effect, rather than dispersion effect, on
466 soil structural stability when compared to the equivalent condition of Ca by reducing K_{sat} and
467 increasing desorption of DOC from the clay surface. An initial increase of K_{sat} was observed
468 for each soil when leached with a successive of $MgCl_2$ solutions (electrolyte concentration
469 above 0.005 M), with the extent and point of occurrence of the highest K_{sat} depending on the
470 dominant soil clay mineralogy and the type of cation in the leaching solution. When electrolyte
471 concentrations of leaching solution were below CFC, rapid reduction in K_{sat} was detected in all
472 four soils, but to a different extent. The low turbidity of leachates in all soils suggested that Mg
473 clay domains are non-dispersive. However, mechanical dispersion testing demonstrated that
474 the presence of Mg on the exchange sites results in higher dispersion than Ca caused under
475 additional force or intensive cultivation operations.

476 The results of this study demonstrated that Mg has more deleterious effect on soil structural
477 stability than Ca, due to disaggregation processes, and determines with confidence that Mg is
478 not the direct cause of dispersion. The degree of the disaggregation effect of Mg on saturated
479 hydraulic conductivity and the release of the dissolved organic carbon largely depends on the
480 electrolyte concentration of the soil solution and dominant clay mineralogy.

481

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