

# Clay-coated sand grains in petroleum reservoirs: understanding their distribution via a modern analogue

L. J. Wooldridge\*<sup>1</sup>, R. H. Worden\*<sup>2</sup>, J. Griffiths, J. E. P. Utley

<sup>1</sup>*School of Environmental Sciences, University of Liverpool, Liverpool L69*

*3GP, UK*

\*<sup>1</sup>*Corresponding authors (e-mail: [luke.wooldridge@liv.ac.uk](mailto:luke.wooldridge@liv.ac.uk))*

\*<sup>2</sup>*Corresponding authors (e-mail: [r.worden@liv.ac.uk](mailto:r.worden@liv.ac.uk))*

## ABSTRACT

1

2 Clay coated grains can inhibit ubiquitous, porosity-occluding quartz cement in deeply buried  
3 sandstones and thus lead to anomalously high porosity. A moderate amount of clay that is  
4 distributed in sandstones as grain coats is good for reservoir quality in deeply buried  
5 sandstones. Being able to predict the distribution of clay coated sand grains within petroleum  
6 reservoirs is thus important to help find and exploit such anomalously good reservoir quality.  
7 Here we have adopted a high resolution, analogue approach, using the Ravenglass Estuary  
8 marginal-shallow marine system, in NW England, UK. Extensive geomorphic mapping,  
9 grain size analysis and bioturbation intensity counts were linked to a range of scanning  
10 electron microscopy techniques to characterise the distribution and origin of clay-coated sand  
11 grains within surface sediment. Our work shows that grain coats are common within this  
12 marginal-shallow marine system but they are heterogeneously distributed as a function of  
13 grain size, clay fraction and depositional facies. The distribution and characteristics of  
14 detrital-clay coated grains can be predicted with knowledge of specific depositional  
15 environment, clay fraction percentage and grain size. The most extensive detrital-clay coated  
16 grains are found within sediment composed of fine-grained sand containing 3.5 to 13.0 %  
17 clay fraction, associated with inner estuary tidal flat facies. Thus, against common  
18 convention, the work presented here suggests that, in deeply buried prospects, the best  
19 porosity may be found in fine-grained, clay-bearing inner tidal flat facies sands and not in  
20 coarse, clean channel fill and bar facies.

## INTRODUCTION

21

22 Porosity and permeability generally decrease with increasing depth of burial in sandstones,  
23 although a significant number of deeply buried sandstone reservoirs have unusually high  
24 porosity and permeability (Bloch et al. 2002). Such anomalously high porosity and  
25 permeability have most commonly been linked to the presence of chlorite clay coated grains  
26 that inhibit the growth of porosity-occluding quartz cement (Ajdukiewicz and Larese 2012;  
27 Ehrenberg 1993; Worden and Morad 2000).

28 The term clay coat encompasses both detrital and diagenetic origins (Ajdukiewicz and Larese  
29 2012). Detrital-clay coated grains occur at or near the surface of the sediment, and are the  
30 primary focus of this study.

31 Diagenetic clay coats either develop from the thermally-driven recrystallization of low-  
32 temperature, detrital precursor clay coats or they grow in situ due to the authigenic alteration  
33 of detrital or early diagenetic minerals interacting with the pore fluids during burial  
34 (Ajdukiewicz and Larese, 2012; Wise et al., 2001; Worden and Morad, 2003).

35 Chlorite and illite clay coatings are considered to preserve reservoir quality by reducing the  
36 nucleation area on detrital quartz grains that is available for authigenic quartz cementation  
37 (Ehrenberg 1993; Pittman et al. 1992). Porosity can be at least 10 % higher than expected  
38 where grain-coating clays are abundant (Ehrenberg 1993). Experiments undertaken by  
39 Ajdukiewicz and Larese (2012); Billault et al. (2003) and Lander et al. (2008) led to the  
40 conclusion that clay crystals within the clay coat act as barriers, inhibiting epitaxial quartz  
41 cement growth and subsequent coalescence to form thick quartz overgrowths. The primary  
42 factors controlling the effectiveness of clay coated grains for the inhibition of authigenic,  
43 porosity-occluding quartz cement are the extent, completeness and distribution of the detrital  
44 precursor clay coated grains (Billault et al. 2003).

45 Oil field-based studies which collectively show that clay coats are most common in fluvial to  
46 marginal marine sediments including: Jurassic sandstones on the Norwegian continental shelf  
47 (Bloch et al. 2002), Jurassic-Triassic fluvial, lacustrine-deltaic sandstones of the Ordos basin,  
48 China (Luo et al. 2009), marginal marine Jauf Formation, eastern Saudi Arabia (Al-Ramadan  
49 et al. 2004), the Upper Cretaceous Tuscaloosa Formation, USA (Pittman et al. 1992), and see  
50 review by Dowey et al. (2012). However there is no model capable of predicting the  
51 occurrence of clay coated grains or the degree of completeness of grain coats within fluvial to  
52 marginal marine sediments.

53 The positive influence of chlorite and illite clay-coated grains on reservoir quality in deeply  
54 buried sandstone has resulted in extensive reservoir core-based research (Ajdukiewicz et al.  
55 2010; Gould et al. 2010; Pittman et al. 1992) and laboratory experiments (Ajdukiewicz and  
56 Larese 2012; Billault et al. 2003; Pittman et al. 1992). Chlorite coated grains have been  
57 observed to inhibit quartz cement and the need to understand the origin of chlorite coated  
58 grains was the driving force that led to the current study. Notable chlorite clay coated  
59 reservoir units include the Tilje Formation, Norwegian continental shelf (Ehrenberg 1993),  
60 Tuscaloosa Formation, U.S. Gulf Coast (Ajdukiewicz and Larese 2012) and the Rotliegend  
61 Sandstone, northern Netherland (Gaupp and Okkerman 2011). Sandstones which contain  
62 illite and mixed layer illite-smectite clay coated grains have been less commonly advocated  
63 but include the Garn Formation, Mid-Norway (Storvoll et al. 2002), Williams Fork  
64 Formation, Colorado (Ozkan et al. 2011) and Jauf Formation, Eastern Saudi Arabia (Al-  
65 Ramadan et al. 2004; Cocker et al. 2003).

66 Aagaard et al. (2000) showed that low temperature, discontinuous, detrital-clay coated grains  
67 recrystallized during experiments at 90 °C to form thick, continuous, diagenetic clay coats  
68 that are morphologically consistent with naturally occurring reservoir examples. In some  
69 examples, euhedral clay minerals grow out into the pore from an underlying, unstructured

70 clay coat (Gould et al. 2010). Such clay coat stratigraphy could be the result of a detrital, or  
71 very early diagenetic, clay coat acting as a seed for deep burial diagenetic clay coat  
72 neoformation.

73 Despite the importance of being able to predict the occurrence and distribution of detrital-  
74 clay coated grains, there is no all-encompassing model that is useful for ranking prospects or  
75 populating reservoir models with the completeness of clay coats in marginal marine  
76 sandstones. Relatively little fundamental work has been undertaken on the controls on clay  
77 coat growth in sediments although Wilson (1992) and Matlack et al. (1989) undertook early  
78 studies focused upon environments (aeolian, marine-shelf, marginal marine, fluvial) in which  
79 clay-coated sand grains occur and potential mechanisms of formation (bioturbation,  
80 infiltration, inheritance). In order to predict anomalously high porosity in the subsurface,  
81 there is a need to focus on the origin and spatial distribution of detrital-clay coated grains  
82 since clay coats inhibit quartz cement in deeply buried sandstones (Bloch et al. 2002)  
83 Anomalously high porosity has also been shown to derive from other processes such as early  
84 oil charge, over pressure and microquartz coatings (Bloch et al. 2002).

85 The four main ways to develop a fundamental understanding of primary sedimentary  
86 environment and mineral distribution, and thus the processes that lead to clay coats, are: core-  
87 based studies, outcrop based studies, experimental studies and modern analogue studies.  
88 Core based studies have problems of limited spatial resolution of samples (wide spacing  
89 between wells and the lack of abundant cores in most fields) and the abiding uncertainty  
90 about both the primary mineralogy and exact environment of deposition due to subsequent  
91 diagenetic modifications. Outcrop based studies overcome the spatial resolution problem but  
92 typically suffer from weathering-related recent changes to mineralogy, plus outcrop-  
93 diagenesis studies routinely have problems in seeing through the long history of burial,  
94 heating and then uplift. We have here adopted a modern analogue approach, linking the

95 distribution of detrital-clay coated grains to sedimentary processes and characteristics (grain  
96 size, percentage clay fraction) and biological processes (bioturbation). The detailed study of  
97 sediment from modern environments permits a high resolution investigation into the  
98 distribution of detrital-clay coated grains, removing the limited spatial distribution,  
99 stratigraphic coverage and ambiguous depositional environment interpretations of subsurface  
100 core-based studies. This study addresses the following questions, focussed on the marginal-  
101 shallow marine Ravenglass Estuary system (Fig. 1).

- 102 1. What are the textural characteristics of detrital-clay coated grains within a  
103 modern marginal-shallow marine setting?
- 104 2. What are the mineralogical characteristics of clay-coated sand grains within a  
105 modern marginal-shallow marine setting?
- 106 3. How variable is the coverage of detrital-clay coated grains within a modern  
107 marginal marine system?
- 108 4. What controls the formation and distribution of detrital-clay coated grains?
- 109 5. Are the clay coats in this modern, marginal-shallow marine system, texturally  
110 comparable to other modern or subsurface examples?
- 111 6. What is the potential impact of using modern analogues for the prediction of  
112 reservoir quality in ancient and deeply buried sandstones from the same  
113 primary environment?

## 114 **STUDY SITE GEOMORPHOLOGY**

115 The Ravenglass Estuary is located in Cumbria, NW England. The mid to upper portions of  
116 the Ravenglass Estuary are fed by three rivers the Esk, Mite and Irt, with the lower, western  
117 part of the estuary connected by a single channel to the Irish Sea (Bousher 1999) (Fig. 1).  
118 Ravenglass sediment is quartz-dominated (Daneshvar 2011; Daneshvar and Worden 2016)

119 with depositional environments translatable to marginal-shallow marine petroleum reservoirs.  
120 Ravenglass is a modern analogue equivalent to the environment of deposition for many  
121 ancient and deeply buried, chlorite-coated sandstone reservoirs such as the tidally-influenced,  
122 shallow marine-deltaic Tilje Formation, Norway (Ehrenberg 1993), braid delta margin with  
123 foreshore and shoreface deposits Garn Formation, Norway (Storvoll et al. 2002), and  
124 shallow-marine to deltaic Lower Vicksburg Formation, USA (Grigsby 2001).

125 The 5.6 km<sup>2</sup> estuary has a maximum tidal range of 7.55 m and is 86% intertidal (Bousher  
126 1999; Lloyd et al. 2013). The estuary has extensive back barrier tidal flats and tidal bars,  
127 fringed by well-established saltmarsh vegetation (Bousher 1999). The estuary is connected to  
128 the Irish Sea through a single, 500m wide, tidal inlet that dissects a fringing coastal barrier  
129 which is topped with eolian dunes. The three fluvial channels, fluvial overbank, foreshore  
130 and ebb delta complex provide a complete fluvial to marine transect that we have investigated  
131 in terms of depositional environments, and detrital-clay coat abundance, with analysis of  
132 detrital-clay coat mineralogy (Fig. 1). Despite the high spring tidal range, the estuary  
133 contains geomorphological elements consistent with a mixed energy (wave-tide) regime,  
134 following the estuary classification scheme proposed by Ainsworth et al. (2011). This  
135 indicates a tidal hydrodynamic dominance within the inner estuary and wave-dominated  
136 processes occurring along the foreshore coastal side of the barrier spits.

137 The marginal- shallow marine Ravenglass system can be divided into fluvial-, estuary-,  
138 shallow marine- and eolian dune-dominated regimes, with the results of this study subdivided  
139 by sub-environment. The estuary has a clay mineral sediment assemblage consisting of  
140 chlorite, illite and kaolinite, largely derived from suspended fluvial sediment, originating  
141 from incision and weathering of the hinterland geology (Daneshvar 2011; Daneshvar and  
142 Worden 2016). The southern River Esk drains the Palaeozoic Eskdale Granite; the northern  
143 River Irt drains the Triassic Sherwood Sandstone Group and the Borrowdale Volcanic Group;

144 the central, but minor, River Mite drains a combination of Eskdale Granite, Triassic  
145 Sherwood Sandstone Group and the Borrowdale Volcanic Group (Moseley 1978).

## 146 **MATERIALS AND METHODS**

### 147 *Field-Based Mapping of the Estuary*

148 The estuary was initially mapped by identifying each depositional environment via world  
149 imagery and Google Earth. Extensive field mapping and sampling of all geomorphological  
150 elements enabled ground-truthing of mapped depositional elements and interpolation using  
151 ArcGIS. Tidal flats were further subdivided using the scheme proposed by Dyer (1979),  
152 based upon component volume clay fraction (< 2  $\mu\text{m}$  fraction):

153 0-10 % clay fraction is classed as sand flat,

154 10-30 % is muddy sand flat,

155 30-80 % is sandy mud flat.

156 Surface sediment grain size (approximately 2 cm depth) was determined at 3151 sites in the  
157 field using grain size cards and mapped using interpolated in ArcGIS. Lugworm faecal cast  
158 density (number per square metre) was recorded in the field using a 1m<sup>2</sup> quadrat, randomly  
159 thrown at 3182 sites within the estuary. Lugworm density was mapped across the entire  
160 intertidal exposed area, and also mapped using interpolated in ArcGIS. Polished thin sections  
161 were constructed from samples across a tidal flat succession to allow mineralogical  
162 quantification via automated scanning electron microscope-energy dispersive spectrometry  
163 (SEM-EDS). Sediment clay fraction mineralogy was established through X-ray diffraction  
164 analysis (XRD).

### 165 *Determination of Clay Coat Coverage*



166 This study is primarily focused on a suite of 181 surface sediment samples which were  
167 subject to grain coat petrography. The sample sites were chosen to provide sufficient spatial  
168 coverage and to encompass a fluvial- shallow marine transect incorporating all depositional  
169 environments. The clay size fraction volume (weight percentage) was established for 95 of  
170 the 181 sites.

171 Approximately 50 cm<sup>3</sup> of surface sediment was collected at each of the 181 sites. The  
172 sediment was then sub-sampled and dried at room temperature. Quantification of detrital  
173 clay coverage was achieved using scanning electron microscope (SEM) analysis of grain  
174 mounts on a 1 cm diameter stub. The grain mount stubs were examined by SEM petrography  
175 in backscattered electron (BSE) imaging.

176 A complete traverse across each SEM stub was collected by stitching together nine or more  
177 BSE images taken for each sample to produce a representative image of approximately 200  
178 grains. In comparison to thin-section based approaches for the study of grain coats, this  
179 approach permitted the investigation of detrital-clay coated grains in three dimensions. It  
180 also allowed for detailed classification of each sample (Fig. 2). Here we have adopted a  
181 novel approach that initially categorises the samples in terms of absence (group 1) or  
182 presence (groups 2-5) of clay coat and then subdivides those with coats into the degree of  
183 coat coverage (by surface area). Detrital-clay coats within this study were thus categorised  
184 into five principle classes:

- 185 1) Complete absence of attached clay coats.
- 186 2) Less than half of the grains have a small (~ 1-5 %) surface area of attached clay coats.
- 187 3) Every grain exhibits at least ~ 5-15 % surface area of attached clay coats.
- 188 4) Clay coats observed on every grain with the majority exhibiting extensive (~ 15-30 %)  
189 surface area grain coverage.
- 190 5) Extensive > 30 % surface area covered by clay coats observed on every grain.

191 To ensure reliability of the method and interpretation, duplicate SEM stub preparation and  
192 analysis was undertaken for 38 of the 181 samples to check the consistency of the  
193 classification method. We here note that all replicates faithfully reproduced the initial  
194 classification. Critical point drying (Jernigan and McAtee 1975) was not applied to the  
195 samples, owing to the absence of delicate fibrous clays associated with authigenic growth.

### 196 *Clay coat mineralogy*

197 Mineralogical quantification of clay coated sand grains from a mixed sand-mud tidal flat was  
198 undertaken via SEM-EDS using an FEI-QEMSCAN® (Armitage et al. 2016). This approach  
199 was selected to enable in-situ imaging of clay mineralogy, distribution characteristics and  
200 define the link between sediment clay mineralogy and that of clay coats. Three polished thin  
201 sections were constructed from surface sediment. The QEMSCAN® system comprises a  
202 scanning electron microscope coupled with fast energy dispersive spectrometers (EDS), a  
203 microanalyzer and an electronic processing unit, which integrates the data to provide  
204 information about the micron scale texture, chemical and mineral composition. The step size  
205 for the analysis was 1  $\mu\text{m}$  to ensure that the fine fraction in the sediment was analyzed as well  
206 as framework grains.

207 The data are presented as a combination of a backscatter secondary electron image, and fully  
208 quantitative mineralogical content image (framework grains) and quantitative clay  
209 mineralogy (total clay, illite, chlorite, kaolinite) to represent the sediment assemblage and  
210 component clay-coated sand grains.

### 211 *Determination of clay fraction*

212 The percentage of the clay fraction ( $< 2 \mu\text{m}$ ) was established via homogenised sediment sub-  
213 samples, dried at 60°C. A few grams of sample were added to 200ml of water and then  
214 ultrasonicated for 20 minutes with vigorous stirring at 5 minute intervals. Gravity settling

215 removed sand and silt sized particles, with the supernatant water (containing the clay grain  
216 sized particles) decanted and settled by centrifugation to obtain the clay fraction. The  
217 separated clay fraction was dried at 60°C, crushed in an agate pestle and mortar and then  
218 weighed, revealing the percentage clay fraction within the sediment sample.

### 219 *Determination of bulk sediment clay fraction mineralogy*

220 Classification of the clay fraction (<2 µm) mineralogy was undertaken by X-ray diffraction  
221 analysis (XRD). The clay sized fraction was detached from framework grains using an  
222 ultrasonic bath and isolated using centrifuge settling, at 5000 rpm for 10 minutes. The  
223 separated clay fraction was dried at 60 degrees and scanned as a randomly orientated powder,  
224 using a PANalytical X'Pert Pro MPD X-ray diffractometer. XRD analysis was carried out  
225 for the same samples that were mineralogy mapped through (SEM-EDS) analysis.

## 226 **RESULTS**

### 227 *Surface Sedimentary Characteristics and Distribution of Biological Activity*

228 Sedimentary environments were identified in the field, with further subdivision of the tidal  
229 flats based upon the lab-derived clay fraction data sets into sand-flat, muddy sand-flat and  
230 sandy mud-flat (Fig. 1).

231 High resolution, spatial distribution maps of sediment grain size reveal a wide range of mean  
232 grain sizes, from very fine to coarse sand sized sediment (Fig. 3A). There is a large scale  
233 trend of decreasing grain size away from the ocean, and smaller scale patterns of decreasing  
234 grain size with increasing distance from the main ebb channel, towards the tidal limit (Fig.  
235 3A).

236 A heterogeneous distribution of lugworms occurs in the estuary, as denoted by the widely  
237 varying lugworm cast density (Fig. 3C). The lugworm density at the sediment surface is

238 taken to indicate the intensity of bioturbation in the biotic zone of the sediment (McIlroy et  
239 al. 2003; Needham et al. 2005). The highest density of lugworms (31 to > 50 per m<sup>2</sup>) was  
240 observed within the outer sand tidal flats and non-vegetated tidal bar depositional  
241 environments (Figs. 1 and 3C). Comparing the sediment grain size map (Fig. 3A) to the  
242 lugworm population map (Fig. 3B) suggests that well-developed lugworm populations tend to  
243 be confined predominantly to the inner estuary where the sediment grain size tends to be  
244 between 88 and 177 µm.

245 The percentage sediment clay fraction data have been split into eight classes (Fig. 3B).  
246 Samples that contain > 1.5 % clay fraction are confined to the inner estuary. Samples that  
247 contain < 1.5 % clay fraction sit within the seaward portion of the estuary and outer tidal-flats  
248 (Fig. 3B). This pattern suggests that there is an inverse relationship between overall grain  
249 size and the amount of co-deposited clay fraction, i.e. there is an increased percentage of the  
250 clay fraction with decreasing grain size.

#### 251 *Clay fraction mineralogy*

252 The sediment samples have a clay fraction composed of illite, chlorite and kaolinite, with an  
253 average 7.6 % clay fraction in the sediment . X-ray diffraction shows that the clay fraction is  
254 dominated by illite (62 % of the clay fraction) clay with chlorite (17 % of the clay fraction)  
255 and kaolinite (21 % of the clay fraction) expressing similar values. (Fig.4).

#### 256 *Characteristics of Detrital-clay coats*

257 The observed detrital-clay coated grains are generally characterised by thin and discontinuous  
258 accumulations of individual but interlocking (overlapping and aligned clay platelets) clay  
259 minerals (Fig. 5). This study has focussed on the morphology of the coat and here we do not  
260 rely on a differentiation based on internal structure. Each sample was characterised by the  
261 morphology of the coat, the extent (degree) of grain coverage and abundance (proportion of

262 grains that contain coats) (Figs. 2). The clay coats occur on both convex and concave grain  
263 faces but the coats with the greatest thickness (maximum of about 5  $\mu\text{m}$ ) occur in grain  
264 indentations (Fig. 5G, 6E, 7). Clay coats occupy up to about 60 % surface area of individual  
265 grains in a given sample.

266 Detrital-clay coats are composed of individual interlocking clay minerals with a mixed  
267 mineralogy even along a singular ridge structure and a range of accessory impurities  
268 consisting of silt-sized quartz and bioclastic debris. Clay coats have been observed on all  
269 component framework grains within the sediment assemblage (quartz, feldspar, dolomite,  
270 calcite). The sand grains within this study are coated with a mixture of clay minerals (Fig. 7),  
271 dominated by illite (9.1 image area percentage), with minor chlorite (1.7 image area  
272 percentage) and kaolinite (1.1 image area percentage). There was no identified variability  
273 between clay mineralogy and component clay coat morphological classes (ridged, bridged,  
274 and clumped).

275 Detrital-clay coats occur with a variety of morphologies (Fig. 5). Here, we have grouped the  
276 samples into three principle morphological classes: ridged, bridged and clumped (Fig. 6).

277 Ridged clay coats consist of elongate intergrowths of plate-like clay minerals, orientated at  
278 high angles to the grain surface (Fig. 6A). Ridged coats have variable lengths ( $< 200 \mu\text{m}$ )  
279 and are preferentially observed upon relatively flat grain surfaces with minimal (silt)  
280 impurities. Ridged clay coated grains predominantly occur within the coarser, cleaner  
281 sediment assemblages that are associated with outer tidal flat and non-vegetated tidal bar  
282 environments.

283 Bridged clay coat textures occur between detrital grains. Bridged clay coats consist of  
284 elongate clay mineral aggregates that connect two grains. Bridged clay coats are relatively  
285 uncommon within surface sediment, possibly as result of the sampling procedure (Fig. 6B).

286 Clumped clay coats are highly variable both in extent and thickness (Fig. 5 and 6C).  
287 Clumped coatings are commonly reach sizes of up to 200  $\mu\text{m}$ , and contain silt-sized  
288 fragments as well as clay grade material. Clumped clay coats are most abundant within the  
289 upper estuary intertidal muddy sand flats, tidal bars and salt marsh depositional  
290 environments.

### 291 *Spatial Distribution of Detrital-Clay Coated Grains*

292 There is a high degree of variability in the distribution of detrital-clay coated grains, although  
293 most outer estuary sediment exhibits no more than minor attached clay coats (Fig. 8). The  
294 proportion of detrital-clay coated grains in the estuary tends to increase with distance from  
295 the open ocean and with distance from the main ebb channel. Clay coats are most extensive  
296 within the upper reaches of the three estuary channels. There is a strongly heterogeneous  
297 distribution of clay coat classes within the southern Esk estuary arm, while the northern Irt  
298 and central Mite estuary arms show more homogeneous distributions. In the central and sea-  
299 ward portions of the estuary, clay coats tend to be either absent or present in trace amounts  
300 (classes 1 and 2).

301 The surface sediment samples have here been plotted against depositional environment, with  
302 the aim of allowing the modern clay coat data to be compared to ancient, deeply buried  
303 sediments (Fig. 9). Detrital-clay coated grains are present within the fluvial channel  
304 sediments ranging from absent (class 1) to extensive (class 4) depending upon the position of  
305 the sample relative to the channel axis. Grains from inner meander and point bars samples  
306 typically have better developed clay coats representative of class 3-4. Grains from fluvial  
307 overbank samples tend to have the best developed detrital-clay coats on grains (class 3-5).

308 Inner estuary tidal depositional environments have a heterogeneous pattern of detrital-clay  
309 coated grain coverage. Clay coats are more extensively developed on detrital grains within

310 vegetated, as opposed to non-vegetated, tidal bars (Fig. 9). Tidal flats (sand flat, muddy sand  
311 flat, sandy mud flat) represent the only inner estuary depositional environment in which the  
312 full spectrum of clay coat grain coverage has been observed (classes 1 to 5). Samples that  
313 contain >10% clay fraction correspond to muddy sand flats. All grains in all samples from  
314 muddy sand flats contain some degree of clay coating. Samples from sandy mud flat (with  
315 >30% clay fraction) contain extensive (class 4-5) detrital-clay coat grain coverage. Saltmarsh  
316 sediment assemblages have uniformly well-developed detrital-clay coats (class 5). The  
317 observed variability in detrital-clay coat characteristics within tidal environments correlates  
318 to grain size; the more extensively developed detrital-clay coats (class 4-5) occur within very  
319 fine sand grain size dominated sediment (e.g. compare Fig. 8 to Fig 3A).

320 The samples from foreshore, ebb delta, tidal inlet and eolian dune depositional environments  
321 largely do not contain detrital-clay coated grains. Most samples from the vegetated, dune-  
322 topped spits and sheltered region within the tidal inlet contained no clay coat coverage (class  
323 1) and the remainder had minor clay coat coverage (class 2) (Fig. 9).

#### 324 *Detrital-Clay Coated Grains: Grain Size, Clay Fraction and Bioturbation*

325 Bin class intervals have been plotted against average grain size, percentage clay fraction and  
326 lugworm density (Fig. 10). This confirms that there is increasing percentage clay fraction  
327 with decreasing grain size. This also shows that increasing the percentage of the clay fraction  
328 correlates with increasing clay coat coverage (class number). Thus, clay coat class 3 (every  
329 grain exhibiting at least ~5-15 % attached clay coats) corresponds to sediment with a 2.5%  
330 clay fraction, while clay coat class 5 (extensive, >30 %, clay coats observed on every grain)  
331 corresponds to sediment with 10% clay fraction (Fig. 10). The coverage of clay coats does  
332 not seem to simply relate to lugworm density with the two highest clay coat classes found in  
333 association with low lugworm densities (Fig. 10).

334 Detrital-clay coats vary systematically within a given depositional environment (Fig. 9).  
335 Extensive detrital-clay coated grains are observed within the inner estuary tidal depositional  
336 environments, and they increase in extent towards the upper tidal limit (Figs. 8 and 9).  
337 Variations in grain size and clay fraction are secondary controls, with a lower fine sand grain  
338 size and >5 % clay fraction required to form uniform-extensive detrital-clay coats upon  
339 grains (class 3-5). There are negligible attached clay coats (class 1-2) observed within the  
340 high energy (upper fine-lower medium grain size), clean (<2% clay fraction) sand  
341 assemblages of the outer sand tidal flat, foreshore, ebb delta and eolian dune environments.

## 342 **DISCUSSION**

### 343 *Origin of Detrital-clay coat Textures*

344 The internal fabric and outer morphology of clay coats in deeply buried reservoir have been  
345 described in a few studies. Clay coats tend to be composed of an inner, densely packed,  
346 tangentially oriented, root layer that tends to be overlain by an outer coat composed of  
347 perpendicular euhedral flakes that grow into open pore spaces (Ajdukiewicz and Larese 2012;  
348 Wise et al. 2001). It has been proposed that the inner layers are the result of thermally-driven  
349 recrystallization of precursor detrital-clay coats (Aagaard et al. 2000; Billault et al. 2003).  
350 The clay coats from the Ravenglass Estuary, described here, are therefore analogues for the  
351 inner layer of clay coats reported from deeply buried reservoirs.

352 The observed ridged and bridged textures within this study (Fig. 6A, B, 7) have been reported  
353 previously in a range of case studies (Dowey 2013; Franks and Zwingmann 2010;  
354 Houseknecht 1992; Matlack et al. 1989; Moraes and De Ros 1992; Wilson 1992) and in  
355 synthesis experiments (Matlack et al. 1989). Ridged detrital-clay coat textures have been  
356 interpreted to derive from infiltration processes (Wilson 1992); bridge structures have been  
357 reported to form where ridges join two adjacent grains; initially bridged structures develop



358 distinct ridged texture when the sediment is disaggregated (Matlack et al. 1989). The  
359 sediment from the Ravenglass Estuary exhibits many of the textural characteristics that have  
360 been reported to result from clay infiltration into sand-dominated sediment (Wilson 1992).  
361 Infiltrated ridged detrital-clay coat textures have been reported within the Brazos River and  
362 Galveston marginal marine system, Texas (Matlack et al. 1989), as well as in the Anllons  
363 Estuary, Spain and Leiravogur Estuary, Iceland (Dowey 2013).

364 Infiltration occurs when water that contains suspended clay and silt flows into partially water-  
365 saturated sandy sediment. Within estuarine settings, infiltration is driven by a hydraulic  
366 gradient produced by the effect of the tidal range. This gradient drives suspended clay  
367 through the sediment at falling tide, towards the low tidal main ebb channel or during times  
368 of flooding due to increased rainfall in the hinterland (Santos et al. 2012). Reduction of flow  
369 velocity results in the deposition of the suspended clay and silt particles on to the sand grains  
370 (Dowey 2013; Worden and Morad 2003).

371 Clumped clay coat textures, that are comparable to those illustrated in this study (Fig. 6C),  
372 have been reported within the sediment of the Mandovi Estuary, India (Mohan Kessarkar et  
373 al. 2010), with similar clump sizes and textures. The subtropical Mandovi Estuary clay coats  
374 are composed of clay particles, bioclasts and organics that produce a heterogeneous  
375 mineralogy that is reported to be fluvially-derived from weathering products in the hinterland  
376 (Mohan Kessarkar et al. 2010). Clumped clay accumulations have also been reported within  
377 the fluvial-estuarine Rappahannock River, Virginia (Pierce and Nichols 1986). In both the  
378 Rappahannock and Mandovi examples, clumped textures were interpreted to originate from  
379 the deposition of biogenic (faecal) pellets and flocculated estuarine aggregates (Crone 1975)  
380 under stagnant pore water conditions in the estuary.

381 A comparison of clay coat textures found in the Ravenglass Estuary to other modern  
382 analogues, as well as experimental-based results, suggests that clay coats derive from a

383 combination of infiltration, resulting in the ridged-bridge textures, and flocculation with the  
384 deposition of biogenic faecal pellets resulting in clumped textures.

### 385 *Origin of detrital-clay coat mineralogy: internal or external to the estuary?*

386 The illite-dominated, mixed mineralogy of the clay-coated sand grains, determined by  
387 spatially-resolved SEM-EDS (Fig. 7), is consistent with the clay fraction mineralogy  
388 identified by XRD (Fig. 4). Had the clay coats formed in the hinterland, a much more varied  
389 clay-coat mineralogy would be expected than revealed by micro-studies using SEM-EDS and  
390 bulk-studies using XRD. Therefore, the observation that the clay coat mineralogy reflects the  
391 bulk clay mineralogy of the estuary implies that the clay coats were formed in the estuary  
392 itself rather than in the hinterland.

### 393 *Detrital-Clay Coat Distribution and Origin*

394 It has been reported that the primary depositional environment of a clastic sediment exerts a  
395 strong control on subsequent diagenetic processes, via the sediment texture, primary  
396 mineralogy, organic content and aqueous chemistry (Ehrenberg 1997; Morad et al. 2010;  
397 Worden and Morad 2003). The concept of a depositional control on the occurrence, type and  
398 subsequent diagenetic evolution of detrital-clay coats is reasonably well established (Bloch et  
399 al. 2002; Dowey et al. 2012; Ehrenberg 1993; Luo et al. 2009; Matlack et al. 1989). The  
400 results of this study confirm a depositional environment control but reveal, for the first time,  
401 systematic variability of the extent and completeness of clay coat coverage on a marginal  
402 marine depositional sub-environments scale.

### 403 *Comparison of Clay Coats in Ravenglass to Modern Estuary Studies*

404 In the Ravenglass marginal-shallow marine system, the most extensive detrital-clay coated  
405 grains are confined to the inner estuary tidal flat, tidal bar, saltmarsh and fluvial point bar

406 depositional environments. In contrast, detrital-clay coated grains are effectively absent  
407 within the coarse, clean sand that is associated with outer tidal flats, foreshore, dune topped  
408 spits, fluvial channel axis and main ebb channels. The distributions that are illustrated in  
409 Figures 8 to 10 have similarities to that of detrital-clay coated grain distribution along the  
410 Texas Gulf Coast, Galveston and within the Brazos River (Matlack et al. 1989). The Texas  
411 study reported clay coated grains from fluvial point bars, but an absence of detrital-clay  
412 coated grains within beach, delta beach, flood tidal delta, and delta plain surface sediments.  
413 Studies of the Anllons Estuary, Spain and Leiravogur Estuary, Iceland undertaken by Dowey  
414 (2013), support the observed distribution within this study, with detrital-clay coated grains  
415 being best developed within the less marine-influenced, middle and upper estuary reaches  
416 related to muddy tidal flats.

#### 417 *Comparison of Ravenglass Clay Coats to Ancient, Deeply Buried Clastic*

#### 418 *Sediment*

419 Reservoir studies, based on cored wells and interpretation of primary depositional  
420 environments, tend to be hampered by a lack of high resolution facies interpretation and  
421 relatively poor definition of the spatial and stratigraphic distribution of clay coated grains.  
422 To date, there is no published subsurface reservoir dataset that compares to the high spatial  
423 resolution and the complete certainty of the depositional environment used in this modern  
424 analogue study.

425 Although morphologically dissimilar, occurring as discontinuous clumps and ridges, broad  
426 textural and mineralogical similarities are identifiable between the precursor detrital-clay  
427 coats of this study and clay coats in diagenetically-altered reservoirs. Mixed mineralogy has  
428 been reported in several reservoirs, for example the Lower Cretaceous Mississauga  
429 Formation (Gould et al. 2010) and the Jurassic Garn formation (Storvoll et al. 2002), in which

430 the inner (tangential) diagenetic clay coats consist of a mixed illite-chlorite- mineralogy that  
431 is broadly similar to the mixed mineralogy of the detrital-clay coats in Ravenglass (Fig. 7 ).  
432 In the Upper Carboniferous submarine-fan and marine slope facies of the Arkoma Formation,  
433 USA it has been reported that muddy clay coated grain facies offer the best reservoir quality  
434 prospects compared to the well-sorted, clean sandstones (with little or no dispersed clays).  
435 (Houseknecht 1992). In the Arkoma Formation, amalgamated sandstone units contain beds  
436 with clay coated grains and no quartz overgrowth and adjacent clean sandstone beds that are  
437 devoid of clay coated grains but with pervasively quartz overgrowth, and therefore have  
438 negligible remaining porosity (Houseknecht 1992). Although the environment of deposition  
439 is different, the Arkoma example illustrates that a small quantity of clay that is co-deposited  
440 with sand can lead to improved reservoir quality.

## 441 **CONTROLS ON THE FORMATION AND DISTRIBUTION OF** 442 **DETRITAL-CLAY COATS**

443 In this study, we have produced a high resolution, modern analogue data set and established  
444 the distribution patterns of detrital-clay coats relative to surface sedimentary and biological  
445 facies. Percentage clay fraction, grain size and bioturbation have all been advocated as  
446 controls on the origin of clay coated grains in ancient, deeply buried sandstones.

### 447 *Role of Grain Size*

448 From this study, the observed inverse relationship of increasing detrital-clay coats coverage  
449 with decreasing grain size (Fig. 10) is consistent with previous observations by Wilson  
450 (1992), that clay coats are more extensively developed within finer grained sandstones in  
451 Holocene eolian dune and marine-shelf settings. The Permian-Carboniferous Unayzah  
452 sandstones, Saudi Arabia, also have a reported relationship between mean grain size and the

453 average percentage coverage of grains, with fine- to very fine-sandstone exhibiting the  
454 greatest degree of clay coat coverage (Shammari et al. 2010).

#### 455 *Role of Percentage Clay Fraction Control*

456 The role that percentage clay fraction ( $< 2 \mu\text{m}$ ) plays in the formation and distribution of  
457 detrital-clay coated grains is not well established within the literature. However, the Anllóns  
458 Estuary, Spain, has a clay fraction percentage that increases in marginal areas towards the  
459 upper tidal limit (Dowey 2013), consistent with the present study. The Anllóns example  
460 identified a trend comparable with Ravenglass of increasing clay coats coverage with  
461 increasing co-deposited clay fraction percentage (Dowey 2013). Furthermore, in the Texas  
462 Gulf Coast at Galveston and within the Brazos River, virtually no clay-coated grains occur in  
463 environments that are characterised by low suspended sediment concentrations (assumed here  
464 to be proportional to the percentage clay fraction) (Matlack et al. 1989).

#### 465 *Bioturbation Control*

466 Sediment bioturbation (specifically ingestion and excretion) has been experimentally shown  
467 to lead to the creation of clay coats on detrital sand grains (McIlroy et al. 2003; Needham et  
468 al. 2006; Needham et al. 2004; Needham et al. 2005). This mechanism works through the  
469 production of a mucus membrane on sand grains which then adheres finer clay-silt sized  
470 sediment on to the sand grains.

471 In the present study, the distribution of clay-coated grains does not spatially correlate with the  
472 degree of bioturbation observed in the estuary (compare Fig 3, 8 and see Fig. 10). It is also  
473 notable that a similar conclusion can be drawn from the Lower Cretaceous Missisauga  
474 Formation, Scotian Basin, where the coverage of clay coated grains does not positively  
475 correlate with the degree of bioturbation (Gould et al. 2012). The lack of correlation between  
476 bioturbation and the degree of clay coated grains in this study may result from the limited

477 environmental grain size niche of the utilized lugworm biogenic proxy. To address this, a  
478 focused study on the abundance distribution of estuarine macro- and microorganisms would  
479 be required.

## 480 **IMPLICATIONS FOR HYDROCARBON EXPLORATION**

### 481 *Target Reservoir Quality Prospects*

482 At depths > 3 km (temperatures > 90°C) pervasive authigenic quartz typically starts to  
483 become a dominant cement in sandstones (Bloch et al. 2002). Such sandstones risk becoming  
484 extensively quartz cemented if grain coats are absent, or poorly developed, upon grain  
485 surfaces (Ajdukiewicz and Larese 2012).

486 Based upon the surface distribution patterns of detrital-clay coats presented here (Figs. 8-10),  
487 the best prospects for anomalously high reservoir quality due to the presence of clay coated  
488 grains in deeply buried sandstones (> 3km), should be sought within the fine sand sized  
489 sediment that also contains approximately 5% clay fraction percentage. Specifically targeting  
490 clay-bearing sandstones in the hunt for elevated porosity is against common convention,  
491 which would typically target the cleanest, most clay-free sandstones. Our interpreted  
492 optimum value of approximately 5 % clay fraction is based upon the likelihood of producing  
493 extensive clay coats within sandstones. However we note that highly elevated clay content  
494 would of course produce a detrimental effect on permeability and porosity (see next section).  
495 Sites with fine sand-sized sediment that also contain approximately 5% clay fraction  
496 correspond to inner estuary tidal bar, tidal flat and fluvial point bar facies in the Ravenglass  
497 system. In contrast, coarse, clean sand from tidal channels, outer sand flat and foreshore  
498 facies would, upon deep burial, potentially experience pervasive quartz cementation due to  
499 the lack of inhibiting detrital-clay coated grains if the sediment reached temperatures  
500 sufficient for quartz cementation.

501 *Goldilocks Zone of Optimum Detrital-Clay Coat Coverage*

502 The high resolution, marginal-shallow marine model for the distribution of detrital clay-  
503 coated grains presented in this study may be used, by analogy, to help in the prediction of  
504 clay coated sandstones in the deep subsurface. Too much clay is highly detrimental to  
505 sandstone reservoir quality (Armitage et al. 2016; Houseknecht and Pittman 1992; Worden  
506 and Morad 2003) since abundant clay minerals fill pores and block pore throats between sand  
507 grains. The quantity of clay in a sandstone that is sufficient to coat grains (and thus inhibit  
508 quartz cement) but not enough to block pore throats, surprisingly, remains poorly resolved,  
509 and is addressed below.

510 Bloch et al. (2002) noted that a minor amount of clay (as little as 1 to 2% of the rock volume)  
511 can coat a relatively large surface area of sandstone grains, but the optimum amount for  
512 specific clay minerals has not been precisely defined. Here examples from previous studies  
513 were used to help constrain broad percentages of total clay quantities as clay coats that can  
514 lead to the development of diagenetic coats which can successfully inhibit quartz cement.  
515 Pittman et al. (1992) suggested an optimum range of 5 to 13 % sediment volume of clays  
516 occurring as chlorite grain coats for the Tuscaloosa Formation and 4 to 7 % for the Berea  
517 Sandstone. Heald and Baker (1977) reported an optimum range of 3.5 to 6.5 % volume of  
518 illite clay coats for reservoir quality within the Rose Run sandstone.

519 Here, we tentatively propose (from the observed association of clay fraction occurring  
520 predominantly as clay coatings) lower and upper threshold values of 3.5 and 13.0 % total  
521 volume of clay minerals (chlorite, illite and mixed) as the optimum range for the eventual  
522 development of clay coats that can form continuous barriers that prevent quartz cementation  
523 and so preserve reservoir quality. Using the 3.5 to 13.0 % range of total volume of clays, we  
524 have mapped out regions within the Ravenglass Estuary that would lead to the best reservoir

525 quality, were this sedimentary system to be deeply buried (Fig. 11). These optimum regions,  
526 termed “Goldilocks zones”, encompass the central tidal flat region, non-vegetated and upper  
527 estuary tidal bar depositional environments.

528

## CONCLUSIONS

- 529 1. The work presented here, from the Ravenglass Estuary, UK, represents the first high  
530 resolution study of the distribution of detrital-clay coated grains within a modern  
531 marginal-shallow marine setting.
- 532 2. Sedimentary environment is the main control on the absolute quantity of clay minerals  
533 and detrital-clay coat sand grain coverage in these sand-dominated sediments.
- 534 3. Detrital-clay coats in recent sediments have discontinuous ridged, bridged and  
535 clumped textural morphologies. The coats on sand grains are formed of individual  
536 interlocking clay minerals with silt-sized lithic and bioclastic accessory components  
537 and were probably derived from a combination of infiltration (of clay-bearing water  
538 into sand-dominated sediment), flocculation and biogenic processes. Clay coats range  
539 from being absent to covering > 30% of sand grain surfaces in a given sample.
- 540 4. The observation that the illite-chlorite-kaolinite clay coat mineralogy reflects the bulk  
541 clay mineralogy of the estuary implies that the clay coats were formed in situ within  
542 the estuary rather than in the hinterland.
- 543 5. The distribution of detrital-clay coated grains is primarily a function of sediment grain  
544 size and clay fraction percentage. In the Ravenglass case study, a sediment  
545 assemblage composed of fine-grained sand containing > 5 % clay fraction percentage  
546 is necessary for the development of uniform-well developed clay coats on detrital  
547 grains.



548       **6.** The best prospects for anomalously high reservoir quality in deeply buried marginal  
549       marine sandstones (i.e. with inhibited growth of quartz cement) should most likely be  
550       sought within clay-rich inner estuary tidal facies.

551 **FIGURE CAPTIONS**

552 Figure 1. Location maps. A) The Ravenglass Estuary, within the UK. B) Regional map  
553 showing the study area and component depositional environments. Tidal flats have been  
554 subdivided based upon their component clay fraction ( $< 2 \mu\text{m}$ ); 0-10 % sand flat, 10-30 %  
555 muddy sand flat, 30-80 % sandy mud flat. Classification modified from the scheme initially  
556 proposed by Dyer (1979). The black square indicates the sediment sample location from  
557 which clay coat (SEM-EDS) and sediment clay fraction (XRD) mineralogical analyses (Fig.4  
558 and Fig. 5) were undertaken.

559 Figure 2. SEM electron images showing the variable extent of attached clay coats observed  
560 within surface sediment samples, which define the basis of the utilized classification scheme.  
561 1) Complete absence of clay coats. 2) ~1 to 5 % attached clays on less than half of the grains.  
562 3) Every grain exhibits ~5 to 15 % clay coats coverage. 4) Clay coats observed on every grain  
563 with the majority exhibiting extensive ~15 to 30 % coverage. 5) Extensive,  $> 30$  % clay  
564 coats coverage observed upon every grain.

565 Figure 3. Distribution maps of surface sedimentary features. A) High resolution map of  
566 surface sediment grain size ( $n = 3150$ ). B) Surface distribution of clay fraction percentage ( $n$   
567  $= 2000$ ) C) High resolution map of lugworm population in surface sediment ( $n = 3182$ ).

568 Figure 4. X-ray diffractogram used to quantify the bulk sediment clay fraction mineralogy of  
569 surface sediment within the Ravenglass Estuary (for location, see Fig. 1).

570 Figure 5. Representative SEM electron images of the textural characteristics of surface clay  
571 coated sand grains. Arrows indicate regions of clay coat coverage. Note the extent of the  
572 ridged clay coat morphologies composed of interlocking and aligned clay particles (A, B, C,  
573 and E). The clumped clay coat aggregates composed of clay minerals, lithics and organics  
574 are illustrated within (A, D, G, and F). The textural clay coat characteristic of extending pore

575 ward are observed within (A, C and F) and with greatest accumulation (thickness) observed  
576 within grain indentations (E and G).

577 Figure 6. Clay coat textures showing the main morphological feature classification observed  
578 within surface sediment samples. A) Ridged clay coat. B) Bridged clay coat structure. C)  
579 Clumped clay coat. Note greatest thickness of attached coating within the grain indentation  
580 (enlarged in F). Arrows indicate regions of clay coat coverage.

581 Figure 7. Scanning electron microscope-energy dispersive spectrometry (SEM-EDS) image,  
582 showing clay coat and bulk sediment mineralogy for within muddy sand flat sediment (for  
583 location, see Fig. 1). A) Backscattered electron image. B) SEM-EDS image of framework  
584 grain mineralogy. C) SEM-EDS image of the component clay fraction mineralogy. D to F)  
585 SEM-EDS images of the distribution of illite, chlorite, and kaolinite. Arrows indicate regions  
586 of attached clay coating.

587 Figure 8. Distribution map of surface clay coated sand grains within the Ravenglass Estuary  
588 (n = 195). Plotted are the surface distribution of classified areas in light grey signify at least  
589 partial clay coat coverage, with dark grey-black regions indicating extensive surface clay  
590 coated sand grains.

591 Figure 9. Frequency histograms for all sediment samples, divided by depositional  
592 environment and clay coat bin class: (A) Total number of samples in each clay coat class. (B)  
593 Normalised data to reveal relative importance of different environments for optimum clay  
594 coat coverage. Clay coat class 1 and 2 have minimal to complete absence of clay coats.

595 Figure 10. Average grain size, lugworm population and clay fraction percentage plots for  
596 each representative clay coat bin class.

597 Figure 11. Distribution map indicating the literature-constrained Goldilocks zone of the  
598 optimum quantity of total clay (i.e. detrital-clay coated sand grains inhibiting quartz cement  
599 but not blocking pore throats).

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