ABSTRACT

The specific mineralogy of clay grain coats controls the ability of the coat to inhibit quartz cementation in sandstones during prolonged burial and heating. How and why clay-coat mineralogy varies across marginal marine systems is poorly understood, even though these eogenetic phenomena strongly influence subsequent mesodiagenesis and reservoir quality. The novel development of the ability to predict the distribution of clay-coat mineralogy would represent an important development for sandstone reservoir quality prediction. In marginal marine sediments, clay minerals occur as grain-coats, flocs, mud intraclasts, clay-rich rock fragments or as dispersed material. However, the relationships between clay mineralogy, the amount of clay, and its distribution is poorly understood. This study focused on the Ravenglass Estuary, UK. The key aim was to develop and apply a novel methodology utilising scanning electron microscope – energy dispersive spectrometry, for the first time, on grain coats in modern sediments, to differentiate the clay-coat mineral signature from that of the bulk sediment, and reveal the distribution of clay minerals across marginal marine sediments. The study showed that marginal marine sediments principally have their clay mineral assemblage present as clay-coats on sand grains. These clay-coats have a mixed clay mineralogy and are spatially heterogeneous across the range of marginal-marine depositional environments. The study further showed that clay-coat mineralogy is governed initially by the hydrologically-controlled segregation of the clay minerals within inner estuarine depositional environments, and subsequently by the selective abrasive removal of specific clay mineral types during reworking and transport into the outer estuary and the marine environment. The highest relative abundance of grain-coating chlorite was in sand-flat and tidal-bar depositional environments. The availability of an analogue data set, and an understanding of the controlling processes of clay-coat mineralogy, offer crucial steps in building a predictive capability for clay-coat derived elevated reservoir quality in deeply buried sandstones.

Keywords clay-coat mineralogy, clay minerals, modern analogue, sandstone reservoir quality prediction, sediment heterogeneity.

INTRODUCTION

Clay-coats have long been demonstrated to inhibit the growth of quartz cement in deeply buried sandstones (Heald & Larese, 1974; Ehrenberg, 1993; Bloch et al., 2002; Lander et al., 2008). The principal factor controlling the effectiveness of clay-coats (whether chlorite, illite or mixtures of the two) to
inhibit growth of quartz cement, and thus preserve elevated primary porosity deep in sedimentary basins, is the completeness of grain coat coverage (Bloch et al., 2002; Lander et al., 2008; Ajdukiewicz & Larese, 2012). Clay-coat mineralogy is an important secondary control on quartz cement growth because typically there are morphological differences between coats composed of different clay minerals (Ajdukiewicz & Larese, 2012).

**Terminology and evolution of clay-coat morphology**

The term clay-coat encompasses both detrital (pre-diagenesis) and diagenetic clay-coats (Ajdukiewicz & Larese, 2012; Dowey et al., 2017). Detrital clay-coated sand grains form at, or near, the surface of the sediment in the eodiagenetic realm and are the focus of this study (Worden & Morad, 2003; Ajdukiewicz & Larese, 2012; Dowey et al., 2017; Wooldridge et al., 2017b). It has been concluded that diagenetic clay-coats (those found in many ancient and deeply buried sandstone reservoirs) form via: (i) the thermally-driven recrystallization of attached detrital, precursor, clay-coat material (known as diagenetic-recrystallized coats); and (ii) authigenic in situ precipitation (accretion) through diagenetic mineral interactions (grain dissolution and re-precipitation) with pore fluids. (These are known as diagenetic-authigenic coats – Bloch et al., 2002; Gould et al., 2010; Ajdukiewicz & Larese, 2012; Wooldridge et al., 2017b). Authigenic coats have been interpreted to typically nucleate on existing detrital-clay mineral root-layers (Bloch et al., 2002; Gould et al., 2010; Stricker & Jones, 2016).

In order to help terms reference reference of future studies on clay-coats, a framework and definition of terms for the classification of clay-coats in sands and sandstones is presented in Table 1.

The timing of transformation from detrital clay-coats to diagenetic clay-coats (Fig. 1) remains relatively poorly understood. The reported occurrence of chlorite clay-coats that inhibit quartz cements suggests that transformation (thermally driven neoformation from discontinuous detrital-clay-coats to continuous diagenetic coats) occurs at temperatures lower than 80°C (the typical onset temperature of quartz cement precipitation; Ehrenberg, 1993; Worden & Morad, 2000; Ajdukiewicz & Larese, 2012). From various sources in the literature (Ehrenberg, 1993; Aagaard et al., 2000; Billault et al., 2003; Ajdukiewicz & Larese, 2012), it has been possible to produce a synthesis of the relative timing and morphological changes during clay-coat evolution (Fig. 1). Stage 1: the early attachment of detrital-clay minerals to grain surfaces at or near the sediment surface prior to the onset of compaction. Note that stage 1 is the focus of this research. Stage 2: the transformation from discontinuous detrital-clay-coats to continuous diagenetic grain coats (if sufficient attached detrital-clay material is available for neoformation) prior to the onset of extensive quartz cementation and feldspar dissolution. Stage 3: the precipitation of authigenic clay-coat components via the dissolution of detrital grains and re-precipitation of the dissolved material.

**Quartz cementation in sandstones**

In order to assess the effectiveness of clay mineralogy in promoting the preservation of primary porosity (through the inhibition of quartz cement), it is important to understand the process of quartz growth and the mechanism of clay-coat-derived inhibition. The quantity of quartz cement (Worden & Morad, 2000; Lander et al., 2008) is the main control on porosity, after compaction, in many deeply buried sandstones (Worden & Morad, 2000). Quartz cementation occurs initially as scattered anhedral crystallites at isolated sites across a quartz grain surface (Pittman, 1972; Lander et al., 2008; Ajdukiewicz & Larese, 2012). Homoeptaxial growth of the initial quartz crystallites causes the lateral spreading of quartz cement across the grain surface until they mutually contact and coalesce with one another to lead to a continuous quartz cement layer (Lander et al., 2008; Ajdukiewicz & Larese, 2012). Continued growth of the coalesced quartz produces the characteristic euhedral termination associated with authigenic quartz cements (Lander et al., 2008).

Clay-coats have been reported to inhibit quartz cementation via a two-step process: (i) at moderate temperatures (ca 115°C), clay-coats (if complete) physically isolate quartz grain surfaces from silica saturated pore fluids, thus retarding initial quartz cement nucleation (here defined as nucleation inhibition); and (ii) at high temperatures (115 to >160°C), isolated nanocrystals of quartz cement begin to nucleate on quartz grain surfaces between the clay mineral particles in the coat (here defined as growth inhibition; Lander et al., 2008; Ajdukiewicz & Larese, 2012). At these high temperatures, clay particles in the coat act as barriers (discontinuities) which prevent epitaxial quartz growth and subsequent coalescence and growth of nanoquartz crystals.
The result is a vastly reduced quartz cement growth rate (Billault et al., 2003; Ajdukiewicz & Larese, 2012). Clay-coats start to lose their ability to inhibit quartz cement at elevated temperatures (>160°C), or when there are mechanically-induced breaks in the clay-coat coverage (for example, during a fracturing event; Lander et al., 2008; Ajdukiewicz & Larese, 2012).

The ability of the attached clay-coats to inhibit quartz cement growth in deeply buried sandstones is a function of clay-coat morphology and completeness, which, in turn, are controlled by clay mineralogy (Ajdukiewicz & Larese, 2012). It is apparent that the morphology of the coat directly controls quartz cement inhibition. Chlorite, which tends to occur as platy to curly crystals, has been reported to better inhibit quartz cement than illite, which typically occurs as fibrous hair-like crystals (Worden & Morad, 2003; Ajdukiewicz & Larese, 2012).

Elevated porosity, clay-coats in deeply buried sandstones, has commonly been associated with chlorite grain-coats (Dowey et al., 2012); however, illite and mixed mineralogy coats (for Table 1. Definitions of clay-coat terminology, modified from Dowey et al. (2017)

| Terminology | Suggested definitions | Distinguishing morphology |
|-------------|-----------------------|--------------------------|
| Surface-Shallow burial | A clay mineral-dominated coat on a sand grain that formed before, during or immediately after deposition via the physical attachment of clay minerals (typically a divergent mixture of phyllosilicates); i.e. low temperature and while still in the depositional environment | Discontinuous, micron scale, accumulations of clay to silt-sized material (clay particles and organics) typically occurring as: clumps, tangentially attached coats, or bridging strands of clay material between grains. Creates a diverse mixture of phyllosilicates |
| Diagenetic-clay coats | A clay mineral-dominated coat (where any clay mineral could be present) on a sand grain that formed during burial and diagenesis (typically >2000 m and/or >70°C) | Tangentially orientated (parallel to the grain surface), densely packed mass of platey-anhedral mineralogically heterogeneous clay minerals |
| Diagenetic-recrystallized clay coat | Depositional controlled (i.e. recrystallization of the primary attached clay mineral assemblage, emphasizing the importance of detrital-clay coats formed in depositional environments) | A detrital clay mineral-dominated coat that formed before, during or immediately after deposition, that has been mineralogically and texturally modified via thermally-driven (burial) recrystallization when the sediment entered a different geochemical and thermal regime |
| Diagenetic-authigenic clay coat | Provenance controlled (i.e. mineralogy of the sediment assemblage controlling the availability and mineralogy of the dissolved material necessary for authigenic coat formation) | In situ authigenic growth (somewhat deeper burial), through detrital mineral interactions with pore fluids and direct re-precipitation of the dissolved material; most commonly reported to occur on an existing recrystallized detrital root structure (nucleation site) |

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example, illite–chlorite–smectite have also been reported (Storvoll et al., 2002; Martinius et al., 2005; Stricker & Jones, 2016). Mixed mineralogy coats occur in sandstones deposited in marginal marine settings and have significantly enhanced reservoir quality in several notable sandstone reservoirs from the Norwegian Continental Shelf (Storvoll et al., 2002; Martinius et al., 2005), the Ordos Basin, China (Luo et al., 2009) and the Ghawar Field, Saudi Arabia (Al-Ramadan et al., 2004).

Modern detrital-clay-coats have been shown to consist of a diverse mixture of phyllosilicates (chlorite, illite, kaolinite, berthierine, smectite and odinite) that are bound to framework sand grain surfaces (Ehrenberg, 1993; Daneshvar, 2011; Dowey et al., 2017; Wooldridge et al., 2017b). Detrital-clay-coats are composed of aggregates of clay minerals, silt to clay-sized lithics and bioclasts (for example, diatoms) which result in discontinuous accumulations attached to sand grain surfaces (Dowey et al., 2017; Wooldridge et al., 2017a,b).

The initial mineralogy of the detrital-clay-coat is the principal control on the mineralogy of the final diagenetic coats (Worden & Morad, 2003) which thus influences the potential effectiveness of the inhibition of subsequent quartz cement. There is a growing understanding of the distribution of clay-coats in modern systems (Dowey, 2013; Dowey et al., 2017; Wooldridge et al., 2017a,b). However, there is relatively limited knowledge about the specific spatial distribution of clay-coat mineralogy in marginal-marine sediments. This represents a shortfall in our ability to develop a credible model for the prediction of reservoir quality as a function of clay-coat presence and mineralogy.

The goal of this study was therefore to document clay-coat mineralogy across a marginal-marine system using an automated scanning
electron microscope – energy dispersive spectrometry (SEM–EDS) methodology. This approach has involved the development of a novel ability to differentiate the clay-coat mineralogy from that of the total sediment clay mineralogy. Focused on the Ravenglass Estuary, UK, this study addresses the following questions, (Fig. 2):

1. How variable is clay-coat mineralogy in a modern marginal-marine system?
2. What controls the distribution of clay-coat mineralogy across a marginal-marine system?
3. Are patterns in clay-coat mineralogy comparable to other modern or ancient and deeply buried marginal-marine sedimentary systems?
4. Can clay-coat mineralogy be predicted in modern and ancient deeply buried marginal marine sandstones?

**MATIERALS AND METHODS**

**Study site**

The aim of the study was to establish the origin and distribution of clay-coat mineralogy in ancient and deeply buried sandstones by studying, at high resolution, a modern sedimentary analogue. The Ravenglass Estuary is a 5-6 km², macro-tidal (tidal range <7-55 m) system located
in Cumbria, UK, composed of three rivers, the Esk, the Mite and the Irt, that merge in a central estuary basin: (Wooldridge et al., 2017b). Previous studies of the estuary defined the depositional environment and sedimentary framework which is utilised in this study (Fig. 2B; Wooldridge et al., 2017b; Griffiths et al., 2018a,b). The inner estuarine depositional environments of tidal-flats (mud, mixed and sand) and tidal-bars (vegetated and non-vegetated) are consistent with a back-barrier, tidal-dominated hydrodynamic regime (Wooldridge et al., 2017b). The outer estuary (the area seaward of the dune-topped barrier spits) is characterized by wave-dominated foreshore and pro-ebb delta depositional environments (Fig. 2B).

The Ravenglass Estuary has an overall clay mineral assemblage composed of illite, chlorite and kaolinite, reported to have been derived principally from a suspended fluvial sediment source (Daneshvar, 2011; Daneshvar & Worden, 2018). Previous studies of the estuary have identified a heterogeneous distribution of clay-coated sand grains (Wooldridge et al., 2017b; Fig. 2C) across the full range of marginal-marine depositional environments.

Many ancient and deeply buried, chlorite, illite and mixed mineralogy (chlorite-illite) clay-coated sandstones were initially deposited in estuarine environments so that studying modern estuaries may unlock the ability to predict clay-coat distribution in reservoir sandstones (Ehrenberg, 1993; Dowey et al., 2012; Skarpeid et al., 2017). The study area thus offered the ability to: (i) explore the spatial distribution of clay-coat mineralogy; and (ii) elucidate the controlling mechanisms to aid prediction and understanding of clay-coat derived reservoir quality in ancient and deeply buried sandstones.

The fluvially-derived clay mineral assemblage at Ravenglass originates principally from the weathering and incision of the hinterland Palaeozoic Eskdale Granite, Triassic Sherwood Sandstone, Borrowdale Volcanic Group geology, and outcropping glacial units (Moseley, 1978). The chloritized Eskdale Granite (Moseley, 1978) and chloritized pyroxenes in the Borrowdale Volcanic Group have been proposed by Griffiths (2018) and Griffiths et al. (2018a,b) to be the source of detrital chlorite. Mica-bearing rocks in the hinterland (Eskdale Granite and Borrowdale Volcanic Series) provide the source for illite, and kaolinite was derived as an alteration product of feldspar minerals or other aluminous silicate minerals.

Sample suite and methodology
The study focused on a suite of 38 surface sediment samples (i.e. collected from the top <2 cm of sediment) from which polished thin sections were constructed as grain mounts. Samples encompassed the range of intertidal depositional environments to produce a complete fluvial to marine-transect of clay-coat mineralogy. Spatial distribution maps were plotted using the interpolation function in ArcGIS (https://www.arcgis.com). Clay mineralogy is here reported as index maps [for example, illite/(illite + chlorite + kaolinite)] to define any clay mineral distribution trends or spatial patterns.

Quantification of clay-coat mineralogy was undertaken via scanning electron microscope–energy dispersive spectrometry (SEM–EDS), using a FEI WellSite QEMSCAN® system (FEI Company, Hillsboro, OR, USA), operating at 15 kV, with two Bruker EDS detectors (Bruker Corporation, Billerica, MA, USA; Armitage et al., 2010; Wooldridge et al., 2017b). This methodology permitted the in situ mineralogical imaging and quantification of the sediment (for example, framework grains and clay mineral assemblage) to a spatial resolution of ca 1 μm. The SEM–EDS QEMSCAN® system digitizes the thin sectioned sediment by undertaking chemical mapping via two fast energy dispersive spectrometers (EDS detectors). Each analysed point (with an area of 1 μm²) was compared to a library of spectra and assigned to a specific mineral (Armitage et al., 2010). Mineral mapping also produces a backscatter electron image and quantitative digital maps of clay mineralogy (image-area percentage and imaged area mass).

The granulate function in the QEMSCAN® system provides the ability to digitally sieve component sediment minerals (for example, chlorite and illite) based on particle size (long axis; Fig. 3) post-measurement. The QEMSCAN® technique thus enables the digital-disaggregation of all of the clay mineral components of a sample based on particle size (Fig. 3C).

Previous clay-coat analysis of the Ravenglass Estuary defined the following typical maximum dimension of monomineralic clay mineral accumulations that are present in clay-coats (illite <32 μm, chlorite <16 μm and kaolinite <16 μm) that are present in clay-coats (Wooldridge et al., 2017b).

A fundamental assumption behind the methodology is that any monomineralic clay particle, or monomineralic clay aggregate, that is
in excess of the user-defined clay-coat maximum size, is present as either: (i) clay-rich lithic fragments; (ii) large clay floccules; or (iii) clay-rich intraclasts in the sediment, and not as components of clay-coats (Worden & Morad, 2003). The ability to digitally size-sieve minerals and separate out large clay-rich grains from clay-coats thus enables quantification of: (i) the volume (image-area percentage) of clay minerals in a sample that occur as clay-coats and (ii) the clay mineralogy of grain coats.

Note that previous studies of clay-coat mineralogy used X-ray diffraction analysis (XRD) of carefully separated fine and coarse sediment fractions (Dowey, 2013; Dowey et al., 2017; Wooldridge et al., 2017b). The alternative methodology adopted here provides the novel ability to define the clay-coat mineralogy and differentiate it from
the clay mineralogy of the bulk sediment during one analysis step followed by computational size-sieving. The SEM–EDS approaches have their limitations, explicitly their inability to analyse items smaller (using QEMSCAN®) than about 1 or 2 μm. However, there is no technique, or combination of techniques, other than SEM–EDS, that can uniquely and repeatedly define clay mineralogy, quantify clay mineral proportions and image the location of the clay mineral in the sediment (or in sedimentary rocks).

RESULTS

Distribution of clay minerals in marginal-marine sediments

The whole sediment clay mineral abundance map (Fig. 4A) shows an overall pattern of increasing clay mineral content with increasing distance from the open ocean and so towards the upper tidal limit. Sediment clay mineral percentages are summarized in Table 2. Note that there is no visible clay matrix in any of the imaged samples (Fig. 4C to F).

Samples which contain a clay mineral content >4% are found predominantly in inner estuarine environments (compare Fig. 4A to Fig. 2B). Clay minerals are heterogeneously distributed in the inner estuary, ranging from ca 18% to 2%, with a higher average clay mineral percentage in samples from the northern Irt arm of the estuary than the southerly Esk arm of the estuary (compare Fig. 4A to Fig. 1B). Inner estuarine samples that contain <4% clay minerals occur principally in deposits from non-vegetated tidal-bars and sand-flats (compare Fig. 2B to Fig. 4A). Mud-flat samples contain the highest average sediment clay mineral abundance (12-7%). The outer estuarine depositional environments (foreshore and pro-ebb delta) have a fairly uniform distribution of total clay minerals, ranging from 4-4% to 2-2% (Table 2).

The internal distribution and textural characteristics of clay minerals in estuarine sediment are documented in Fig. 4C to F. Clay minerals in the marginal marine sediments studied here occur predominantly as: (i) coats on and between framework grains; (ii) clay-rich lithic clasts (allochthonous clay-rich sand grains); (iii) clasts (flocules) composed predominantly of clay minerals; (iv) mud intraclasts (autochthonous clay-rich sand grains) composed of aggregated clay and lithics; and (v) in association with feldspars. Clay-rich rock fragments enter the systems (Fig. 4D) in the fluvial arms. Inner estuary sediment contains clay mineral linkages (bridging structures; Wooldridge et al., 2017b) between grains (Fig. 4C) whereas the outer estuary sediment contains no clay mineral linkages (Fig. 4F).

The fraction of clay mineral abundance present as clay-coats and its distribution

In order to avoid confusing the use of percentages of total clay and percentages of clay that occur as coats, the latter is here expressed as a fraction of 100 to emphasize that this unique feature has been quantified. The amount of the total clay mineral abundance that is present as clay-coats varied from 93/100 to 61/100 across the estuary, with pronounced heterogeneity (Fig. 4B, Table 2). Figure 4B documents the internal distribution of clay minerals within marginal marine sediments. For example, regions with a high fraction of the total clay mineral abundance that is present in clay-rich lithics or as clay floccules, have a reduced fraction of clay mineral abundance present as clay-coats (Fig. 4).

The spatial distribution reveals that, on average, 81/100 of the total estuarine sediment clay mineral content is present as clay-coats, rising to 89/100 and 90/100 in the deposits consisting of mud flat and tidal inlet depositional environments, respectively (Table 2). Tidal bars (non-vegetated) have the lowest fraction of clay mineral abundance present as clay-coats (61/100). The tidal bars located in the lower Esk arm of the estuary (Fig. 2B) represent a depositional environment in close spatial correspondence to a surface armoured by pebbles (compare Fig. 2B to Fig. 4B). The sediment assemblage here consists of an elevated proportion of clay-rich lithic fragments (Fig. 4D).

The spatial distribution of whole sediment clay mineralogy

The clay mineral assemblage of the Ravenglass Estuary is dominated by illite which contributes a minimum of 0-47 of the clay material across the estuary, with chlorite (0-01 to 0-48) and kaolinite (0-01 to 0-22) contributing smaller proportions. Whole sediment clay mineralogy is distinctly heterogeneous across the marginal-marine heterogeneous across the marginal-marine sediments of the Ravenglass Estuary (Fig. 5).

Clay mineralogy is here reported as index values [for example, illite/(illite + chlorite]...
Fig. 4. Maps of total clay mineral percentage and the fraction of the total clay minerals present as clay-coats. (A) Map of total clay mineral percentage. Filled circles on map (A) indicate sample locations. (B) Map of clay mineral content present as clay-coats. (C) to (F) SEM–EDS images of sediment bulk mineralogy, showing the progression in the abundance and distribution styles (for example, coats or floccules) of clay minerals.
Table 2. Summary information for the scanning electron microscope-energy dispersive spectrometry (SEM-EDS) derived: (i) sediment clay mineral percentages; (ii) total clay minerals present as clay-coats; (iii) total sediment clay mineralogy percentages; and (iv) total clay-coat mineralogy percentages.

| Clay minerals in sediment % | Fraction of clay minerals as clay-coats | SEM-EDS whole sediment clay mineralogy (image area) % | SEM-EDS clay-coating, clay mineralogy (image area) % |
|-----------------------------|-----------------------------------------|------------------------------------------------------|------------------------------------------------------|
|                             | Total sediment-illite | Total sediment-kaolinite | Total sediment-chlorite | Clay coating-illite | Clay coating-kaolinite | Clay coating-chlorite |
| PRO-EBB DELTA               |                          |                         |                          |                        |                        |                        |
| Average                     | 2.5                      | 76/100                  | 1.34                     | 0.22                   | 0.81                   | 1.29                    | 0.19                    | 0.26                    |
| Maximum                     | 3.0                      | 83/100                  | 1.38                     | 0.25                   | 1.27                   | 1.38                    | 0.24                    | 0.30                    |
| Minimum                     | 2.2                      | 66/100                  | 1.28                     | 0.20                   | 0.56                   | 1.19                    | 0.17                    | 0.23                    |

| FORESHORE                   |                          |                         |                          |                        |                        |                        |
| Average                     | 3.0                      | 79/100                  | 1.47                     | 0.42                   | 1.00                   | 1.45                    | 0.39                    | 0.37                    |
| Maximum                     | 4.4                      | 89/100                  | 1.78                     | 0.51                   | 2.17                   | 1.75                    | 0.50                    | 0.59                    |
| Minimum                     | 2.2                      | 62/100                  | 1.17                     | 0.28                   | 0.44                   | 1.14                    | 0.28                    | 0.26                    |

| TIDAL-BAR VEGETATED         |                          |                         |                          |                        |                        |                        |
| Average                     | 6.2                      | 82/100                  | 4.46                     | 0.33                   | 0.87                   | 3.94                    | 0.31                    | 0.39                    |
| Maximum                     | 6.6                      | 89/100                  | 4.49                     | 0.51                   | 0.89                   | 4.32                    | 0.48                    | 0.42                    |
| Minimum                     | 5.9                      | 75/100                  | 4.42                     | 0.15                   | 0.85                   | 3.55                    | 0.14                    | 0.36                    |

| NON-VEGETATED TIDAL BAR     |                          |                         |                          |                        |                        |                        |
| Average                     | 3.9                      | 75/100                  | 2.02                     | 0.39                   | 1.19                   | 1.96                    | 0.25                    | 0.44                    |
| Maximum                     | 8.0                      | 89/100                  | 3.96                     | 0.74                   | 2.93                   | 3.93                    | 0.64                    | 0.97                    |
| Minimum                     | 2.4                      | 61/100                  | 1.05                     | 0.08                   | 0.41                   | 1.04                    | 0.07                    | 0.20                    |

| MUD-FLAT                    |                          |                         |                          |                        |                        |                        |
| Average                     | 12.7                     | 89/100                  | 9.33                     | 1.25                   | 1.68                   | 8.25                    | 1.02                    | 1.23                    |
| Maximum                     | 17.8                     | 93/100                  | 13.20                    | 1.68                   | 2.63                   | 11.94                   | 1.48                    | 2.26                    |
| Minimum                     | 8.7                      | 85/100                  | 4.80                     | 0.81                   | 1.27                   | 4.78                    | 0.67                    | 0.64                    |

| MIXED-FLAT                  |                          |                         |                          |                        |                        |                        |
| Average                     | 8.4                      | 84/100                  | 5.52                     | 0.59                   | 1.62                   | 5.29                    | 0.53                    | 0.61                    |
| Maximum                     | 13.7                     | 92/100                  | 10.24                    | 1.22                   | 4.56                   | 9.56                    | 1.12                    | 1.24                    |
| Minimum                     | 3.8                      | 63/100                  | 2.51                     | 0.23                   | 0.71                   | 2.44                    | 0.21                    | 0.37                    |

| SAND-FLAT                   |                          |                         |                          |                        |                        |                        |
| Average                     | 3.2                      | 78/100                  | 1.82                     | 0.36                   | 0.98                   | 1.69                    | 0.30                    | 0.38                    |
| Maximum                     | 4.7                      | 89/100                  | 2.58                     | 0.82                   | 1.54                   | 2.53                    | 0.82                    | 0.49                    |
| Minimum                     | 2.0                      | 61/100                  | 0.92                     | 0.06                   | 0.43                   | 0.87                    | 0.05                    | 0.25                    |

| TIDAL INLET                 |                          |                         |                          |                        |                        |                        |
| Average                     | 3.4                      | 90/100                  | 2.15                     | 0.26                   | 0.60                   | 2.10                    | 0.20                    | 0.37                    |
| Maximum                     | 3.5                      | 92/100                  | 2.21                     | 0.28                   | 0.69                   | 2.14                    | 0.20                    | 0.50                    |
| Minimum                     | 3.3                      | 87/100                  | 2.08                     | 0.23                   | 0.51                   | 2.05                    | 0.2                    | 0.24                    |

Kaolinite clay mineral content of the whole sediment has two areas of elevated relative

+ kaolinite]. Illite is the principal component of the clay mineral assemblage (illite index ranges from 0.47 to 0.87) with the highest relative abundance present in the mud-flat and mixed-tidal-flat sediments of the Irt and Mite arms of the estuary, and the southern foreshore (compare Fig. 5A to Fig. 2B). Inner estuarine whole sediment has a higher illite proportion than outer estuarine depositional environments (for example, foreshore and pro-ebb delta) although local variability occurs.

The highest whole sediment chlorite content occurs in the inner-central and Esk arm of the estuary, corresponding to tidal-bar and tidal-flat (mixed and sand) depositional environments (Fig. 5B). Elevated relative chlorite abundance occurs in the tidal inlet and northern foreshore sediments. Chlorite is relatively depleted in the sediments from the tidal-flats (mud and mixed) of the Irt and Mite arms of the estuary.

Kaolinite clay mineral content of the whole sediment has two areas of elevated relative

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kaolinite concentration that are restricted to either the inner-central estuarine tidal-flats and tidal-bars, or the outer foreshore and pro-ebb delta depositional environments. Sediments that contain high proportions of kaolinite (i.e. >0-12) are restricted to the tidal-bars and sand-flats of the River Esk arm of the Estuary (compare Fig. 2B to Fig. 5C); represented in Fig. 4D and E.

Focusing on the tidal flat environment (Fig. 5) of the inner-central estuary reveals the local variability in clay mineralogy across mud-flats, mixed-flats and sand-flats, with the greatest proportion of chlorite in the sand-flats and the greatest proportion of illite and kaolinite in the mud-flats and mixed-flats. Figure 6 illustrates patterns of clay-coat mineralogy and the internal distribution of the clay.

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Fig. 6. Photographs, clay-coat mineralogy index maps, and scanning electron microscope–energy dispersive spectrometry (SEM–EDS) images of tidal-flat sediments. (A) to (D) Photographs illustrating the surface sediment features and environments (the quadrat is 1 m²). Arrows on (E) to (G) indicate the direction of images in (A), (B) and (D). (E) to (G) Clay mineral index maps of relative clay-coat mineralogy. For location of the Saltcoats tidal-flat see Fig. 2B. (H) to (K) SEM–EDS images of bulk sediment mineralogy. (L) to (O) SEM–EDS images of clay mineralogy with other minerals shaded collectively as grey. (P) to (S) enlarged SEM–EDS images of clay mineralogy. Purple rectangles on parts (L) to (O) indicate the location of images in the larger sample. Arrows linking (E) to (H) and (I), (F) to (J) and (G) to (K) indicate the relative position of the imaged samples. (H), (L) and (P) Mud-flat. (I), (M) and (Q) Mixed-flat upper. (J), (N) and (R) Mixed-flat lower. (K), (O) and (S) Sand-flat.
mineral assemblage across a tidal flat sedimentary environment. The highest relative concentrations of illite at Ravenglass occur in low energy settings (for example, mud flats, Fig. 6E). Chlorite abundance is greatest from Ravenglass in the high energy environments (for example sand flats, Fig. 6G).

The spatial distribution of clay-coat mineralogy

Clay-coat mineralogy is heterogeneously distributed across the marginal-marine sediments at Ravenglass. Clay coats are dominated by illite, which contributes a minimum 0.50 of the attached clay coat material across the estuary, with less chlorite (0.05 to 0.25) and kaolinite (0.04 to 0.25; Fig. 7). There is an increasingly mixed mineralogy (chlorite and kaolinite) at inner estuarine sites in the higher energy settings, representing sand-flat and tidal-bar (non-vegetated) depositional environments (Fig. 7). Clay-coats in outer estuary settings have a low chlorite content and are relatively enriched in illite and kaolinite (Fig. 7).

The illite component occurs typically as irregular accumulations of crystals forming both clay mineral accumulations on grain surfaces and bridging structures between grains (Fig. 6). Chlorite principally has a ‘flake like’ morphology occurring as prominent projections away from the grain. Kaolinite is present both as larger isolated clasts and in irregular accumulations associated with illite (Fig. 6H to S).

Clay-coats have an elevated illite percentage in the tidal-flats (mixed and mud) and tidal-bar depositional environments from the rivers Irt and Mite compared to the river Esk arm of the estuary (compare Fig. 2B to Fig. 7A). Focusing on the tidal flat environment reveals the local variability in clay-coat mineralogy across mud-flats, mixed-flats and sand-flats with most illite in the mud-flats and least illite in the sand-flats (Figs 7A and 6E).

Clay-coats have decreasing chlorite content from the inner-central to the outer estuary (pro-ebb delta; 0.25 to 0.01; Fig. 7B). Clay-coats with the highest chlorite content occur in the inner-central zone and Esk arm of the estuary, corresponding to tidal-bar and tidal-flat (mixed and sand) depositional environments (compare Fig. 2B to Fig. 7B). There are two areas of elevated concentration of kaolinite in clay-coats with clay-coats containing >0.16 kaolinite restricted to either the inner-central estuarine tidal-flats, tidal-bars, or the outer foreshore and pro-ebb delta depositional environments (Fig. 7C).

Correlation between whole sediment and clay-coat mineralogy

The difference between clay minerals found in bulk sediment and clay-coats is illustrated in Fig. 8. The close correlation between total sediment and clay-coats illite abundance in marginal marine sediments (compare Fig. 5A to Fig. 7A) suggests that illite principally occurs as clay-coats (Fig. 8), especially in outer estuarine sediments (for example, foreshore and pro-ebb delta). In mud-flat sediments, an additional ca 10% illite is present not as clay-coats but as either clumps, that potentially resulted from flocculation (Hill et al., 2013), or clay rich lithics (Figs 4C, 6H, 6L, 6P and 8).

In outer estuarine environments, kaolinite is principally present as components of clay-coats especially in foreshore (ca 92%) and pro-ebb delta (ca 86%) sediments (Fig. 8B). Chlorite in tidal flat sediments is present, typically, as clumps that potentially resulted from flocculation (Fig. 6L), in clay-rich lithics (Fig. 6N) and as components of clay-coats (Fig. 6O and S) with approximately half (50%) of the total chlorite present as clay-coats. Tidal bar, foreshore and pro-ebb delta deposits are unusual in having between 63% and 68% of their chlorite present as clay-rich lithics rather than clay-coats (Table 2, Fig. 4D and F).

Sedimentary environments and clay-coat mineralogy

Clay-coat mineralogy of the Ravenglass sediments has here been differentiated by depositional environment (Fig. 9A) to allow comparison between modern and ancient marginal marine sediments. Clay-coats with the highest relative chlorite content occur in sediments from sand-flat and non-vegetated tidal-bar depositional environments (Fig. 9A). The lowest relative chlorite content (typically <15%) is present in clay-coats in vegetated tidal-bar sediments (Table 2). Clay-coats with the highest relative illite content occur in samples from vegetated tidal-bars, tidal inlet tidal-bars, and mud and mixed tidal-flats. The highest relative contribution of grain coating-kaolinite occurs in sand-flat and foreshore depositional environments (Fig. 9A).

There is no simple graphical relationship between clay-coat mineralogy and clay-coat
coverage (Fig. 9B). There is a minor relative increase in chlorite with increasing grain size (which is a proxy for energy regime; Fig. 9C); this is broadly consistent with spatial distribution patterns of clay-coat mineralogy (Fig. 7) and grain size (Fig. 2D). Overall, lower energy settings tend to have higher degrees of clay-coat coverage and also have a greater quantity of illite (Figs 2C and 7).

**DISCUSSION**

**Origin of sediment clay minerals**

This study has revealed that the majority of the sediment clay mineral volume, in excess of 61/100 (the lowest recorded proportion), is present as clay-coats in the Ravenglass sediment.
(Table 2). This fraction is highest in the sand-dominated assemblages of the mixed tidal flat (84/100), sand tidal flats (78/100), tidal bars (non-vegetated; 75/100), tidal inlet (90/100), mud flats (89/100) and foreshore (79/100) depositional environments (Table 2).

The origin of the additional clay minerals (i.e. those not present as clay-coats) occur principally as: (i) floccules of clay (salinity-mediated and biologically-mediated; Fig. 6) in sediment from reduced energy conditions (for example, mud-flat, Fig. 10A); (ii) clay-rich rock fragments (Fig. 10F); and (iii) in association with feldspars (Fig. 10B), which can develop via the rapid early diagenetic alteration to illite and kaolinite clay minerals (Daneshvar & Worden, 2018). The predominant role of grain-coating clay in the overall clay mineral budget in the sediments studied here highlights the fundamental role of clay-coat forming processes (Wilson, 1992; Worden et al., 2006; Wooldridge et al., 2017b).

**Origin of clay minerals in clay-coats**

The mineralogy of estuarine clay minerals is mainly governed by the geology of the hinterland, climate, relief and the length of the fluvial transport system (Worden & Morad, 2003; Wooldridge et al., 2017b; Daneshvar & Worden, 2018; Worden et al., 2018). The similar mineralogy and spatial patterns of the total sediment clay mineral (Fig. 5) and clay-coat mineralogy (Fig. 7) for inner-estuarine sediments implies that coat mineralogy is principally a function of the fluvial inputted clay mineral assemblage.

A comparison between whole sediment clay mineralogy and clay-coat mineralogy reveals that the spatial patterns are broadly similar for both illite (Figs 5A and 7A and chlorite (Figs 5B and 7B), especially for inner estuarine sediments. Discrepancies between the whole sediment clay mineral assemblage and clay-coat mineralogy (for example, kaolinite content in the River Esk; Fig. 5C to Fig. 7C), result from the inclusion of clay minerals present in clay-rich lithics (Fig. 4D) in the calculations of total sediment clay mineralogy. The consistent relationship between whole sediment clay mineralogy and clay-coat mineralogy for illite and chlorite in the Ravenglass Estuary suggests that clay-coats were formed in the inner estuary via the attachment of the hydrodynamically-fractionated clay mineral assemblage (thus explaining the cause of the similarity of the whole sediment and clay-coat mineral distribution patterns in Figs 5 and 7). The correlation between whole sediment and clay-coat mineralogy suggests that clay-coat mineralogy is principally governed by the processes that control the depositional fractionation of clay minerals supplied to the estuary (see later).

The local differences between the whole sediment clay mineral abundance (Fig. 5) and the proportion of clay-coating minerals (Fig. 8), suggests that techniques which quantify the total sediment (for example, XRD) may potentially over-predict the abundance and mineralogy of clay-coats (for example, chlorite, Fig. 8C). In summary, it is important to examine clay-coat mineralogy as...
Fig. 9. Plots comparing total clay-coat mineralogy to depositional environment, clay-coat coverage and grain size. (A) Plot of normalized relative clay-coat mineralogy, segregated by depositional environments. (B) Cross-plot of indexed clay-coat mineralogy and clay-coat coverage. (C) Cross-plot of clay-coat mineralogy and mean sediment grain size.
Fig. 10. Schematic model showing relative trends in clay-coat mineralogy and clay-coat coverage across a typical marginal marine transect of depositional environments, with the distribution of sedimentary environments modified from Dalrymple et al. (1992). (A) Mud-flat. (B) Mixed-flat. (C) Sand-flat. (D) Distal (from ocean or vegetated) tidal-bar. (E) Proximal (to ocean) tidal-bar. (F) Pro-ebb delta. Yellow arrows on (A) to (F) indicate the location of attached clay-coats.
well as whole sediment composition. This approach requires high resolution SEM–EDS analysis with a minimum step size of 2 μm.

**Origin of inner estuarine clay-coat mineralogy heterogeneity**

Previous work on the Ravenglass Estuary (Wooldridge et al., 2017b), concluded that clay-coat formation (i.e. clay mineral attachment to grain surfaces) occurred preferentially in an inner estuary ‘clay-coat factory’, therefore clay-coat mineralogy must be derived from inner-estuarine clay mineral fractionation and depositional processes. Griffiths (2018) and Griffiths et al. (2018a,b) reviewed the potential processes that govern the fractionation of the fluvially-derived clay minerals in estuarine systems and concluded that the heterogeneity of illite and chlorite (Fig. 6) partly derives from differential settling rates, partly governed by particle sizes (Whitehouse et al., 1960).

In the Ravenglass Estuary, chlorite typically sits in coarser sediment than illite (compare Fig. 5 to Fig. 2D), and therefore must have dropped out of suspension close to the main ebb channel. Differential settling rates could explain the systematic variations of chlorite and illite abundance across the tidal flats sequences (Figs 5 and 6) with chlorite typically present as larger ‘flakes’ (compared to illite) in clay-coats (Fig. 6).

The mineralogy of inner estuarine clay-coats is thus governed, first by the fluvial inputted clay mineral assemblage (heterogeneous between the fluvial arms), and second by the differential settling rates of the fluvial derived clay mineral assemblage. The ability to define whether the identified patterns in clay-coat mineralogy are generally applicable is hampered by the palatable lack of comparable studies.

**Origin of outer estuarine patterns of clay-coat mineralogy**

In natural systems, multiple processes occur simultaneously, including the formation of clay-coated sand grains (limited to inner estuary depositional environments) along with continued erosion, transport and deposition of clay-coated sand grains (Wooldridge et al., 2017a,b). Evidence for the transport of clay-coated sand grains in the Ravenglass Estuary from the inner to outer estuary include: (i) the presence of partial clay-coats in samples from the pro-ebb delta; (ii) the systematic decrease in the extent of clay-coat coverage from inner to outer estuarine sediments (Fig. 2C); and (iii) the fact that clay-coats from sand-flats, tidal-bars, tidal-inlet and the outer estuarine sediments (Fig. 4C to F) display textural characteristics consistent with abrasive transport (i.e. grain to grain collision; Wilson, 1992). Clay-coats from foreshore and pro-ebb delta samples have remaining coats within grain indentations, a lack of clay-coat projections, and elevated clay-coat grain coverage on the finer grain size sediment components (Fig. 10F). Abrasive transport (remobilization of the sediment) would remove grain projections but preserve the greatest clay-coat thickness in grain indentations; coats would be preferentially retained in the finer grain size sediment due to a reduced abrasive effect (Wilson, 1992).

Abrasive transport of inner-estuary, clay-coated grains may explain the evolution in clay-coat mineralogy from the inner to outer estuarine sediments. The larger ‘flakes’ of chlorite in clay-coats (Fig. 6) typically form prominent perpendicular projections which, upon grain to grain collision during transport, would experience preferential early detachment compared to more tangential and finer-grained illite and kaolinite. The overall consequence of preferential early detachment during transport out of the inner estuary, is that chlorite coat constituents will be removed (note the decrease in chlorite from inner to outer estuarine depositional environments; Fig. 7B) resulting in the increased relative illite content of the remaining attached coat material (Fig. 7A).

**Comparison of clay-coat mineralogy to the Anllóns Estuary, Spain**

There are few high resolution published studies of clay mineralogy in estuaries (Algan et al., 1994; Brockamp & Clauer, 2012; Dowey, 2013; Dowey et al., 2017). The most detailed study was by Dowey et al. (2017) of the Anllóns Estuary, Spain, using XRD techniques on clay fraction separates, although details of clay-coat mineralogy were not reported. The Anllóns study has a clay mineral assemblage dominated by kaolinite, illite and chlorite (Dowey et al., 2017). Illite is dominant in the inner estuary, kaolinite content increases towards the marine end of the system. Chlorite concentration is highest in sediment from the central estuary, close to the main ebb channel, in sand-flat depositional environments. Kaolinite concentrations...
are lowest in the open marine-influenced shoreface and sand-flat areas of the estuary (Dowey et al., 2017).

The elevated chlorite clay-coat content in inner estuarine sand-flat and tidal-bar depositional settings, reported here for the Ravenglass Estuary, are similar to the total sediment chlorite patterns reported for the Anllóns Estuary (Dowey et al., 2017). The relative increase in the kaolinite clay-coat content in outer estuarine sediments, reported here for the Ravenglass Estuary, is not consistent with the clay mineral distribution pattern of the Anllóns Estuary (Dowey et al., 2017).

A significant outcome from this comparison is that spatial patterns in clay-coat mineralogy from Ravenglass are broadly consistent (i.e. illite and chlorite) with those reported for the bulk clay mineral assemblage of the Anllóns Estuary (Dowey et al., 2017). Differences between the bulk sediment clay fraction and clay-coat mineralogy (i.e. kaolinite content), potentially result from the different analytical methodologies adopted; SEM-EDS analysis and computational (digital) size sieving data from Ravenglass are unlikely to provide exactly comparable data to physical separation and XRD analysis.

RESERVOIR QUALITY IMPLICATIONS AND PREDICTION OF CLAY-COAT MINERALOGY

Mixed mineralogy clay-coats have been reported in sandstones that were deposited in marginal-marine environments (Storvoll et al., 2002; Martinius et al., 2005). Most clay-coats seem to contain chlorite and a mixture of other clay minerals.

Detrital-clay-coated sand grains form via the attachment of the locally available clay minerals, mainly in the inner-estuarine tidal-flat and tidal-bar depositional environments (Wooldridge et al., 2017b). It has been suggested that clay-coats (i.e. detrital-clay-coats) represent precursors to those found in some deeply buried sandstone reservoirs (Bloch et al., 2002; Ajdukiewicz & Larse, 2012; Wooldridge et al., 2017b; Fig. 1). The precursor detrital-coat material exerts a fundamental control on the diagenetic evolution and final mineralogy of clay-coats in deeply buried sandstones (diagenetic clay-coat; Worden & Morad, 2003). In turn, diagenetic clay-coats directly influence the effectiveness of quartz cement inhibition (Ajdukiewicz & Larse, 2012). If, as reported (Aagaard et al., 2000; Bloch et al., 2002; Worden & Morad, 2003; Ajdukiewicz & Larse, 2012), diagenetic clay-coats form via the recrystallization of a precursor detrital-clay-coat (Table 1), then an analogue data set of detrital-clay-coat mineralogy will aid in the prediction of reservoir quality in deeply-buried marginal-marine sandstones.

This study has revealed that clay-coat mineralogy reflects that of the bulk clay mineral assemblage. Pre-drill predictions of clay-coat mineralogy (whether chlorite, illite or mixed mineralogy) should consider the initial bulk clay assemblage at the time of deposition (which in turn is influenced by palaeo-climate and the geology of the fluvial hinterland).

The results presented here suggest that facies deposited in sand-flat and tidal-bar environments in the inner estuary have clay-coats that are relatively enriched in chlorite (Fig. 7B). Facies deposited from mixed-tidal-flat and mud-tidal flat environments are relatively enriched in illite (Fig. 7A). Foreshore facies have the greatest relative enrichment in kaolinite (Fig. 7C). Figure 10 is a schematic diagram illustrating the identified spatial patterns in clay-coat mineralogy and coat grain coverage (area of grain covered by attached clay material; Wooldridge et al., 2017a,b) across a marginal-marine transect of depositional environments.

The effect of the specific clay-coat mineralogy (i.e. the ratio between illite and chlorite) on quartz cement inhibition remains uncertain. On the assumption that increasing chlorite content, in the mixed mineralogy coats from Ravenglass, will be advantageous for quartz cement inhibition, this work suggests that, in deeply-buried marginal marine sandstones, the best porosity should be found in the inner estuarine sand-flat and tidal-bar depositional elements.

The opportunity to test the applicability of such an analogue model for the prediction of clay-coat mineralogy in the subsurface is hampered by the absence of high-resolution, core-based studies of the distribution of diagenetic clay-coat mineralogy, owing to the typically poor spatial and stratigraphic coverage of cored intervals in chlorite-cemented sandstone reservoirs (Wooldridge et al., 2017b). However, the study by Skarpeid et al. (2017) on the Cook Formation, Knarr Field, Norway, reported that total sediment chlorite abundance and mean grain coat coverage are greatest in estuarine facies compared to fully marine facies. There is therefore broad concurrence between the modern Ravenglass Estuary...
and the Jurassic Cook Formation (Churchill et al., 2016; Skarpeid et al., 2017).

CONCLUSIONS

1 Clay minerals occur principally as clay-coats on sand grain surfaces (with coats representing from 61/100 to 93/100 of the total sediment clay mineral abundance).

2 On average, 81/100 of the total sediment clay mineral assemblage is present as clay-coats.

3 The mineralogy of clay-coats is spatially heterogeneous across marginal marine depositional environments.

4 Clay-coat mineralogy across the Ravenglass Estuary is dominated by illite (with an illite/(illite + chlorite + kaolinite ratio of 0.50)). High energy sediment assemblages from sand-flat and tidal-bar depositional environments have the lowest illite and highest chlorite proportions.

5 With distance towards the open ocean and away from the inner estuary, the clay-coats in outer estuarine sediments (pro-ebb delta and foreshore environments) are increasingly chlorite-deficient and illite-enriched.

6 Clay-coat mineralogy reflects the mineralogy of the bulk sediment clay mineral assemblage. This indicates that clay mineralogy is governed principally by the geology and weathering regime of the hinterland drainage area.

7 Spatial patterns in clay-coat mineralogy are governed firstly by differential settling rates of the fluvially-derived clay mineral assemblage, and secondly by the selective abrasive removal of specific clay mineral types (e.g. chlorite) during reworking and transport.

8 Based on the newly revealed distribution of clay-coating chlorite, this study suggests that, in deeply buried marginal-marine sandstones, the best prospects for elevated reservoir quality, due to clay-coat induced inhibition of quartz cement, should most likely be sought in the coarser, relatively clean, sand-flat and tidal-bar facies.

ACKNOWLEDGEMENTS

This work was carried out as part of the Chlorite Consortium at Liverpool University (UK), sponsored by BP, BG, Shell, Eni, Statoil, Petobras, Woodside and Chevron oil companies. Thanks are given to FEI (and Thermo-Fisher) for the loan of the QEMSCAN® system to the University of Liverpool (UK). The encouragement and constructive comments by reviewers Alan Butcher and Saturnina Henares Ladrón De Guevara are greatly appreciated.

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Manuscript received 30 January 2018; revision accepted 11 September 2018

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