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1 Technical note

2 **Protolith identification of bedding-parallel, smectite-bearing shear zones in**
3 **argillaceous and siliceous marine sediments: discriminating between tephra-derived**
4 **shear zones and host-rock-derived fault gouges**

5

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12 **Abstract**

13 Smectite-bearing shear zones are important in a variety of geoscientific and
14 geoengineering fields owing to the unique physicochemical properties of smectite. Such
15 shear zones occur globally in argillaceous and siliceous marine sediments, and are
16 typically a product of either host-rock deformation, or exploitation of weak, altered tephra
17 horizons within the sedimentary sequence. However, it is difficult to discriminate
18 between a tephra-derived and host-rock-derived origin based on simple fault-rock
19 observations. Here, a method is proposed that integrates two discriminators: the ratio of
20 smectite to illite, $sm/(ill + sm)$ (wt%/wt%), in the clay fractions of shear zone samples;
21 and the ratio of conservative elements, Al_2O_3/TiO_2 (wt%/wt%), in the bulk chemistry of
22 samples. Although high $sm/(ill + sm)$ ratios may indicate a tephra-derived origin,
23 deformation processes can lower the ratio, making protolith identification difficult.
24 Previous studies have suggested that different Al_2O_3/TiO_2 ratios between shear zones and
25 host rocks may also imply a tephra-derived origin. However, the Al_2O_3/TiO_2 ratios within
26 shear zones may be intrinsically comparable with those of the host rock, even if the shear
27 zones were derived from tephra layers. Despite these ambiguities, application of these
28 two indicators to the protolith identification of bedding-parallel, smectite-bearing shear
29 zones in a folded siliceous mudstone (Hokkaido, Japan) demonstrates that combining
30 $sm/(ill + sm)$ and Al_2O_3/TiO_2 ratios is a useful starting point when attempting to identify
31 the protolith of smectite-bearing shear zones in argillaceous and siliceous marine
32 mudstones.

34 **Keywords**

35 gouge, marine sediment, protolith, shear zone, smectite

37 **1. Introduction**

38 Tephra (volcanic ash) deposits are globally intercalated within argillaceous and
39 siliceous marine sediments (Hein and Scholl, 1978; Compton, 1991; Delano et al.,
40 1994). Under low-*T* diagenetic conditions, such tephra deposits undergo alteration to
41 form smectite-bearing sedimentary horizons (Ross and Hendricks, 1945; Bradshaw,
42 1975; Compton, 1991), which are often subsequently exploited as shear zones during
43 crustal deformation (Vrolijk, 1990; Kameda et al., 2015; Ishii and Furusawa, 2017).
44 Smectite-bearing shear zones within argillaceous and siliceous marine sediments are of
45 significant interest to both geoscientists and geoengineers working within the fields of
46 structural geology, hydrogeology, and geochemistry, particularly those studying plate
47 boundary faults (Vrolijk, 1990; Kameda et al., 2015, 2016), landslides (Bromhead,
48 2013; Massey et al., 2016), underground construction (Ishii et al., 2015; Ishii and
49 Furusawa, 2017), and nuclear waste disposal (Iwatsuki et al., 2009; Murakami et al.,
50 2016). This interest arises from the unique physicochemical properties of smectite, such
51 as its low coefficient of friction, cation-exchange capacity, low permeability,
52 expansivity, and variable consistency (Waltham, 2002). However, tephra-derived,
53 smectite-bearing shear zones are similar in appearance to host-rock derived fault
54 gouges, making it difficult to discriminate between them based on meso- and micro-
55 scale observations alone (Ishii and Furusawa, 2017).

56 It is important to be discriminate between tephra-derived shear zones and host-
57 rock-derived fault gouges because they typically differ with regard to their spatial extent
58 and thickness, protolith mineralogy, and the time and process of formation, as well as
59 their utility as marker beds (Ishii and Furusawa, 2017). For example, when a smectite-
60 bearing shear zone is discovered in a pilot borehole at an investigation site and is shown

61 (by mineralogy) to have a tephra-derived origin, the risk that a thick smectite-rich layer
62 is extensively distributed around the borehole should be considered, even if the shear
63 zone is very thin at the borehole. This risk arises because a tephra-derived shear zone is
64 likely to be more extensive than a host-rock-derived fault gouge, and the thickness of
65 the former may vary substantially, in part due to local stratigraphic pinchout of the
66 original tephra layer. Therefore, discriminating tephra-derived shear zones and host-
67 rock-derived fault gouges can provide knowledge of hazards (e.g., washout and
68 groundwater inflow) caused by abundant smectite during underground construction
69 (Ishii et al., 2015; Ishii and Furusawa, 2017), as well as their mitigation, by predicting
70 the distribution of the shear zone based on that of the tephra layer. For distinguishing
71 tephra-derived shear zones from host-rock-derived fault gouges, it is also important to
72 ascertain the protolith mineralogy of shear zones, which is central to establishing
73 thermal histories related to shear-zone seismicity (Kuo et al., 2009) and for studying the
74 past migration of natural trace elements analogous to radionuclides of radioactive
75 wastes (Iwatsuki et al., 2009). Knowledge of the origin of smectite-bearing shear zones
76 also has application to the study of landslides (Bromhead, 2013).

77 Tephra-derived shear zones intrinsically have a higher smectite content than that
78 of the surrounding host rock (Perry et al., 1976b; Vallier and Kidd, 1977; Tribble,
79 1990). Host-rock-derived fault gouges, on the other hand, are more likely to be
80 mineralogically similar to the host rock (Holland et al., 2006). Thus, it should be
81 possible to discriminate between the two by comparing the smectite content of the shear
82 zone with that of the host rock. However, the smectite content in a tephra-derived shear
83 zone may decrease over time as the shear zone becomes contaminated by host-rock
84 materials during deformation. The smectite content may also decrease via syn-tectonic

85 alteration, for example due to frictional heating during shear deformation (Yamaguchi et
86 al., 2011; Schleicher et al., 2015; Kuo et al., 2016), or a combination of mechanical
87 processes and chemical reactions (Vrolijk and Pluijm, 1999; Casciello et al., 2011;
88 Hirono et al., 2014). Consequently, it is also difficult to discriminate between tephra-
89 derived shear zones and host-rock-derived fault gouges by simply comparing their
90 smectite content with that of the host rock.

91 This paper proposes a method utilizing two separate indicators as a means of
92 confidently discriminating between tephra-derived shear zones and host-rock-derived
93 fault gouges in argillaceous and siliceous marine sediments. The first indicator is the
94 content of smectite normalised to that of illite (ill) plus smectite (sm) in the clay fraction
95 of shear zone samples, defined as $sm/(ill + sm)$ (wt%/wt%). The second indicator is the
96 concentration of Al_2O_3 normalised to TiO_2 in the bulk chemistry of shear zone samples,
97 defined as Al_2O_3/TiO_2 (wt%/wt%). Illite is a common clay mineral in argillaceous and
98 siliceous marine sediments, so $sm/(ill + sm)$ ratios are expected to be useful indicators
99 in identifying protoliths of smectite-bearing shear zones. Based on previous studies,
100 $sm/(ill + sm)$ ratios of tephra-derived smectite-bearing layers are typically ≥ 0.9 (Perry
101 et al., 1976a, b; Hein and Scholl, 1978; Tribble, 1990). However, with smectite content
102 possibly decreasing over time due to deformation, the proportion of conservative
103 elements (Al_2O_3/TiO_2) is also proposed. In argillaceous and siliceous marine sediments,
104 the Al_2O_3/TiO_2 ratio of terrigenous detritus falls within a very narrow range (typically
105 22–24), as terrigenous material is homogenised during sedimentation, albeit without
106 significant modification of certain original compositional features (Sugisaki et al., 1982;
107 Yamamoto, 1984; Yamamoto et al., 1986). The Al_2O_3/TiO_2 ratios of volcanogenic
108 materials, on the other hand, exhibit much wider ranges (10–130) and this characteristic

109 remains even after alteration, although the range may reduce somewhat (to ~0.6 times
110 the minimum ratio) (Yamamoto, 1984; Yamamoto et al., 1986). The $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratio is
111 thus expected to be useful in identifying the protolith of smectite-bearing shear zones.

112 To verify the feasibility of using the proposed method for the identification of
113 the protolith of smectite-bearing shear zones, this study applied the method to bedding-
114 parallel, clayey shear zones including soft clayey material intercalated within a Neogene
115 siliceous mudstone in the Horonobe area, northern Hokkaido, Japan. Within this
116 siliceous mudstone, numerous bedding-parallel slip surfaces have been developed by
117 flexural-slip folding, with some being associated with clayey shear zones including soft
118 clayey material (Ishii and Fukushima, 2006; Ishii, 2016; Hayano and Ishii, 2016; Ishii
119 and Furusawa, 2017). Some of the clayey shear zones have already been found to be
120 derived from tephra layers, via detailed mineralogical analyses of glass melt inclusions
121 in mineral grains (Ishii and Furusawa, 2017). However, the protolith of some of the
122 other clayey shear zones is still unknown.

123 In the siliceous mudstone, bedding-oblique slip surfaces are also developed
124 (Ishii et al., 2010; Ishii, 2012) and are rarely associated with clayey shear zones,
125 including soft clayey material observed in drill cores (Ishii, 2016; Ishii and Furusawa,
126 2017). However, the process of formation of soft clayey material in the bedding-oblique
127 shear zones is not fully understood, and therefore such structures are unsuitable for
128 verifying the proposed method. Thus, only bedding-parallel shear zones were
129 investigated in the present study. Other types of clayey zone, unrelated to shear zones,
130 have not been found in the siliceous mudstone.

131

132 **2. Geological setting**

133 The Horonobe area is located on the eastern margin of a Neogene–Quaternary
134 sedimentary basin on the western side of northern Hokkaido, Japan. It is within an
135 active Quaternary foreland fold-and-thrust belt, not far from the boundary between the
136 Eurasian and North American plates (Fig. 1). From stratigraphically lowest to highest,
137 the basin fill consists of the Masuporo, Wakkanai, Koetoi, Yuchi and Sarabetsu
138 formations (Figs 1 and 2; note that the Masuporo Formation is not shown). The
139 Wakkanai Formation, a Neogene siliceous mudstone, consists mainly of a single,
140 massive, homogeneous lithofacies (Mitsui and Taguchi, 1977; Iijima and Tada, 1981),
141 although weakly developed bedding planes are recognised in electrical-micro-imaging
142 borehole logs (Ishii et al., 2006). As for the mineral composition, the mudstone
143 comprises 40–50 wt% silica (mainly opal–cristobalite/tridymite: opal–CT), 19–33 wt%
144 clay minerals (mainly smectite and illite), 9–13 wt% quartz, 7–13 wt% feldspar, 0–2
145 wt% pyrite, and 0–1 wt% carbonate (Mazurek and Eggenberger, 2005; Hiraga and Ishii,
146 2008). Although vitric tephra layers do not occur in the Wakkanai Formation, they are
147 interbedded within the overlying diatomaceous mudstone containing biogenic opal-A
148 (the Koetoi Formation, Figs 1 and 2) (Ishii et al., 2008).

149 Flexural folding of the Wakkanai Formation occurred between 2.2 and 1.0 Ma in
150 response to regional E–W shortening, and subsequent uplift and denudation began at ~1
151 Ma (Ishii et al., 2008; Ishii, 2012). Displacements accrued along the bedding-parallel
152 slip surfaces and clayey shear zones were contemporaneous with flexural-slip folding
153 (Ishii and Fukushima, 2006; Ishii, 2016), with the displacements along the bedding-
154 parallel slip surfaces inferred to be on the scale of decimetres or less (Ishii and
155 Furusawa, 2017).

156

157 3. Samples and methods

158 Bedding-parallel, clayey shear zones including soft clayey material were
159 identified through outcrop observations (Loc. 1 in Fig. 1), and by core logging and
160 borehole wall image mapping in eight boreholes (HDB-1, -5, -6, -8, -9, -10, and -11,
161 and PB-V01 in Fig. 1). The clayey shear zones have a maximum thickness of 30 cm, are
162 light–dark grey in colour, and are associated with slip surfaces and/or fault breccias
163 (Figs 3 and 4; Ishii, 2016; Ishii and Furusawa, 2017). Shear fabrics, such as S-foliations,
164 were also confirmed within the shear zones and in the adjacent wall rocks during thin-
165 section analyses (Ishii, 2016; Ishii and Furusawa, 2017); the S-foliations are typically
166 highlighted by a scaly cleavage (Fig. 4b, c; Ishii, 2016; Ishii and Furusawa, 2017).
167 Although the soft clayey material in shear zones are sticky when wet (Fig. 4d), the
168 cracks highlighting the S-foliations may have been induced by desiccation of the core
169 samples in the shear zones (Fig. 4a; Ishii 2016; Ishii and Furusawa, 2017) and in the
170 adjacent wall rocks (Fig. 4e; Ishii, 2016). Clayey shear zones may be considered
171 analogous to clay gouge (Vrolijk and Pluijm, 1999), but the term “gouge” may also
172 indicate a mechanical formation process due to frictional wear or cataclasis (e.g.,
173 Engelder, 1974; Chester et al., 1985; Scholz, 1987). Because the process of formation of
174 some of the clayey shear zones has not yet been determined, the term “clayey shear
175 zones” is used to refer to discrete sheared layers composed of soft clayey material. This
176 study also analysed bedding-parallel shear zones devoid of soft clayey material, for
177 reference; although these zones do not contain soft clayey material, they are associated
178 with slip surfaces that are lined with cohesive, scaly material (Fig. 4f–h).

179 Table 1 and Fig. 5 show details of the shear zone and host-rock samples that
180 were analysed by X-ray diffraction (XRD) and X-ray fluorescence (XRF, for bulk

181 chemical composition analyses). Samples collected from the outcrop at Loc. 1 were
182 named using the format “KMH-X”, whereas the samples taken from drill cores were
183 named according to the borehole from which they came (e.g., HDB-1) and the depth at
184 which the samples were collected. For example, a sample taken from a depth of 599.4 m
185 in borehole HDB-1 was given the name “HDB-1-599.4”. The soft clayey material,
186 cohesive scaly material, and intact rock were sampled and analysed for the clayey shear
187 zones, shear zones without soft clayey material, and host rock, respectively. Details of
188 the analytical methods are provided below.

189

190 **3.1. XRD analysis**

191 The sm/(ill + sm) ratio for each sample was calculated from the relative
192 proportions of smectite and illite, as determined by XRD analyses of ethylene-
193 glycolated oriented powders. The relative proportions of smectite and illite that are used
194 to determine the sm/(ill + sm) ratio should be the ratio of smectite to illite in smectite–
195 illite mixed-layer clay species, considering that tephra-derived shear zones are expected
196 to have abundant pure smectite (Perry and Hower, 1970; Perry et al., 1976a, b; Hein and
197 Scholl, 1978). However, the method of determining the ratio of smectite to illite in
198 smectite–illite mixed-layer clay species using the peak positions in XRD profiles might
199 not be suitable for mudstone, as detrital illite may mask those peak positions (Reynolds
200 and Hower, 1970; Rettke, 1981). Although using the saddle peak on the low-angle side
201 of the 17 Å peak for smectite is also useful (Rettke, 1981; Inoue et al., 1989), this
202 method has not yet been routinely incorporated as a standard practice, including for
203 determining the baseline of intensity in the XRD profiles, and there is a lack of
204 associated analytical software. In the present study, as a conventional and practical

205 method, the relative proportions of the basal peak areas for smectite and illite in the
206 glycolated oriented samples were used and were weighted according to the method of
207 Biscaye (1965); i.e., using the area of the 17 Å peak for smectite and four times ($4 \times$)
208 the 10 Å peak area for illite. This weighting method has a maximum analytical error of
209 20 wt% (Underwood et al., 1993). As pointed out by Rettke (1981), the 17 Å peak can
210 include both the peaks of discrete smectite and of smectite–illite mixed-layer clay, and
211 the 10 Å peak can include both the peaks of discrete illite and of smectite–illite mixed-
212 layer clay. Thus, the ratio of smectite to illite calculated from the ratio of those peak
213 areas does not directly express the ratio of smectite to illite in smectite–illite mixed-
214 layer clay species. However, a difference in the ratio of smectite to illite in smectite–
215 illite mixed-layer clay may resultantly appear as a difference in the ratio of the 17 Å
216 peak area to the 10 Å peak area (Rettke, 1981), and such a trend can be confirmed in
217 numerous data previously reported (Hein & Scholl 1978; Tribble 1990; Guo and
218 Underwood, 2012). Thus, the present study employed the ratio of the 17 Å peak area to
219 the 10 Å peak area as an alternative indicator of the ratio of smectite to illite in
220 smectite–illite mixed-layer clay, and the $sm/(ill + sm)$ ratios were determined on the
221 basis of those peak areas.

222 Some of the XRD analyses were performed at the Hiruzen Institute for Geology
223 and Chronology, using a RIGAKU Rint-2500V diffractometer under the following
224 conditions: target = Cu; voltage = 40 kV; current = 160 mA; scan speed = 4° min^{-1} .
225 Other analyses were conducted at Earth Science Co. Ltd., Japan, using a Shimadzu
226 XRD-6000 diffractometer under the following conditions: target = Cu; voltage = 30 kV;
227 current = 20 mA; scan speed = 2° min^{-1} .

228

229 **3.2. XRF analysis**

230 The $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratio for each sample was determined using the Al_2O_3 and TiO_2
231 values yielded by XRF analyses. The XRF analyses were performed by ALS Canada
232 Ltd., using a sequential wavelength dispersive XRF spectrometer (PANalytical Axios).
233 The limit of detection (LOD) for each element in the XRF determinations was 0.01
234 wt%.

235

236 **4. Results and discussion**

237 Fig. 6 shows the $\text{sm}/(\text{ill} + \text{sm})$ ratios of the samples. The values for clayey shear
238 zones are $\sim 0.2\text{--}1.0$, whereas those of host-rock samples are $\sim 0.1\text{--}0.6$ (Fig. 6). The $\Delta 2\theta$
239 value between the peak positions of illite (001)/smectite (002) and illite (002)/smectite
240 (003) was close to 5.3° for the clayey shear-zone samples that exhibited the highest $\text{sm}/(\text{ill}$
241 $+ \text{sm})$ ratios (≥ 0.9) (Fig. 7a). This indicates that the smectite-bearing minerals in these
242 samples are almost pure smectite (Watanabe, 1988). This interpretation is consistent with
243 the size of the saddle peaks (Rettke, 1981; Inoue et al., 1989) on the low-angle side of the
244 17 \AA peak ($5^\circ 2\theta$, 001 peak) for the smectite in those samples, whereby the saddle/001
245 peak intensity ratios are very small compared with those of samples exhibiting lower
246 $\text{sm}/(\text{ill} + \text{sm})$ ratios (Fig. 7). Such pure smectite indicates a tephra-derived origin, and thus
247 the samples with $\text{sm}/(\text{ill} + \text{sm})$ values of ≥ 0.9 can be inferred to have been derived from
248 tephra layers (Perry et al., 1976a, b; Hein and Scholl, 1978; Tribble, 1990). The samples
249 with $\text{sm}/(\text{ill} + \text{sm})$ values of $\sim 0.7\text{--}0.9$ also have higher $\text{sm}/(\text{ill} + \text{sm})$ ratios than the host
250 rock samples, and thus are also inferred to have been derived from tephra layers. It is
251 suggested that their $\text{sm}/(\text{ill} + \text{sm})$ ratios have decreased from ≥ 0.9 to $\sim 0.7\text{--}0.9$ by
252 mineralogical disturbance and/or alteration during shear deformation (e.g., contamination

253 with the host rock materials, and/or syn-tectonic alteration). The $sm/(ill + sm)$ ratios
254 determined for the bedding-parallel shear zones devoid of soft clayey material are similar
255 to those of the host rock (Fig. 6).

256 Integration of the $sm/(ill + sm)$ and Al_2O_3/TiO_2 data shows that Al_2O_3/TiO_2 ratios
257 of the clayey shear samples with $sm/(ill + sm)$ ratios of ≥ 0.7 range widely from ~ 19 to
258 ~ 92 , whereas Al_2O_3/TiO_2 ratios of the clayey shear zone samples with $sm/(ill + sm)$ ratios
259 of < 0.7 fall into a much narrower range of ~ 22 – 25 , which is close to the range (~ 21 – 22)
260 of host-rock samples (Fig. 6). These patterns are observed regardless of the instrument or
261 XRD settings (Fig. 6). Considering that the clayey shear zones with $sm/(ill + sm)$ ratios
262 of ≥ 0.7 are inferred to have been derived from tephra layers, the distinct ranges of
263 Al_2O_3/TiO_2 values suggest that they can be used to identify the protolith of bedding-
264 parallel, clayey shear zones in argillaceous and siliceous marine sediments; i.e., clayey
265 shear zone samples with Al_2O_3/TiO_2 ratios different from those of the host rock are
266 inferred to be derived from tephra layers, as suggested previously (Yamamoto, 1984;
267 Yamamoto et al., 1986). However, in some cases the Al_2O_3/TiO_2 ratios of clayey shear
268 zones may intrinsically be the same as those of the host rock, even when shear zones are
269 derived from tephra layers, as shown by some samples in Fig. 6 with $sm/(ill + sm)$ ratios
270 of ≥ 0.7 (Yamamoto, 1984; Yamamoto et al., 1986). Al_2O_3/TiO_2 ratios of the bedding-
271 parallel shear zones devoid of soft clayey material are similar to those of the host rock
272 (Fig. 6).

273 Although the five samples with $sm/(ill + sm)$ ratios of < 0.7 (KMH-4, HDB-6-
274 458.4, HDB-9-104.5, HDB-10-434.6, and PB-V01-285.5) have Al_2O_3/TiO_2 ratios close
275 to those of the host-rock samples, the Al_2O_3/TiO_2 ratios of samples KMH-4, HDB-10-
276 434.6, and PB-V01-285.5 (24.52, 23.92, and 24.33, respectively; Table 1) are slightly

277 higher than the maximum value (22.43) of host-rock samples (Fig. 6). Although this
278 difference appears to be slight, it may be of significance considering the very small
279 measurement error (LOD = 0.01 wt%) of XRF analyses. It is therefore proposed that the
280 clayey shear zones from which these three samples were taken were also originally
281 derived from tephra layers and that their sm/(ill + sm) ratios decreased from ≥ 0.9 to < 0.7
282 via mineralogical disturbance and/or alteration during shear deformation. The clayey
283 shear zones associated with samples HDB-6-458.4 and HDB-9-104.5 have sm/(ill + sm)
284 and $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratios that are comparable with those of the host rock (Fig. 6). It is
285 therefore not possible to determine whether these two shear zones are derived from tephra
286 or host rock based solely on the sm/(ill + sm) and $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratios.

287 Ishii (2016), using detailed microscopic, mineralogical and geochemical analyses,
288 attributed the evolution of the clayey shear zone from which sample HDB-10-434.6 was
289 taken to shear deformation of the host rock; however, it was not clear why shear
290 deformation was initially localised within that particular horizon. The results outlined
291 above suggest that the horizon may have originated as a thin, altered tephra layer, which
292 was exploited as a plane of weakness during shear deformation, and subsequently evolved
293 into a broader, clayey shear zone.

294 Identification of the protolith of bedding-parallel smectite-bearing shear zones in
295 argillaceous and siliceous marine sediments is typically difficult, as the nature of the
296 protolith is largely lost via diagenesis and/or mineralogical disturbances and alteration
297 during shear deformation. However, the method proposed in this paper, based on the
298 integration of two indicators, sm/(sm + ill) and $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratios, is relatively simple to
299 implement and may be a useful starting point when attempting such discriminations.
300 Although high sm/(ill + sm) ratios may indicate a tephra-layer origin, the ratios are

301 susceptible to distortion by both deformation processes and XRD analytical errors. Shear-
302 zone samples yielding $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratios that differ from those of the host rock can also
303 be used as evidence that the shear zone was initially derived from a tephra horizon. In
304 that case, the data are unlikely to be affected by small measurement errors associated with
305 XRF analyses, but the $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratio may be intrinsically similar to that of the host
306 rock, even if the shear zones were derived from tephra layers. Both the proposed
307 indicators thus have their own pros and cons, but integration of the two may facilitate the
308 identification of the protolith of clayey shear zones in argillaceous and siliceous marine
309 mudstones.

310 Finally, it is noted that the applicability of the proposed method also depends on
311 the lithological heterogeneity of the host rock and the amount of pure smectite in the host
312 rock. The investigated host rock is a quite massive, lithologically homogeneous mudstone
313 that yields $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratios of ~21–22 (Fig. 6; Table 1), which are inferred to correspond
314 to the $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratio of terrigenous detritus that is homogeneously distributed
315 throughout the host rock. Pure smectite is also not particularly abundant, and the sm/(ill
316 + sm) ratio is <0.7 (Fig. 6; Table 1). However, if a host rock were highly heterogeneous
317 in lithofacies and contained a high concentration of detrital/ authigenic pure smectite due
318 to specific sedimentation conditions (e.g., Kameda et al., 2015) rather than to particular
319 ash fall events, it might be difficult to discriminate between tephra-derived shear zones
320 and host-rock-derived fault gouges, as the resultant sm/(ill + sm) ratios might be very
321 similar.

322

323 **5. Conclusions**

324 Samples of bedding-parallel, clayey shear zones intercalated within a folded

325 siliceous mudstone, plus host rock samples, were analysed using XRD and XRF to
326 evaluate the feasibility of using $sm/(ill + sm)$ and Al_2O_3/TiO_2 ratios as a tool for
327 identifying the protolith of bedding-parallel, clayey shear zones in argillaceous and
328 siliceous marine sediments, with results as follows.

- 329 • Tephra-derived shear zones were detected by their $sm/(ill + sm)$ ratios, which were
330 higher than those of the host rock; most of them also exhibited different
331 Al_2O_3/TiO_2 ratios to those of the host rock. This suggests that both $sm/(ill + sm)$
332 and Al_2O_3/TiO_2 ratios can be useful indicators of the protolith of smectite-bearing
333 shear zones.
- 334 • Although some clayey shear zones exhibited $sm/(ill + sm)$ ratios similar to those
335 of the host rock, they were also inferred to be derived from tephra layers, as their
336 Al_2O_3/TiO_2 values differed from those of the host rock.
- 337 • The method applied in this study represents a viable initial evaluation tool when
338 attempting to identify the protolith of smectite-bearing shear zones in argillaceous
339 and siliceous marine sediments.

340

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345 grammar and for helpful suggestions regarding the manuscript.

346

347 **Fig. 1** Geological map and cross-section of the Horonobe area (after Ishii, 2012),
348 showing the locations of boreholes and the outcrop illustrated in Fig. 3.

349

350 **Fig. 2** Schematic columnar section through the stratigraphy in the Horonobe area (Ishii
351 et al., 2010), including lithological descriptions of the main stratigraphic units. Sb =
352 Sarabetsu Formation; Yc = Yuchi Formation; Kt = Koetoi Formation; Wk = Wakkanai
353 Formation.

354

355 **Fig. 3** Photographs of bedding-parallel, clayey shear zones and bedding-parallel slip
356 surfaces devoid of soft clayey material exposed in outcrop (the location of the outcrop is
357 shown in Fig. 1). (a, b) KMH-1 and KMH-2; (c) KMH-3; (d) KMH-4; and (e) KMH-4.

358

359 **Fig. 4** (a) Photograph of the bedding-parallel, clayey shear zone with the greatest
360 thickness (HDB-1-432.9). The sample was taken from the ~5 cm thick, light-grey bottom
361 layer. (b, c) Photographs of typical bedding-parallel, clayey shear zones (HDB-6-444.6
362 and PB-V01-285.5). (d) Photograph of sticky, clayey material within the bedding-parallel,
363 clayey shear zone of PB-V01-395.8. (e) Photograph of desiccation cracks highlighting an
364 S-foliation observed in the hanging wall of the bedding-parallel, clayey shear zone of PB-
365 V01-472.0. (f) Photograph of a bedding-parallel shear zone without soft clayey material
366 but exhibiting cohesive scaly material (HDB-6-445.6). (g, c) Photographs of a bedding-
367 parallel shear zone without soft clayey material (HDB-11-883.5). Cohesive scaly material
368 is observed on the footwall slip surface.

369

370 **Fig. 5** Borehole logs summarising the stratigraphic distribution of bedding-parallel,
371 clayey shear zones and tephra layers (modified from Ishii and Furusawa, 2017). For
372 numbers with the format XXX.X(Y), XXX.X refers to the depth of occurrence (metres

373 below ground level) and Y refers to the thickness (centimetres). Data relating to tephra
374 layers are from Ishii et al. (2008). *Not analysed in this study, due to insufficient
375 sample.

376

377 **Fig. 6** $\text{Al}_2\text{O}_3/\text{TiO}_2$ vs sm/(ill + sm) diagram for the analysed samples. * sm/(ill + sm)
378 ratios based on XRD determinations performed by the Hiruzen Institute for Geology and
379 Chronology (this study); ** sm/(ill + sm) ratios based on XRD determinations performed
380 by Earth Science Co. Ltd. (this study); *** sm/(ill + sm) ratios from Milodowski et al.
381 (2004). BPSZ = Bedding-parallel shear zone.

382

383 **Fig. 7** Ethylene-glycolated XRD profiles of: (a, b) the bedding-parallel, clayey shear
384 zones of HDB-6-444.6 and PB-V01-285.5; (c) the bedding-parallel shear zone without
385 soft clayey material of HDB-11-883.5; and (d) the host rock of PB-V01-396.0.

386

387 **Table 1** Details of and analytical results for the shear zone and host rock samples analysed
388 in this study.

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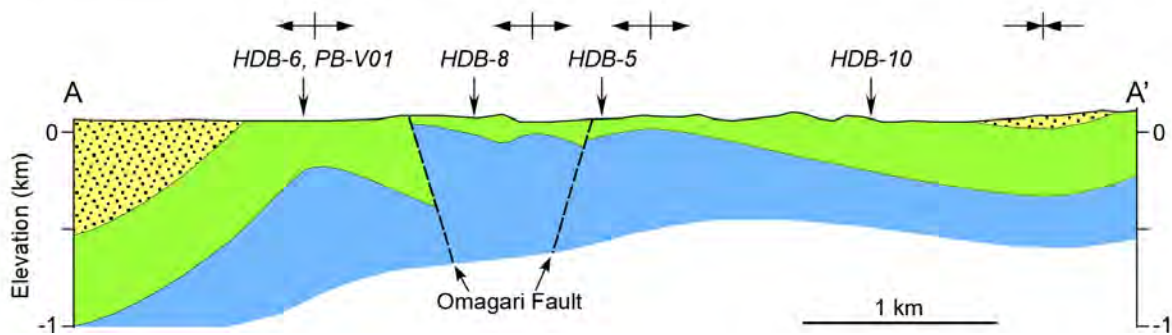
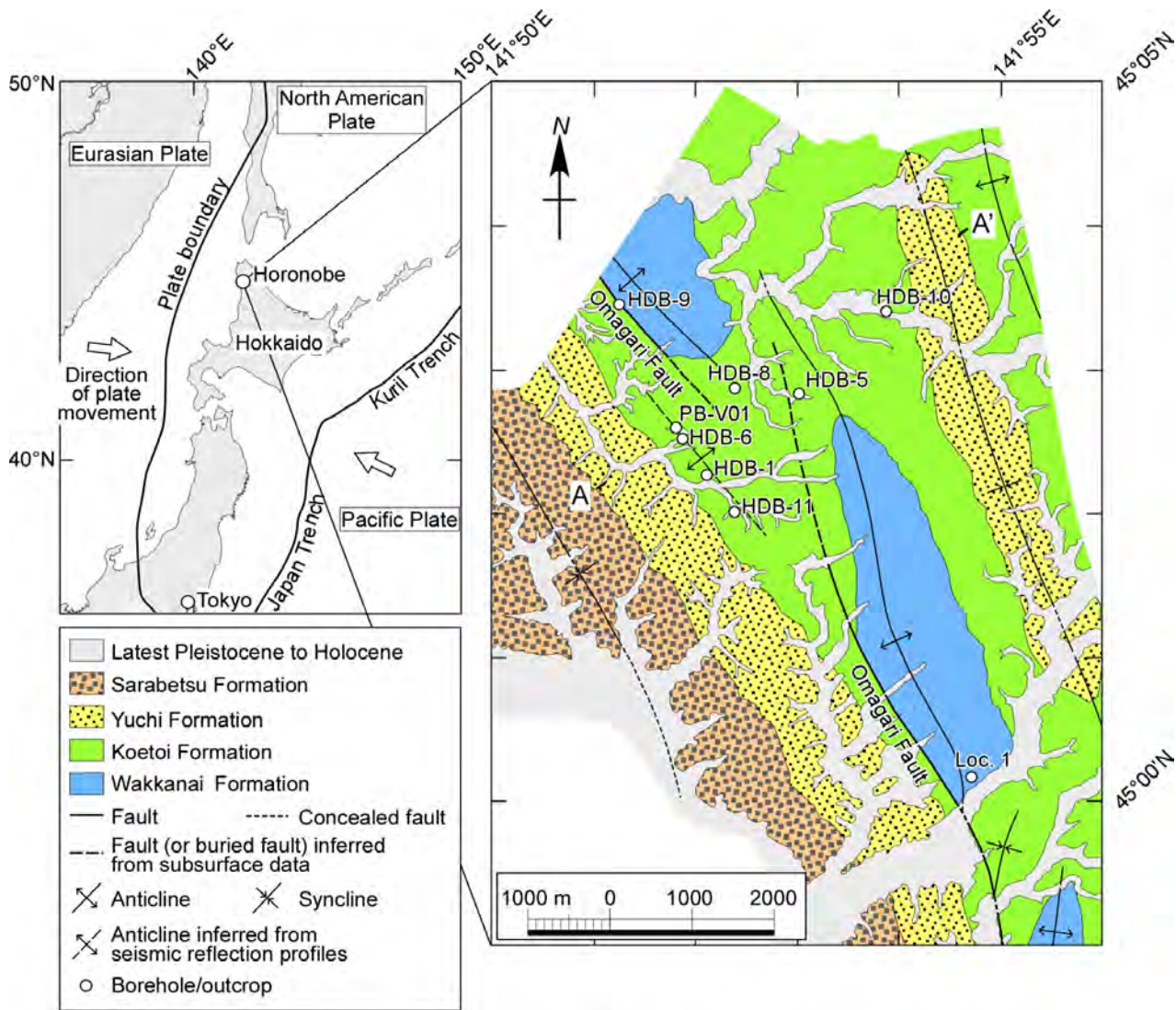
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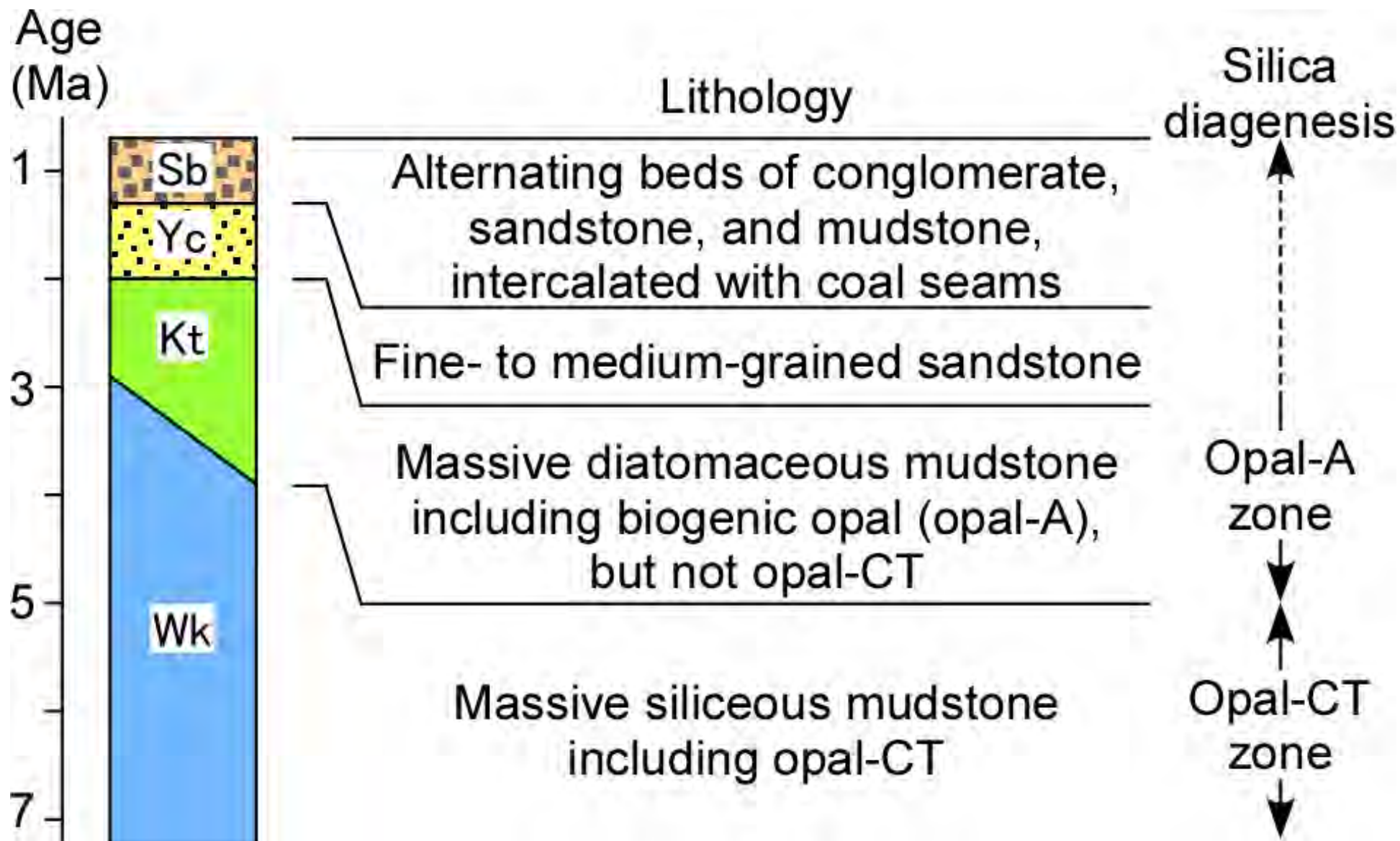
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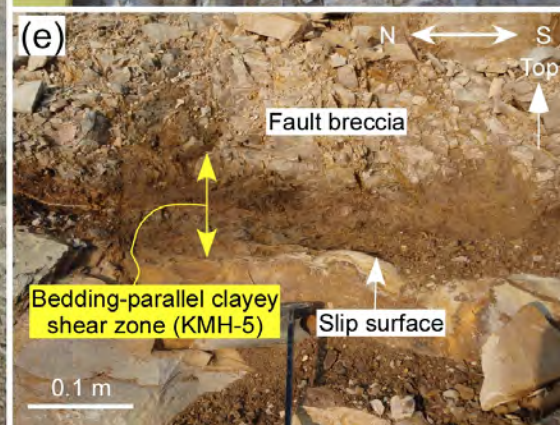
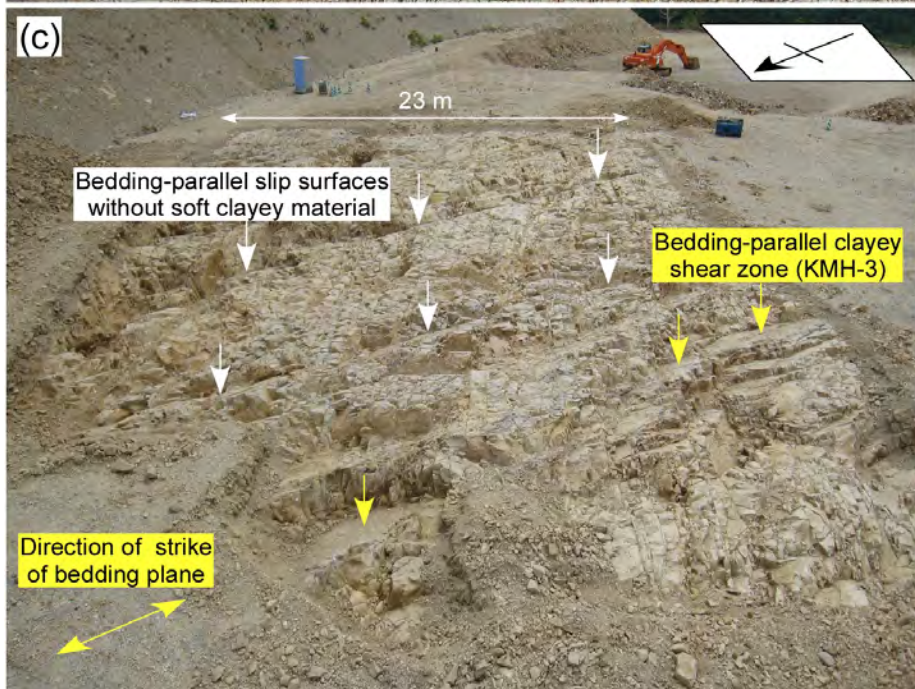
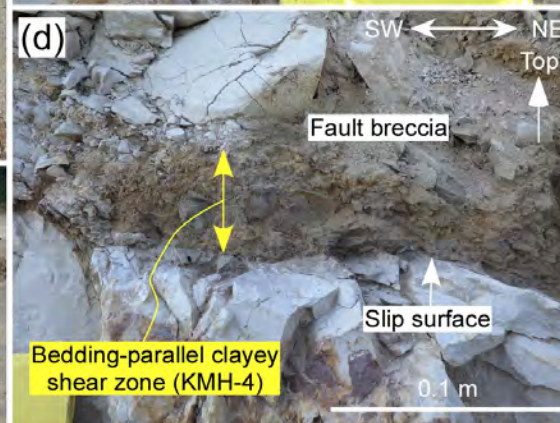
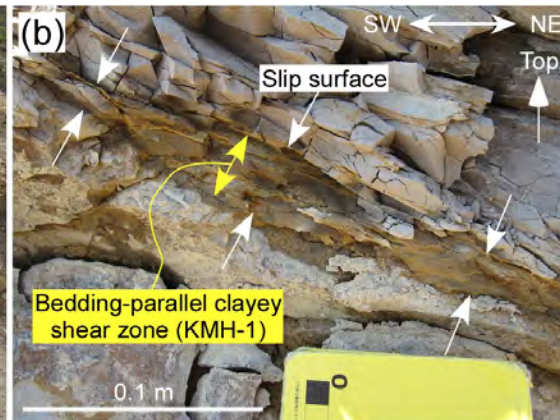
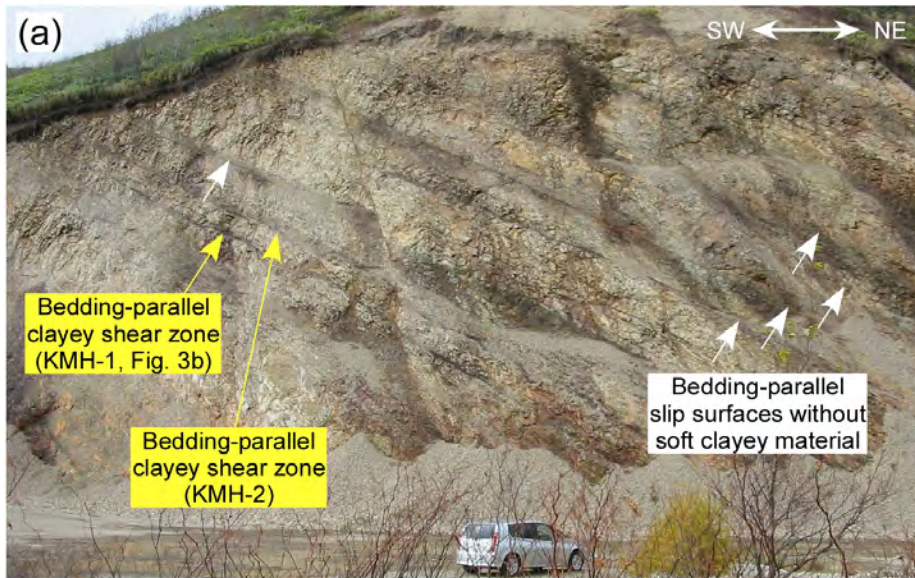
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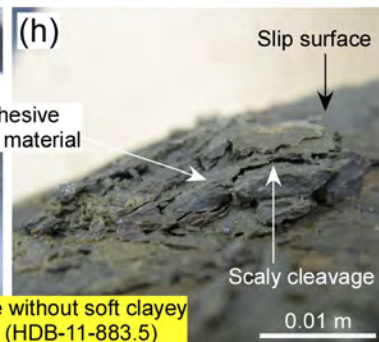
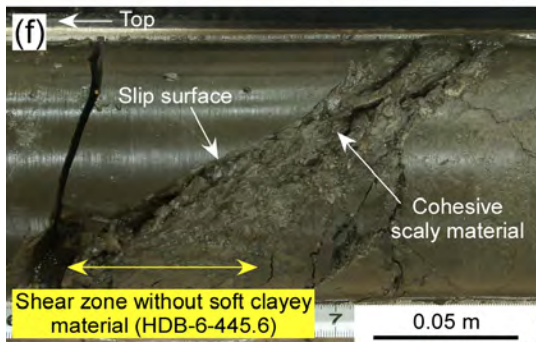
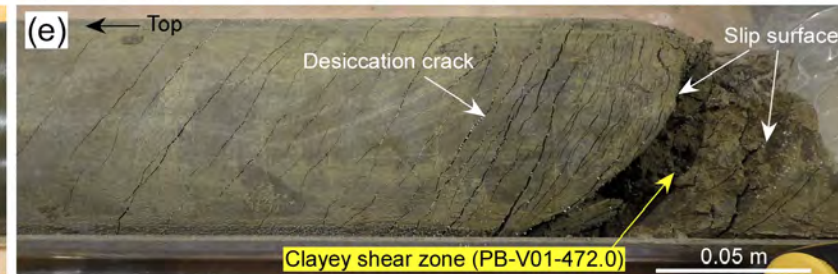
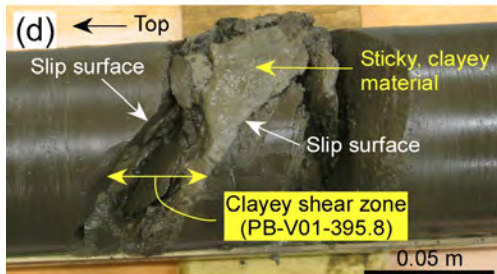
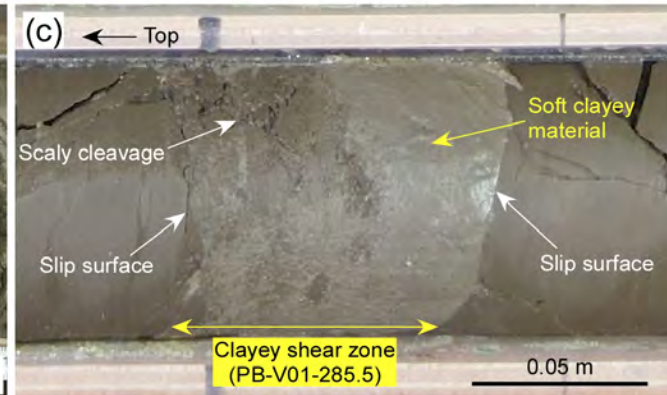
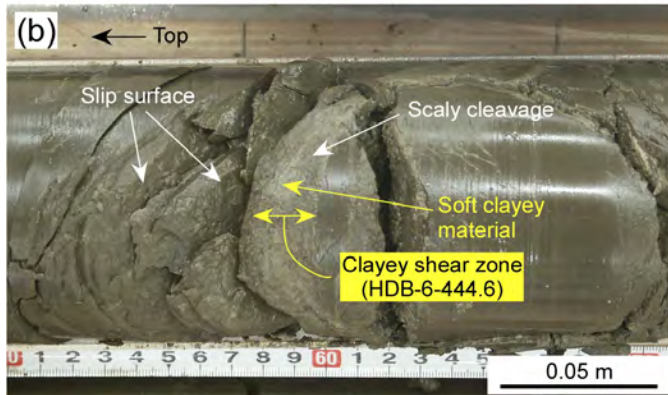
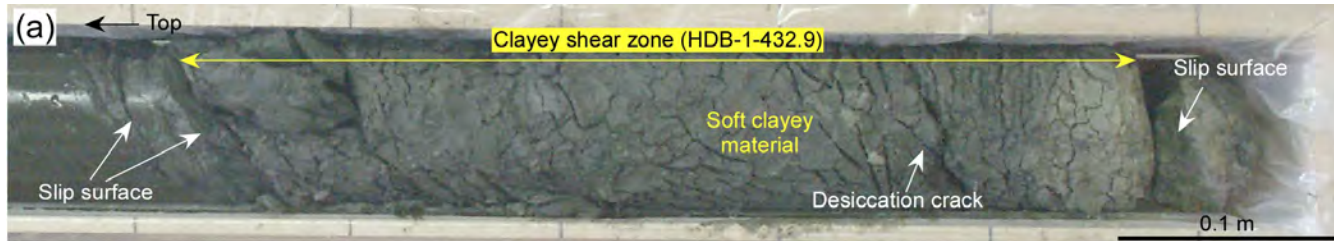
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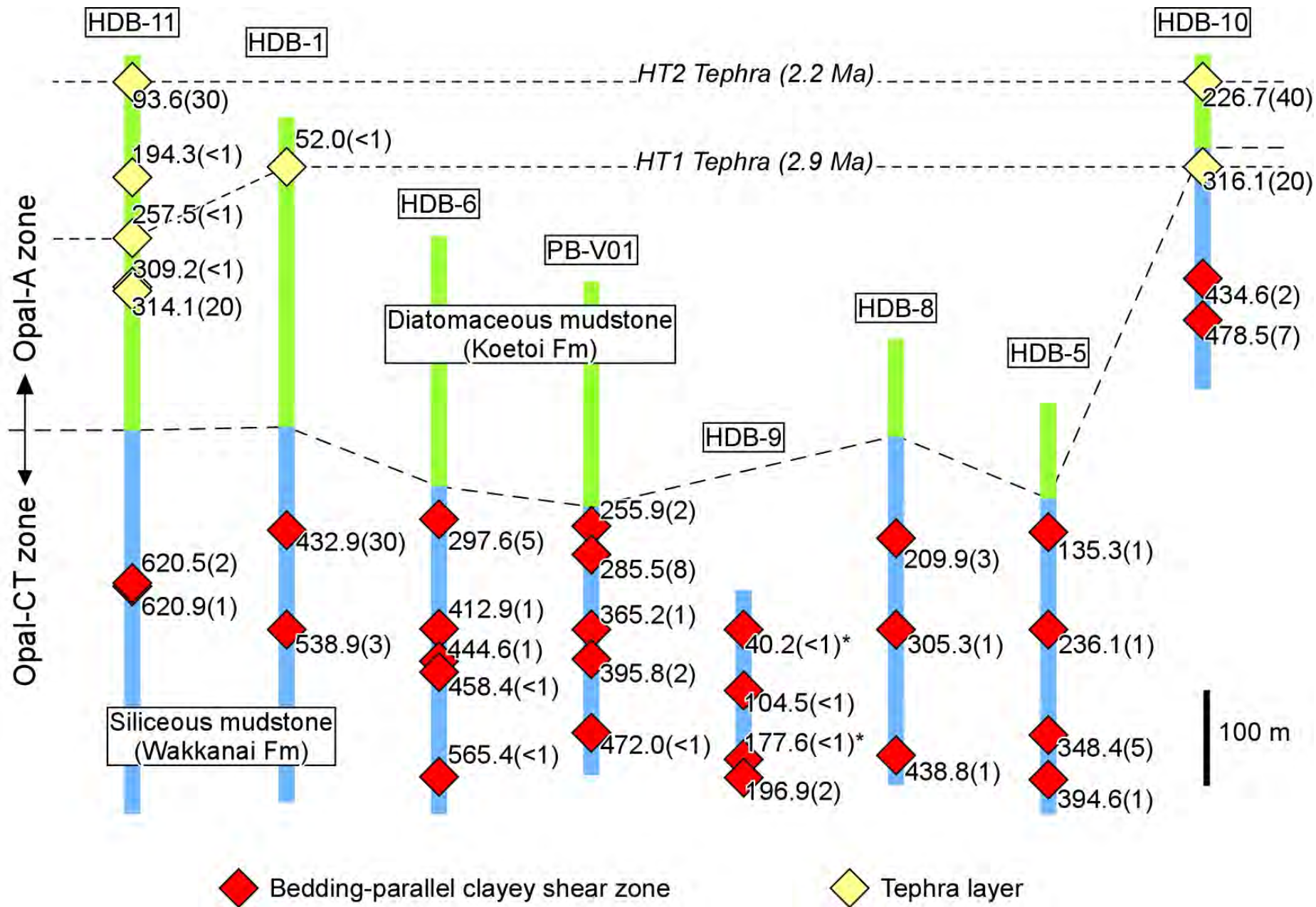
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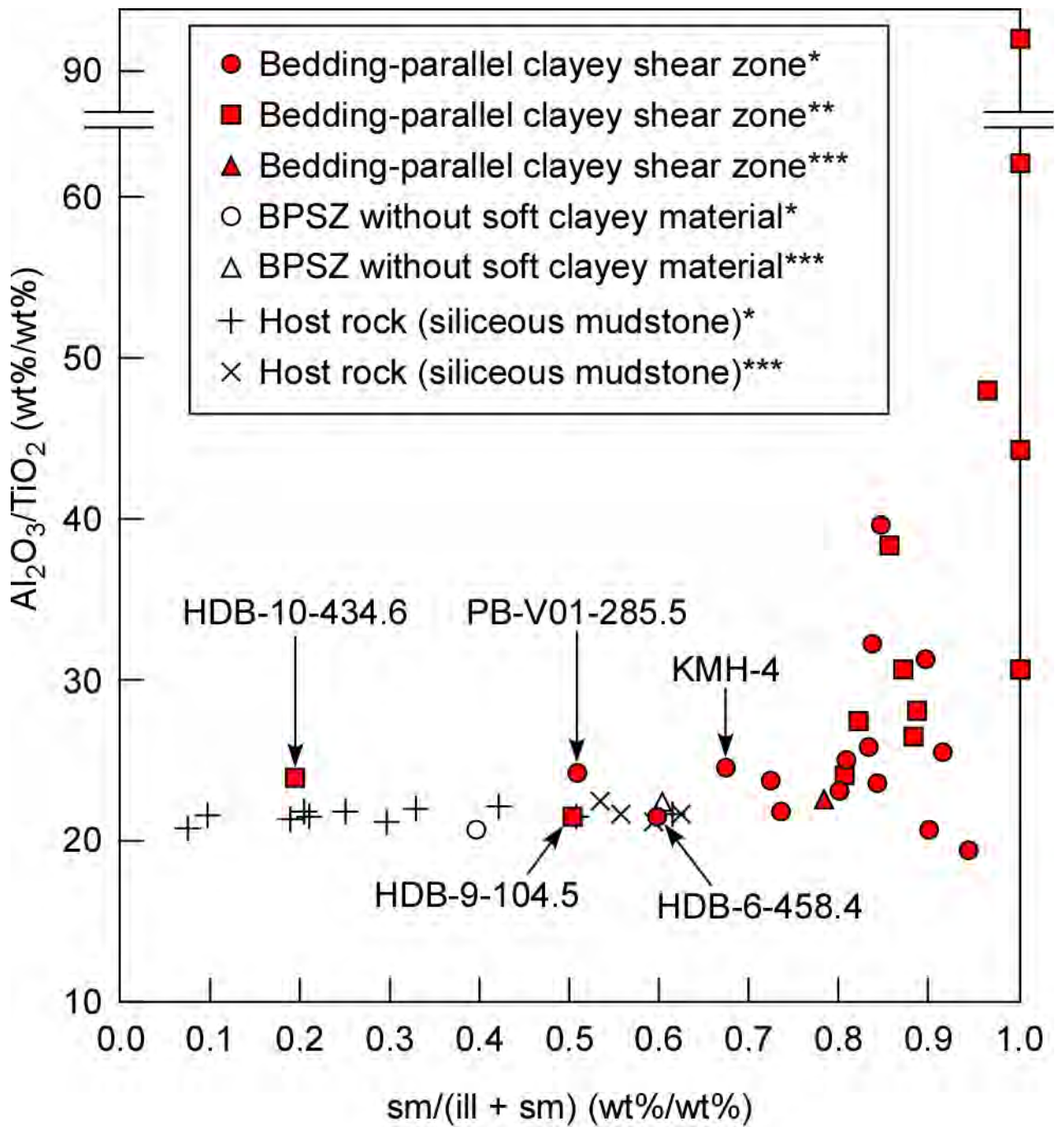












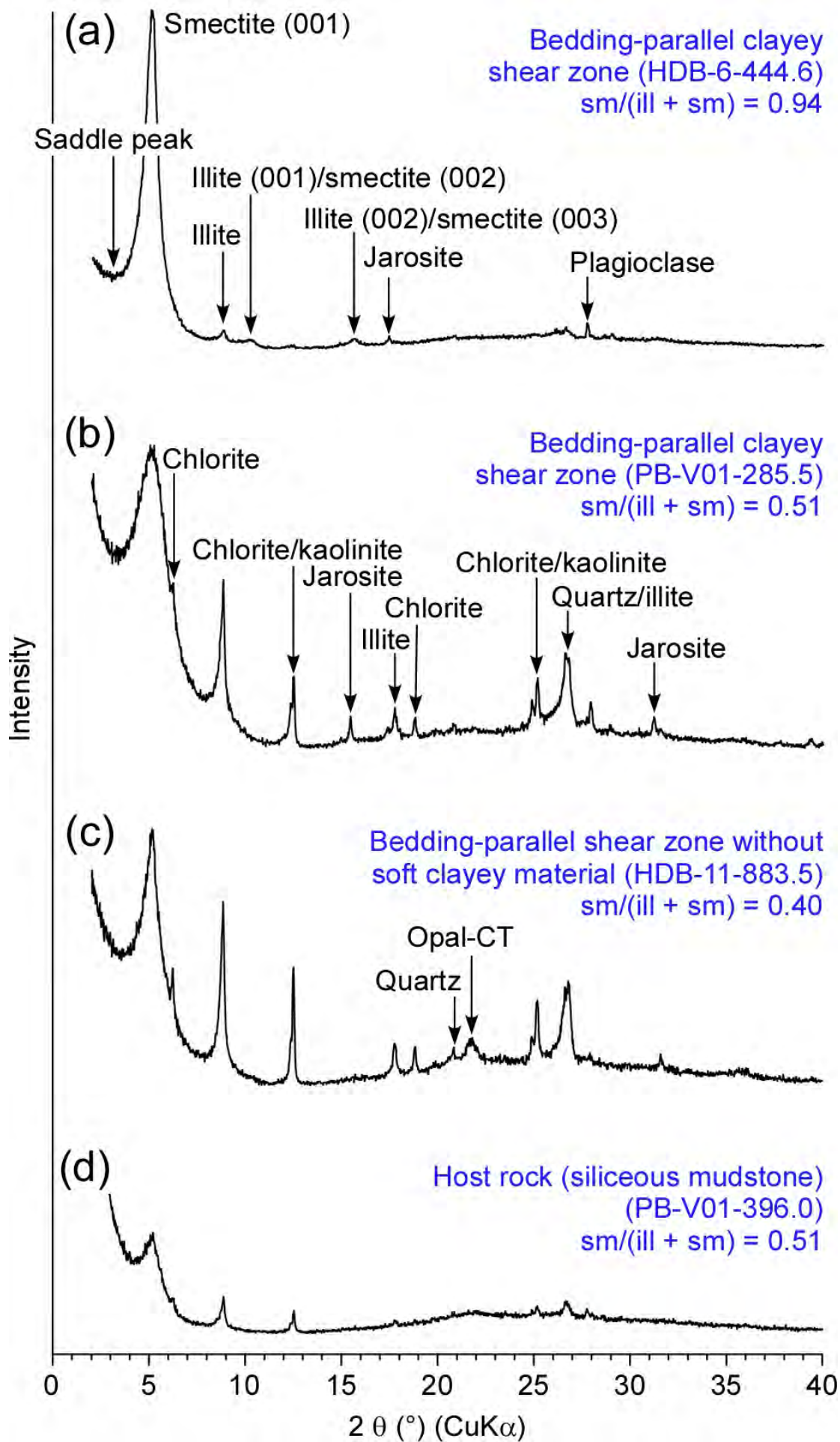


Table 1 Details of and analytical results for the shear zone and host rock samples analysed in this study.

Sample number	Thickness (m)	sm/(ill + sm) (wt%/wt%)	Al ₂ O ₃ /TiO ₂ (wt%/wt%)
Bedding-parallel clayey shear zone			
KMH-1	0.02	0.89**	28.15
KMH-2	0.03	1.00**	44.41
KMH-3	0.02	0.86**	38.41
KMH-4	0.03	0.67*	24.52
KMH-5	0.07	0.89*	31.29
HDB-1-432.9	0.3	1.00**	92.09*
HDB-1-538.9	0.03	0.88**	26.54
HDB-5-135.3	0.01	0.87**	30.68
HDB-5-236.1	0.01	0.91*	25.54
HDB-5-348.4	0.05	0.81*	25.00
HDB-5-394.6	0.01	0.83*	25.93
HDB-6-297.6	0.05	0.84*	32.32
HDB-6-412.9	0.01	1.00**	30.68***
HDB-6-444.6	0.01	0.94*	19.39
HDB-6-458.4	<0.01	0.60*	21.61
HDB-6-565.4	<0.01	0.78****	22.71****
HDB-8-209.9	0.03	0.96**	48.00
HDB-8-305.3	0.01	0.82**	27.54
HDB-8-438.8	0.01	0.80**	24.18
HDB-9-104.5	<0.01	0.50**	21.50*
HDB-9-196.9	0.02	0.72*	23.84
HDB-10-434.6	0.02	0.19**	23.92
HDB-10-478.5	0.07	1.00**	62.19
HDB-11-620.5	0.02	0.73*	21.85
HDB-11-620.9	0.01	0.80*	23.09
PB-V01-255.9	0.02	0.85*	39.62
PB-V01-285.5	0.08	0.51*	24.23
PB-V01-365.2	0.01	1.00**	30.68
PB-V01-395.8	0.02	0.90*	20.76
PB-V01-472.0	<0.01	0.84*	23.58
Bedding-parallel shear zone without soft clayey material			
HDB-6-445.6	0.02	0.60****	22.46****
HDB-11-883.5	<0.01	0.40*	20.76
Host rock (siliceous mudstone)			
HDB-6-326.1	-	0.19*	21.42
HDB-6-445.2	-	0.53****	22.43****
HDB-6-447.4	-	0.59****	21.14****
HDB-6-564.2	-	0.62****	21.71****
HDB-6-579.1	-	0.25*	21.91
HDB-8-111.0	-	0.56****	21.70****
HDB-8-197.5	-	0.21*	21.89
HDB-8-440.1	-	0.30*	21.29
HDB-9-72.10	-	0.08*	20.80
HDB-9-223.15	-	0.10*	21.60
HDB-9-321.4	-	0.42*	22.18
HDB-9-519.5	-	0.61*	21.71
HDB-11-598.1	-	0.33*	22.04
PB-V01-396.0	-	0.51*	21.59
PB-V01-518.3	-	0.21*	21.48

*Performed by Hiruzen Inst. for Geol. and Chron. (this study)

**Performed by Earth Science Co. Ltd. (this study)

***Data from Hiraga and Ishii (2008)

****Data from Milodowski et al. (2004)