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12 **ABSTRACT**

13 Altered pyroclastic (tephra) deposits are highly susceptible to landsliding, leading
14 to fatalities and property damage every year. Halloysite, a low-activity clay mineral, is
15 often associated with landslide-prone layers within altered tephra successions, especially
16 in deposits with high sensitivity, which describes the post-failure strength loss. However,
17 the precise role of halloysite in the development of sensitivity, and thus in sudden and
18 unpredictable landsliding, is unknown. Here we show that an abundance of mushroom-
19 cap-shaped (MCS) spheroidal halloysite governs the development of sensitivity, and
20 hence proneness to landsliding, in altered rhyolitic tephras, North Island, New Zealand.
21 We found that a highly sensitive layer, which was involved in a flow slide, has a
22 remarkably high content of aggregated MCS spheroids with substantial openings on one

23 side. We suggest that short-range electrostatic and van der Waals' interactions enabled
24 the MCS spheroids to form interconnected aggregates by attraction between the edges of
25 numerous paired silanol and aluminol sheets that are exposed in the openings and the
26 convex silanol faces on the exterior surfaces of adjacent MCS spheroids. If these weak
27 attractions are overcome during slope failure, multiple, weakly-attracted MCS spheroids
28 can be separated from one another and the prevailing repulsion between exterior MCS
29 surfaces results in a low remolded shear strength, a high sensitivity, and a high propensity
30 for flow sliding. The evidence indicates that the attraction-detachment model explains the
31 high sensitivity and contributes to an improved understanding of the mechanisms of flow
32 sliding in sensitive, altered tephra rich in spheroidal halloysite.

33 **INTRODUCTION**

34 Most East Asian and western Pacific countries are located in tectonically active,
35 high-rainfall areas where landslides are a major natural hazard. These landslides are
36 typically triggered by rainstorms or earthquakes, and are responsible for fatalities and
37 enormous property damage every year. Many destructive landslides have occurred in
38 pyroclastic deposits in Japan, Indonesia, Hong Kong, and New Zealand (Chau et al.,
39 2004; Chigira, 2014; Moon, 2016), such deposits often containing layers rich in clay
40 minerals formed mainly by chemical weathering either during pedogenesis or diagenesis.
41 In regions with predominantly rhyolitic volcanism, halloysite is a common clay mineral
42 (Churchman and Lowe, 2012) and is therefore potentially a key geological factor
43 increasing the risk of landslides (Kirk et al., 1997; Chigira, 2014). Halloysite is a 1:1
44 Si:Al layered aluminosilicate member of the kaolin subgroup that exhibits various

45 structural morphologies including tubes, spheroids, polyhedrons, plates and books
46 (Joussein et al., 2005; Cunningham et al., 2016).

47 Spheroidal halloysite, in particular, has been recognized in landslide-prone layers
48 of pyroclastic material in Japan (Tanaka, 1992) and New Zealand (Smalley et al., 1980).
49 Smalley et al. (1980) linked a high content of spheroidal halloysite to high sensitivity.
50 Sensitivity refers to the post-failure strength loss in the failure zone during landsliding,
51 and is quantified in the laboratory as the ratio of the undisturbed to remolded undrained
52 shear strength at the same water content (Terzaghi, 1944). High sensitivities were first
53 described for post-glacial, brackish and marine clayey sediments in the Northern
54 Hemisphere (Skempton and Northey, 1952) that are subject to landslides with
55 dimensions and long runout distances difficult to predict. In this study, we investigate
56 processes that have led to high sensitivity in halloysite-rich pyroclastic materials in order
57 to improve landslide-hazard evaluation.

58 **GEOLOGICAL SETTING**

59 Much of the central part of New Zealand's North Island is covered by thick
60 rhyolitic tephra (Lowe, 2011) derived from eruptions in the Taupo Volcanic Zone
61 (Briggs et al., 2005), which are often altered into halloysite-rich successions. We focus
62 here on a coastal flow slide at Omokoroa, Bay of Plenty (Fig. 1A), where ~10,000 m³ of
63 material were transported downslope over long distance into a lagoon in 1979 (Moon et
64 al., 2015), as well as two minor reactivations in 2011 and 2012. The 1979 event was
65 likely initiated in a white, highly sensitive layer with high spheroidal halloysite
66 concentration (Smalley et al., 1980), lacking any detectable allophane (Cunningham et
67 al., 2016).

68 We have analyzed a 40 m-long sediment core, *Omok-1*, which was bored via
69 rotary flush drilling in unfailed material near the headwall (Fig. 1B). The lithology of
70 *Omok-1* was determined by correlation with units of a previously-studied adjacent
71 headwall face (Moon et al., 2015) comprising a succession mainly of Quaternary rhyolitic
72 tephra: overlying lignite at the base of the core, the Pahoia Tephra sequence includes the
73 Te Puna ignimbrite (~0.93 Ma), and a series of altered tephra, which are informally
74 divided into lower and upper Pahoia Tephra units based on two distinct paleosols (P1 and
75 P3). All these deposits and paleosols are overlain by successions of younger altered
76 tephra called Hamilton Ash beds (~0.35 to ~0.05 Ma) and late Quaternary tephra
77 (<~0.05 Ma) (Fig. 1C and 2A). The lower Pahoia Tephra include the 0.3-m-thick, white,
78 highly sensitive clay-rich layer which failed in 1979 (Fig. 1C), having high porosity and
79 high natural water content (Smalley et al., 1980).

80 **METHODS**

81 We performed laboratory vane shear tests on samples from the Pahoia Tephra
82 sequence and Hamilton Ash beds to measure the sensitivity S :

$$83 \quad S = s_u / s_r \quad (1)$$

84 where the undisturbed strength (s_u) was measured on the intact surface of the split
85 core, and the remolded strength (s_r) was measured on core samples with the same water
86 content, which have been kneaded by hand for 10 min (Jacquet, 1990). Halloysite
87 concentration in bulk samples was measured by X-ray diffraction (XRD) using a Philips
88 PW analytical diffractometer and quantification was performed using QUAX (Vogt et al.,
89 2002). Scanning electron microscopy (SEM) was undertaken with a Zeiss Supra40
90 microscope on 24 shock-frozen, freeze-dried, and gold-coated bulk core samples (Reed,

91 2005). The relative abundances of halloysite particles having distinct morphologies were
92 quantified using a point-counting approach (Frolov and Maling, 1969). Six representative
93 SEM-images of planar soil surfaces were chosen for each sample and at least 600
94 particles were counted based on rectangular grids. In the white, highly sensitive layer, the
95 change of halloysite particle arrangement upon remolding was quantified by comparing
96 20 SEM images of undisturbed and remolded material, providing > 1000 counts,
97 respectively. The spheroid diameters were measured from six representative particles per
98 SEM image.

99 **HIGHLY SENSITIVE SLIDE-PRONE LAYER DOMINATED BY SPHEROIDAL**
100 **HALLOYSITE**

101 The sensitivity is low in the upper Pahoia Tephra, especially in the paleosols P2
102 and P3 (Fig. 2A, B). However, the sensitivity tends to increase with depth, reaching
103 values of 15–20 in the lower Pahoia Tephra. The highest sensitivity (Rosenqvist, 1953)
104 of $S = 55$, and the lowest remolded shear strength within the profile of $s_r = 1.4$ kPa, were
105 measured in the white, highly sensitive layer at 23 m depth.

106 The upper Pahoia Tephra have a halloysite content of 10–20 wt.%, and are
107 comprised almost entirely of tubular halloysite (Fig. 2C, D). The lower Pahoia Tephra
108 have 40–50 wt.% halloysite comprising mostly spheroidal particles. In the highly
109 sensitive layer, 76% of the halloysite is spheroidal, and the spheroid sizes are greater than
110 those in the surrounding layers (Fig. 2D). The 3D line plot reveals a clear correlation
111 between high sensitivities and high halloysite bulk concentration, and a high content of
112 spheroids with large diameters (Fig. 2F). The high sensitivity is associated with low
113 remolded shear strength rather than with high undisturbed shear strength (Fig. 2G).

114 We found that deposits with high tubular halloysite content hampered sensitivity
115 development, whereas halloysite spheroids facilitate sensitivity and dominate the highly
116 sensitive layer at 23 m depth within the lower Pahoia Tephra. The highly sensitive layer
117 has low remolded shear strength consequent after failure, which, together with its high
118 water content (Smalley et al., 1980), partly contributed to the long runout distance of the
119 flow slide at Omokoroa.

120 **NEW HALLOYSITE MORPHOLOGY**

121 We present here first observations of a previously unreported halloysite particle
122 morphology that is visible in the SEM images of the remolded halloysite fabrics of the
123 highly sensitive layer. In the undisturbed state, the spheroidal halloysites are distinctly
124 aggregated into networks of well-connected particles (Fig. 3E, F). After remolding,
125 however, most of the aggregates have broken apart into small, loose clusters or individual
126 halloysite particles that are typically ~250–400 nm in diameter (Fig. 3G, H). Individual
127 spheroids have distinctive ‘deformities’ in the form of openings ~80–160 nm in diameter
128 on one side. These openings were previously hidden by contact with other spheroids. The
129 deformities give the particles an ovate “mushroom-cap” appearance. Point-counting
130 individual mushroom-caps in both undisturbed (aggregated) and remolded
131 (disaggregated) samples showed that the observable mushroom-caps were much more
132 abundant in the remolded samples, increasing from $4.4 \pm 3.2\%$ to $44.9 \pm 11.6\%$.

133 **ATTRACTION-DETACHMENT MODEL FOR FLOW SLIDING IN ALTERED** 134 **TEPHRAS**

135 The open-sided, mushroom-cap-shaped halloysite morphology has not been
136 reported previously. Because this particular morphology overwhelmingly occurs in the

137 highly sensitive slide-prone layer, we hypothesize that this unique particle shape controls
138 the mechanical behavior of halloysite clays.

139 Halloysite is composed of an Al-octahedral (aluminol) sheet with a net positive
140 charge and a Si-tetrahedral (silanol) sheet with a net negative charge at pH values
141 between ~2 and ~8 (Fig. 3I) (Churchman et al., 2016). The two sheets have slightly
142 different dimensions, with the silanol sheet being larger. This misfit in the sheet sizes
143 causes the halloysite layer to be curved (Churchman and Lowe, 2012), with the larger
144 negatively-charged silanol sheet on the outside of the curvature and the positively-
145 charged smaller aluminol sheet on the inside. The halloysite spheroids observed in our
146 study are most likely composed of concentrically stacked 1:1 layers, i.e., with an onion-
147 like structure, as shown in numerous studies including those on spheroidal halloysite
148 derived from altered tephra in New Zealand, Japan, and Argentina (Wada et al., 1977;
149 Kirkman, 1981; Cravero et al., 2012; Berthonneau et al., 2015). For a perfect halloysite
150 spheroid, the outermost silanol surface carries a net negative charge and hence the
151 electrostatic interactions between individual spheroids would be repulsive (Fig. 3I). Our
152 study shows, however, a halloysite structure where both silanol and aluminol layers are
153 exposed at spheroid openings and therefore charges within the openings would
154 correspondingly be weakly positive or neutral overall (Fig. 3J), as indicated from charge
155 density-functional tight-binding modeling applied to halloysite nanotubes (Guimarães et
156 al., 2010). If sufficient numbers of positively charged openings are exposed, the
157 electrostatic interactions between them and the negative exterior silanol surfaces would
158 allow the mushroom-cap-shaped spheroids to form stacked aggregates (Fig. 3K). If the
159 paired silanol and aluminol sheets exposed in the openings are neutral overall, then a net

160 increase in particle attraction will still occur because electrostatic repulsion is reduced
161 and the larger contact areas lead to higher van der Waals' forces (Israelachvili, 2011).

162 During diagenesis via hydrolysis of volcanic glass (Cunningham et al., 2016), the
163 halloysite spheroids may form consecutively on top of one another in pore spaces,
164 generating the distinct openings during synthesis. The attractive forces between the
165 openings and the convex exterior surfaces are demonstrably strong enough to allow for
166 the formation of aggregates, but also permit easy disaggregation by mechanical
167 detachment during shear (Fig. 3L). New random contacts between convex silanol
168 surfaces probably lead to a decrease in average attraction between particles. We posit that
169 the detachment of attractive spheroidal particle contacts, in the presence of abundant
170 water having negligible interaction with soil-water ions because of the inactive nature of
171 halloysite (Smalley et al., 1980), leads to the very low post-failure shear strength,
172 facilitating a flow slide with long runout distance. The interparticle, attraction-
173 detachment model appears to successfully explain (at nanoscale dimensions) the post-
174 failure behavior of the highly sensitive tephra layer at Omokoroa that is dominated by the
175 imperfect halloysite spheroids. The question therefore arises if similar altered tephras
176 elsewhere have high contents of spheroidal halloysite with potentially hidden mushroom-
177 cap forms, and if such forms helped mobilize other landslides in the past.

178 **CONCLUSIONS**

179 We investigated a sequence of altered, rhyolitic Quaternary tephras in New
180 Zealand, and the reasons why a landslide-prone layer dominated by spheroidal halloysite
181 was highly sensitive. We explain this high sensitivity with an electrostatic attraction-
182 detachment model. Weakly positive or neutral charges on silanol and aluminol sheet

183 edges exposed in the concave openings of spheroidal halloysite particles were attracted to
184 the negatively-charged convex silanol surfaces of adjacent spheroids. Such short-range
185 attractions between spheroid openings, and the exterior surfaces of adjacent spheroids,
186 stabilize an aggregated halloysite framework. If the aggregates are detached by
187 remolding, the loose arrangement of the spheroids exhibits low remolded shear strength.
188 We suggest that the attraction-detachment model, based on the identification of
189 mushroom-cap halloysite morphologies, provides a potential key for the identification of
190 sensitive altered tephra that are predisposed to sudden failure that triggers landsliding.

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273

274 FIGURE CAPTIONS

275

276 Figure 1. A: Map of Tauranga Harbour, New Zealand, with the Taupo Volcanic Zone
277 (TVZ) as main source for Quaternary tephra at the study site. B: 3D-view of the
278 Bramley Drive flow slide at Omokoroa; red line marks the position of the profile in C. C:
279 Profile through the flow slide with simplified stratigraphy and associated paleosols (P1–
280 4) of core *Omok-1* and ages (in Ma) after Moon et al. (2015).

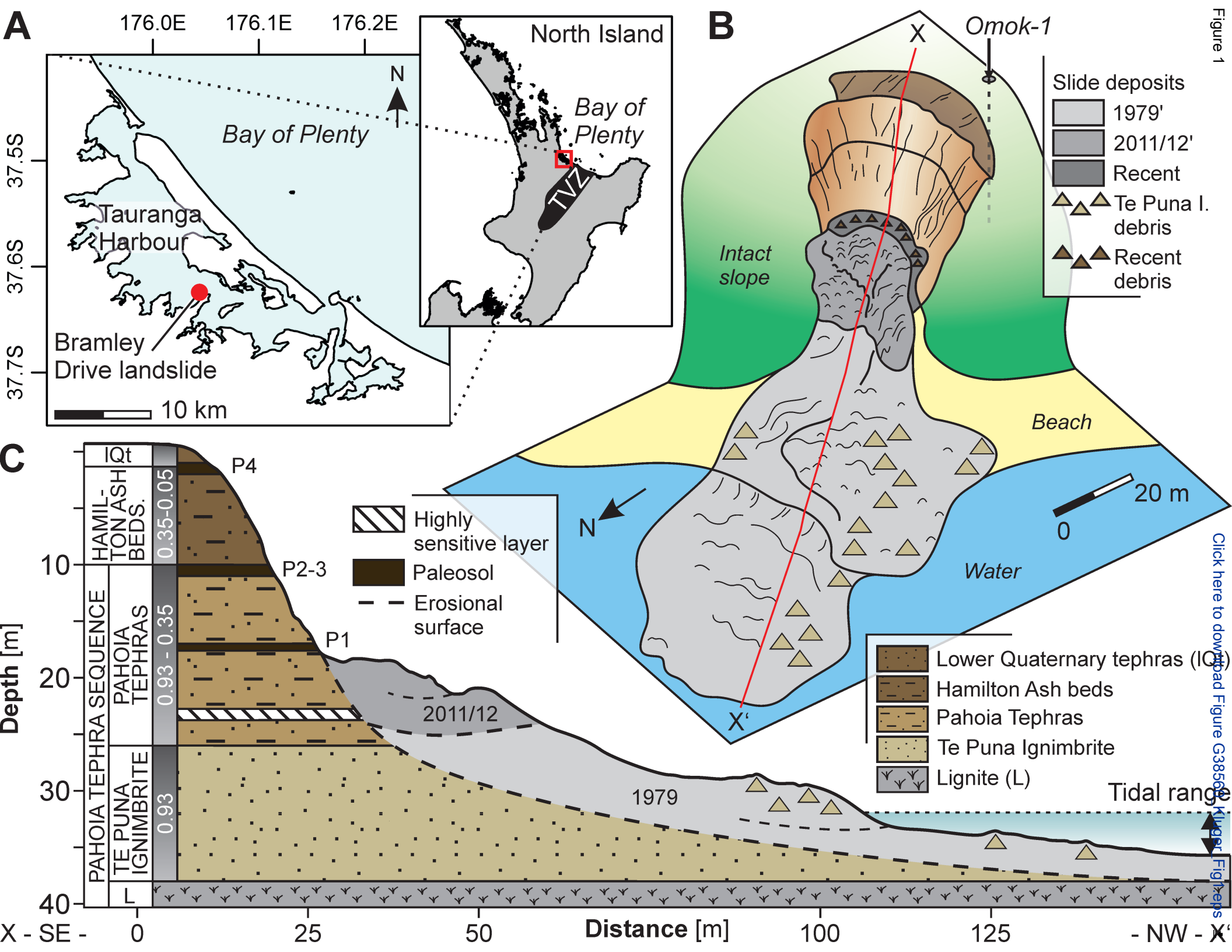
281

282 Figure 2. A: Stratigraphy of core *Omok-1* after Moon et al. (2015) showing the main
283 lithological units as defined in Figure 1, three paleosols (P1–3), and the highly sensitive
284 white layer at 23-m depth (hatched area). B: Undisturbed (s_u) and remolded (s_r) shear
285 strength, and sensitivity (S). C: Halloysite bulk concentration. D: Cumulative volume %
286 (c. %) of halloysite morphologies with bars indicating average standard deviations. E:
287 Average spheroid sizes with standard deviations depicted by fill patterns. F: 3D line plot
288 illustrating the relationship between spheroid content, sensitivity, spheroid size, and
289 halloysite concentration; gray graded areas enable trends in sensitivity to be visualized.
290 G: Dependency between sensitivity and shear strength.

291

292 Figure 3. SEM-images of spheroids (A), polyhedrons (B), tubes (C), and plates (D)
293 representing the main halloysite morphologies in the Pahoia Tephra sequence. SEM-
294 images from the highly sensitive layer of undisturbed and multiply connected halloysite
295 spheroids (E, F) and remolded spheroids (G, H) showing smaller clusters or detached
296 spheroids within a much looser particle network. 1: Exposed layers in spheroid openings.
297 2: Partially separated halloysite spheroids. 3: Detached mushroom-cap-shaped halloysite

298 spheroid. I: Electrostatic field proximal to halloysite nanotubes with colored equipotential
299 surfaces (ES), modified with permission from Guimarães et al. (2010). Copyright 2010
300 American Chemical Society. J: Conceptual mushroom-cap-shaped spheroid cross-section
301 and the weak electrostatic and/or van der Waals' attractions arising between the exposed
302 silanol-aluminol sheets in spheroid openings and the negatively-charged convex exterior
303 surfaces; enlargement is adapted from Berthonneau et al. (2015). Circles with + and –
304 relate to the positive and negative electrostatic field proximal to the spheroid's exterior
305 surface. Mushroom-cap-shaped spheroids connect with one another between concave
306 openings and convex outer spheroid surfaces, forming aggregates (K) which are partly
307 detached because of remolding (L).



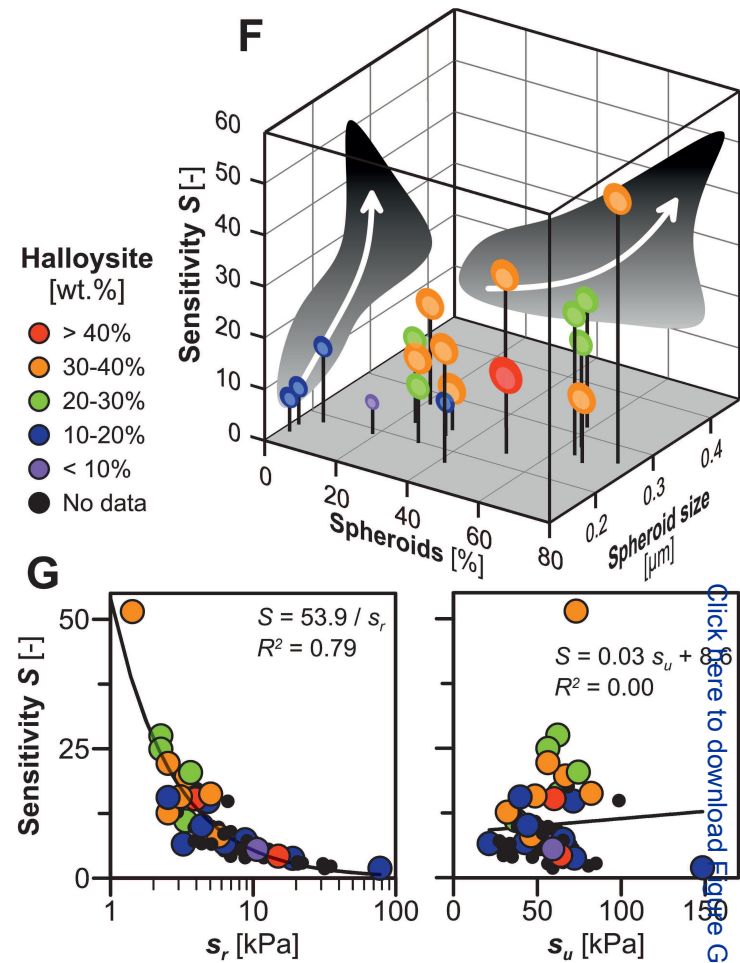
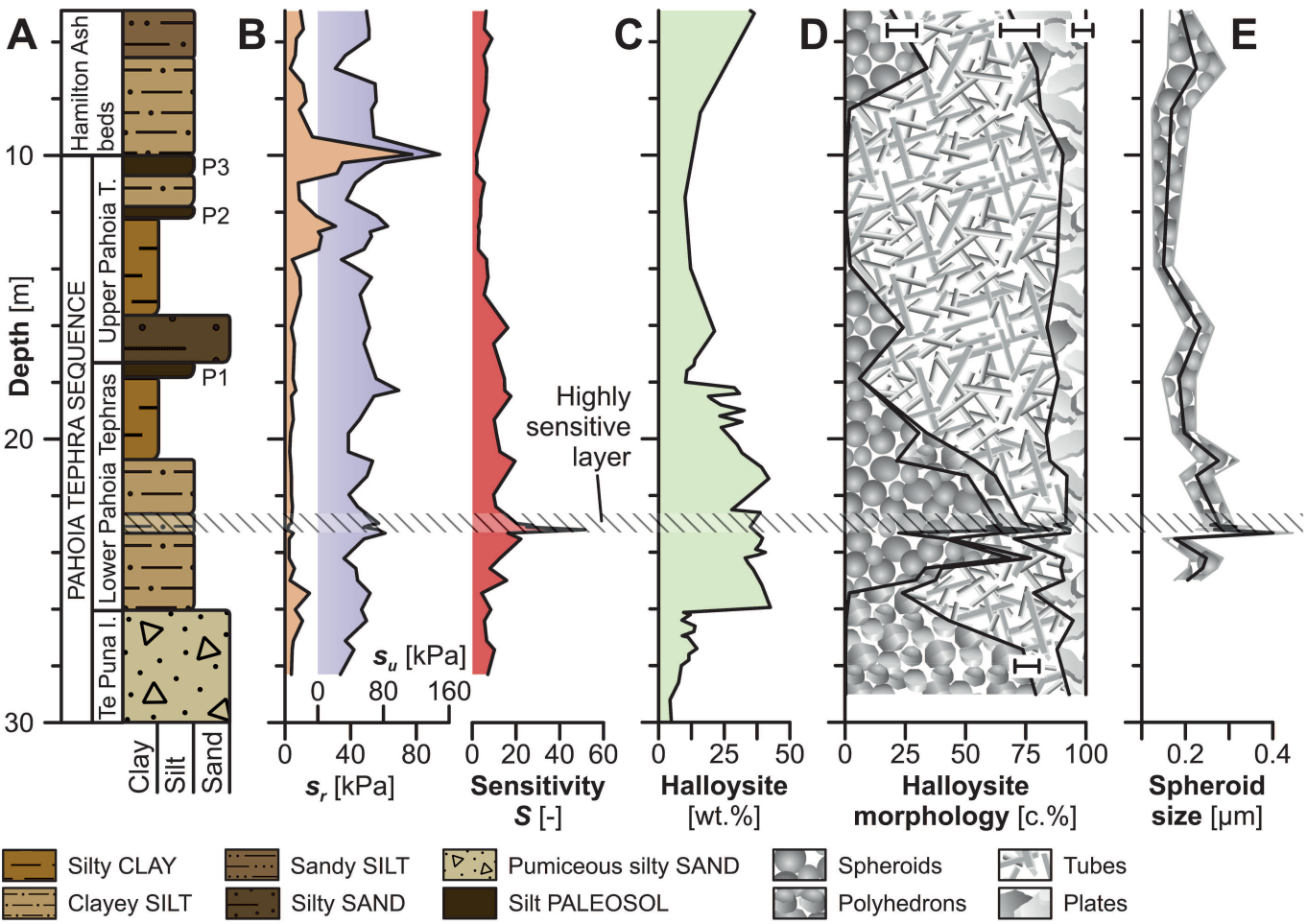


Figure 2

Figure 3

