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Palaeoclimatology

Clay mineral formation and transformation in non-marine environments and implications for Early Cretaceous palaeoclimatic evolution: The Weald Basin, Southeast England

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Abstract Analyses of clay minerals within the Early Cretaceous Weald Basin, Southeast England reveal kaolinite, illite and chlorite as the main detrital clay minerals while glauconite and smectite are subordinates. A kaolinite-rich assemblage which characterized the sand-dominated Ashdown and Tunbridge Wells Sand formations and an illite-dominated assemblage associated mostly with the Wadhurst Clay and Weald Clay formations are recognized. Kaolinite was enriched in the Ashdown and Tunbridge Wells Sand formations during warm and humid climate with high precipitation that encouraged chemical weathering and leaching, while cold and dry conditions favoured the concentration of illite in the Wadhurst Clay and Weald Clay formations. Rainfall patterns associated with warm climate were drastically reduced during the drier climatic conditions. Most clay minerals are detrital in origin, with chlorite being more prominent than previously recognized. Contrary to previous studies and assumptions, this study revealed that authigenic clay minerals are present in the Hastings Beds, with vermiform and mica-replacive kaolinite being the most common, consistent with humid depositional environments. Isolated authigenic illite is also present, along with a chloritized grain, providing evidence for mesodiagenesis. The absence of dickite and occurrence of kaolinite, suggest that authigenic illite formed in relatively shallow burial conditions, indicating a maximum burial depth of 2500 m—3000 m, about 1000 m deeper than previous estimates of 1500 m—2000 m. Authigenic clay minerals

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are absent in the Weald Clay Formation possibly because of hindered flow of meteoric water and limited growth space for authigenic minerals. This study is significant in: 1) reinforcing multiple methods to facilitate a robust and balanced knowledge of formation and transformation of clay minerals; 2) investigating detrital and authigenic clay mineral assemblages when assessing the palaeoenvironments of sedimentary basins.

Keywords Clay minerals, Clay mineral transformation, Palaeoclimate, Early Cretaceous, Weald Basin, Wealden, Southeast England

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1. Introduction

Clay minerals within sedimentary basins could be detrital in origin, formed by diagenetic transformation or a combination of both (De Segonzac, 1970; Eberl, 1984). As a result of their formation modes and transformation processes, clay minerals are very useful for understanding depositional and palaeoenvironmental conditions of sedimentary basins (e.g., Rego *et al.*, 2018; Song *et al.*, 2018). Although most clay minerals are detrital, clay mineral transformation is also common (Weaver, 1956). Consequently, clay minerals are routinely used to understand provenance, palaeoclimate, weathering and post-depositional changes (Akinlotan, 2017a; Tan *et al.*, 2017).

In this study, an integration of X-ray diffraction (XRD), optical microscopy, scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS), and quantitative evaluation of minerals by scanning electron microscopy (QEMSCAN®) datasets is used to examine the formation and transformation of clay minerals from the Weald Basin and describe the influence of clay minerals on interpreting palaeoenvironments. QEMSCAN® provides absolute quantification of minerals within rocks and can serve as an alternative to traditional microscopic description of rocks (Knappett *et al.*, 2011). QEMSCAN® works similarly to the procedure of SEM but provides the chemistry and mineralogy of the examined sample (Knappett *et al.*, 2011). The method has been used to analyse bulk and heavy mineralogy of ceramics (Knappett *et al.*, 2011), loess (Nie and Peng, 2014), ores (Anderson *et al.*, 2014; Edahbi *et al.*, 2018) and sandstones (Zhang *et al.*, 2015).

The Lower Cretaceous is an important geological phase to study palaeoenvironmental changes including eustatic, climatic and sedimentation changes that occurred during the latest Jurassic into the Early Cretaceous in many basins across the world (Hallam *et al.*, 1991; Föllmi, 2012). In Southeast England, the Lower Cretaceous facies are very important geologically and economically. The facies represent important sources of vertebrate and invertebrate fossils (Radley *et al.*, 1998) and they are also targets for conventional and unconventional hydrocarbon exploration and production (e.g., Reeves, 1948; Bray *et al.*, 1998).

Clay mineralogy studies with the Lower Cretaceous facies of England have been largely focused on the palaeoclimatic and palaeoenvironmental implications of the clay minerals (e.g., Tank, 1964; Sladen, 1987; Taylor, 1996; Jeans *et al.*, 2001; Jeans, 2006; Akinlotan, 2017a, 2018). All of these studies are based on the assumptions that the Wealden clays are detrital in origin without any significant transformation of the clay despite the fact that some clay minerals can be authigenic in origin and detrital clays can also be subjected to post-depositional transformation. This study examines the Wealden clays to describe detrital clays and diagenetic and/or post-depositional transformation of these clays and reflect on the depositional controls on their formation and transformation. The aims of the present study are to 1) interpret depositional controls on formation and transformation (diagenesis) of the clay minerals within the Weald Basin and 2) assess the palaeoenvironmental implications of formation and diagenetic transformation of clay minerals in non-marine environments.

2. Geological setting

In southeast of England, including Isle of Wight and Dorset which are not parts of current study, the term Wealden rocks refers to the non-marine sedimentary rocks between the Purbeck Group and the Lower Greensand beds (Fig. 1). Hence English Wealden is commonly used for the Lower Cretaceous facies in southeast of England. The Weald Basin (Figs. 1 and 2) has been described extensively (e.g., Allen, 1975;

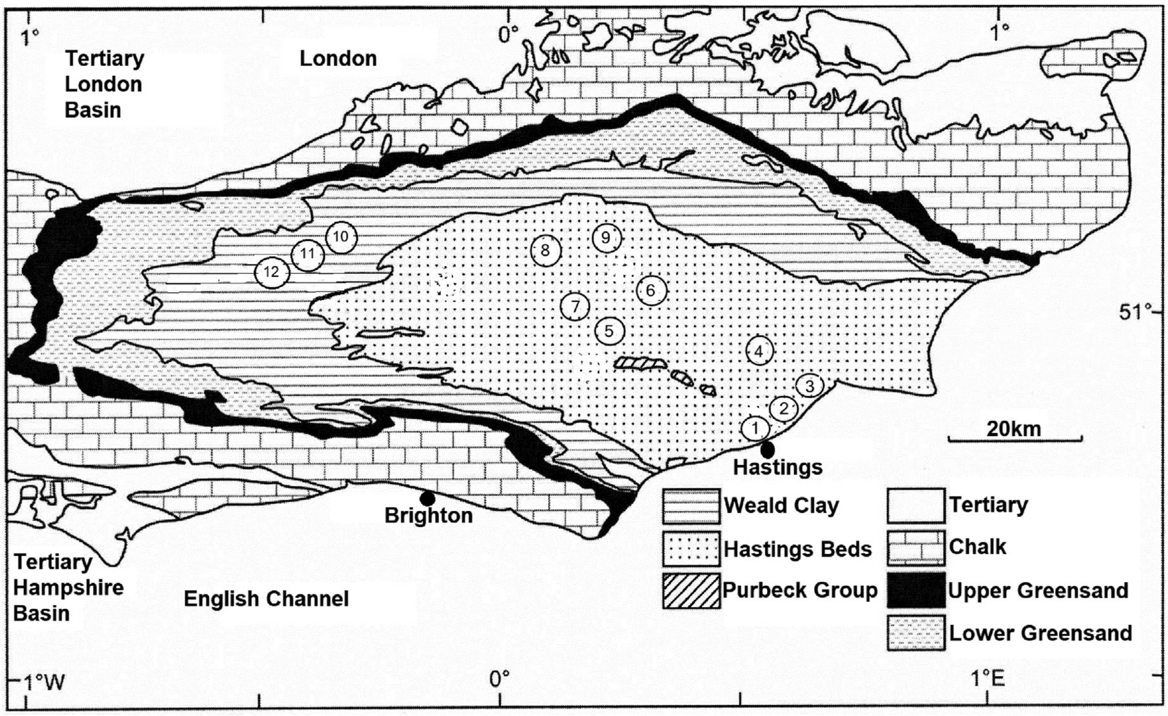

Fig. 1 The geology of the Weald Basin showing approximate study and sampling locations (1–12). Modified after Ruffell *et al.* (2006). 1 — Rock-a-Nore, Old Town, Hastings, East Sussex; 2 — Pett Level including (2a) Haddock's Cottages and (2b) Cliff End, Fairlight, near Hastings, East Sussex; 3 — Galley Hill, Bexhill, East Sussex; 4 — Freckley Hollows, Battle, East Sussex; 5 — Founthill Cutting, Newick, East Sussex; 6 — The Hermitage, High Hurstwood, East Sussex; 7 — Lake Wood, Uckfield, East Sussex; 8 — Philpots Quarry, West Hoathly, West Sussex; 9 — Harrison's Rock, Mott Hill, East Sussex; 10 — Warnham Brickworks, Warnham, West Sussex; 11 — Clock House Brickworks, Capel, Surrey; 12 — Smokejack Brickworks (Ewhurst), Ockley, Surrey.

1981; Stewart, 1981; Lake and Shephard-Thorn, 1987; Hopson *et al.*, 2008; Akinlotan, 2016, 2019), which readers can refer to for detailed information in addition to background information introduced here. The Weald Basin developed as one of the Mesozoic basins in southern England in response to Permo-Triassic rifting that formed several half-grabens (Stoneley, 1982; Chadwick, 1986; Karner *et al.*, 1987; Lake and Karner, 1987). These Mesozoic basins have similar tectonic, structural and rifting episodes (Stoneley, 1982; Chadwick, 1986; Karner *et al.*, 1987; Lake and Karner, 1987). The Weald Basin experienced significant tectonic modification in the Tertiary but the structural changes did not affect the original facies, mineralogy, etc. of the rocks in any significant manner (Stoneley, 1982; Chadwick, 1986; Karner *et al.*, 1987; Lake and Karner, 1987). The Weald Basin consists of Hastings Beds and the Weald Clay (Fig. 1). The Hastings Beds (Late Berriasian—Valanginian, Fig. 2) is an informal term comprising three sand-dominated formations: Ashdown, Wadhurst Clay and Tunbridge Wells Sand that are fluvio-deltaic deposits in nature (Allen, 1975, 1981; Lake and Shephard-Thorn, 1987; Hopson *et al.*, 2008; Akinlotan, 2016, 2017b). The Weald Clay Formation (Hauterivian—Barremian, Fig. 2) is mud-dominated and contains fluvio-deltaic/lacustrine floodplain deposits (shales, mudstones and minor siltstones and sandstones). These sediments were recycled from adjacent massifs and deposited in generally non-marine settings within the basin.

3. Methodology

One hundred and twenty-two surface and unweathered samples were collected from 12 locations (Fig. 1) across the Weald Basin and used for this study. The unweathered samples were subjected to laboratory analyses described below.

3.1. X-ray diffraction (XRD)

In order to examine the mineralogical composition of mudstone and shale, XRD analysis was carried out on 80 samples using a Bruker D8 Advance X-ray diffractometer with a Lynx-eye detector. About 5 g of powdered samples were placed in polymethyl methacrylate sample holders and scanned using 40 kV,

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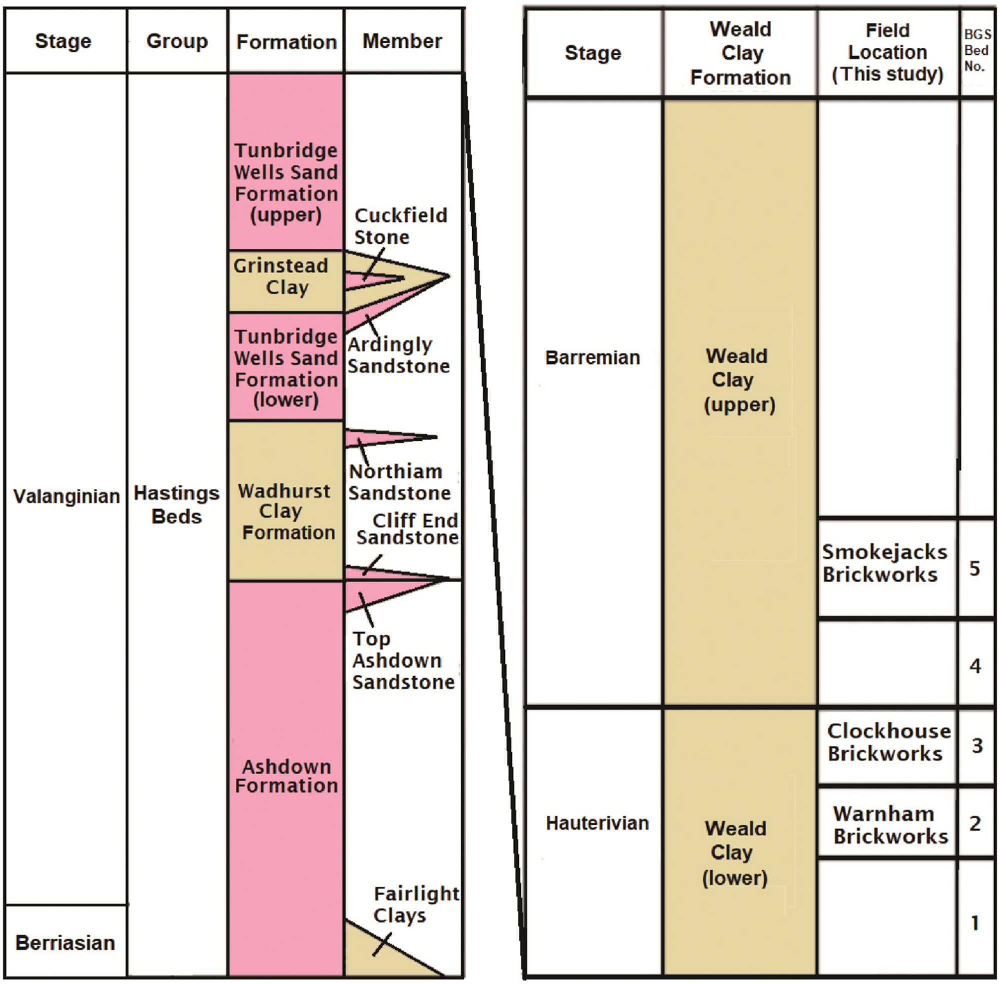

Fig. 2 The Wealden stratigraphy in the Weald Basin. Note that the Hastings Beds is older; and the Weald Clay Formation is younger than the Tunbridge Wells Sand Formation. After Akinlotan *et al.* (2021a)'s figure 2, not drawn to scale.

40 mA and CuKα radiation. The powdered samples were scanned from 5° to 100° and 3°–30° for whole rock and clay-fraction composition respectively, with a step size of 0.009°. Diffrac Commander software was used to collect the data while phase identification was carried out on the Diffrac Eva software with Rietveld refinement performed using Diffrac TOPAS pattern fitting software. These measurements were conducted at the Experimental Techniques Centre (ETC) of Brunel University, London.

3.2. QEMSCAN®, SEM and optical petrography

QEMSCAN® (quantitative evaluation of minerals by scanning electron microscopy), SEM (scanning electron microscopy) and optical petrographic analyses were carried out at CGG Robertson: 122 samples for QEMSCAN® analysis and 17 samples out of the 122 samples for SEM and optical petrographic analyses. The full details of the procedure for these analyses have been provided (Akinlotan and Rogers, 2021; Akinlotan *et al.*, 2021a, 2021b) and summarised below. For QEMSCAN® analysis, samples were either prepared into thin section or into polished resin blocks and analysed using Quanta 650F machine. iDiscover software suite and CGG Robertson's proprietary mineral dictionary were used for data analysis to acquire textural and mineralogical information of the samples. QEMSCAN® technique uses SEM–EDS data to determine the mass of elements at each point. At each point, the relative concentration of elements is used to identify the mineral species. Consequently, the mass data are combined with the known mineral density from the dictionary to obtain the total mineralogy volume. Seventeen samples, from the same samples analysed for the QEMSCAN® procedure described above were subjected to standard optical microscopy and SEM analysis which have been fully described in Akinlotan *et al.* (2021b). Petrographic photographs were taken from these thin sections using a Zeiss AX10 microscope.

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SEM images were acquired using a BSE detector and a secondary electron detector. The acquisition of EDX (energy-dispersive X-ray spectroscopy) data was achieved through Bruker EDX detectors while the data were analysed using Bruker's ESpirit software.

Authigenic minerals were identified through SEM and EDX analyses based on the morphology and chemical composition of clay minerals following the methods described in the literature (e.g., Burley and Macquaker, 1992; Ahmed, 2007; Taylor and Macquaker, 2014). Authigenic clay minerals can be more challenging to detect in mudstones, compared with sandstones, due to their typically finer size and the potential to be obscured by intergrowth with other authigenic phases such as quartz overgrowth cements. Additionally, the small size means when performing EDX analysis the authigenic clays can be smaller than the beam spot size, resulting in a higher degree of contamination in the spectra. However, while the spectra show the elements in the target clay and surround minerals, it can still be indicative of the clay composition, and be used in conjunction with clay morphology to aid in clay mineral identification. Despite the challenges, SEM and EDX analyses have commonly been successfully used to detect and study authigenic clay minerals in mudstones and shales (Burley and Macquaker, 1992; Macquaker and Gawthorpe, 1993; Ahmed, 2007; Taylor and Macquaker, 2014; Gier *et al.*, 2018).

4. Results

4.1. Detrital clay minerals

4.1.1. Occurrence

Optical microscopy reveals that the deposits in Weald Basin contain kaolinite, illite and chlorite (Fig. 3). Thin-section observations show that the clay minerals are commonly distributed evenly as detrital grain-coating clay, with grain-coats typically up to about 10 μm thick (Figs. 3 and 4A). Although clays are heterogeneously distributed, they also concentrate along clay-rich zones or impersistent laminae containing grain-rimming and pore-filling detrital clays (Fig. 4B). Observations from SEM—EDX analyses reveal kaolinite, illite, chlorite and smectite in the Weald Basin (Figs. 5 and 6). SEM—EDX analysis of selected crystals (Fig. 5) reveals K, Al, Si, O, C, Fe, and Mg as the common elements (Fig. 6) and Na, Ti, and Cl as less common. Extrapolation based on elemental combinations of the examined mineral crystals was used to identify kaolinite, illite, chlorite and smectite. Results from qualitative XRD analysis show the presence of kaolinite, illite and chlorite, and kaolinite appears to be the dominant mineral (Fig. 7). Chlorite minerals occur in the form of clinochlore (Fig. 7). QEMSCAN® results show the presence of kaolinite, illite, chlorite, glauconite and smectite (Table 1) with kaolinite, illite and chlorite dominating while glauconite and smectite have the least concentrations (Fig. 8).

4.1.2. Abundance

This section describes the abundance of clay minerals across different stratigraphic divisions of the Weald Basin. Kaolinite (52.1%—54.9%) and illite (37.4%—41.4%) are the main detrital clay minerals in the Ashdown Formation, with kaolinite typically being marginally more abundant than illite (Table 1). Only minor quantities of chlorite are present (Table 1). Samples from the lower part of the Ashdown Formation at sampling location Rock-A-Nore show ca. 55% kaolinite whereas it is ca. 52% at sampling location Haddock’s Cottages (Table 1; Appendix 1). Illite is the second dominant clay mineral in the Ashdown Formation, making up ca. 41% at Rock-A-Nore and decreases to ca. 37% at Haddock’s Cottages (Table 1). Chlorite makes up <5% of the clay minerals throughout the Ashdown Formation (Fig. 8). In the Wadhurst Clay Formation, illite (37.4%) and kaolinite (35.2%) form the dominant clay minerals, with illite typically more abundant than kaolinite in any given samples (Table 1; Appendix 1). Chlorite is present in small quantities, except for within the ironstones and shale sampled at Pett Level, Fairlight (Table 1), where it is a major clay phase (22.1%). Indeed, chlorite comprises the main clay mineral within the ironstones (Table 2).

In the Ardingly Sandstone Member within the Tunbridge Wells Sand Formation, kaolinite (38.7%), chlorite (43.8%), and to a lesser extent, illite (16.7%) form the main clay mineral phases (Table 2). In the Ardingly Sandstone Member samples, kaolinite and illite typically display similar concentrations, with chlorite being less abundant. At Founthill Cutting (Newick), kaolinite dominates whereas in the samples from The Hermitage (High Hurstwood), it is chlorite. The Tunbridge Wells Sand Formation shows high concentration of kaolinite, chlorite and illite at different locations and intervals. Conditions favouring the production of chlorite were observed in chlorite-dominated clay minerals in the lower part of the Ardingly Sandstone Member (The Hermitage) (ca. 50%), with kaolinite and illite making up ca. 34% and ca. 14% of the clay minerals. Chlorite abundance is notably reduced in the

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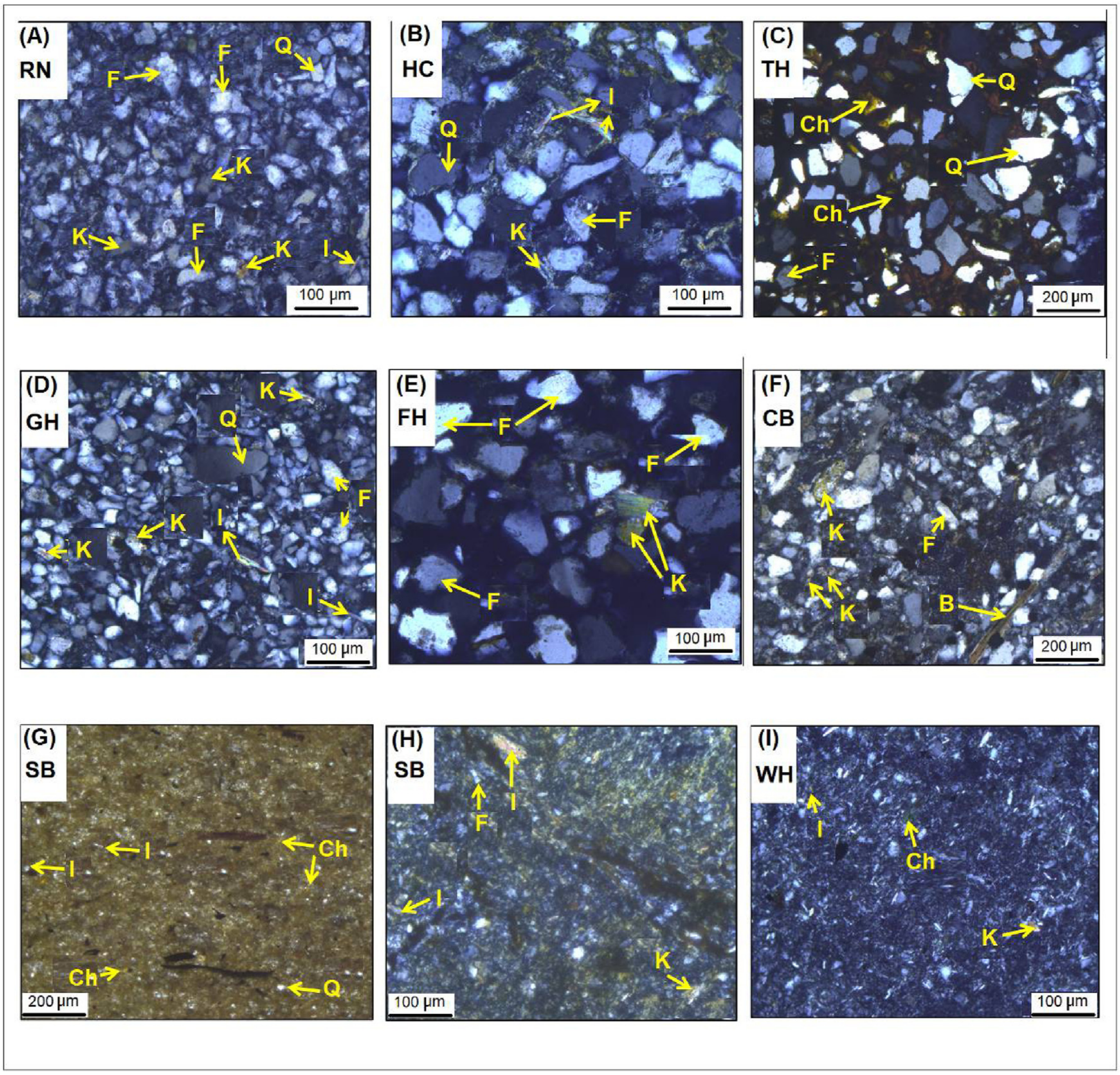

Fig. 3 Thin section photomicrographs of selected rocks from the Ashdown, Tunbridge Wells Sand, and Weald Clay formations. **A)** and **B)** Ashdown Formation — Rock-A-Nore (RN), Haddock's Cottages (HC); **C—E)** Tunbridge Wells Sand Formation — The Hermitage (TH), Galley Hill (GH), Freckley Hollows (FH); **F—I)** Weald Clay Formation — Clock House Brickworks (CB), Smokejack/Ewhurst Brickworks (SB), Warnham Brickworks (WH). Identified minerals include kaolinite (K), illite (I), chlorite (Ch), quartz (Q), feldspar (F) and biotite (B).

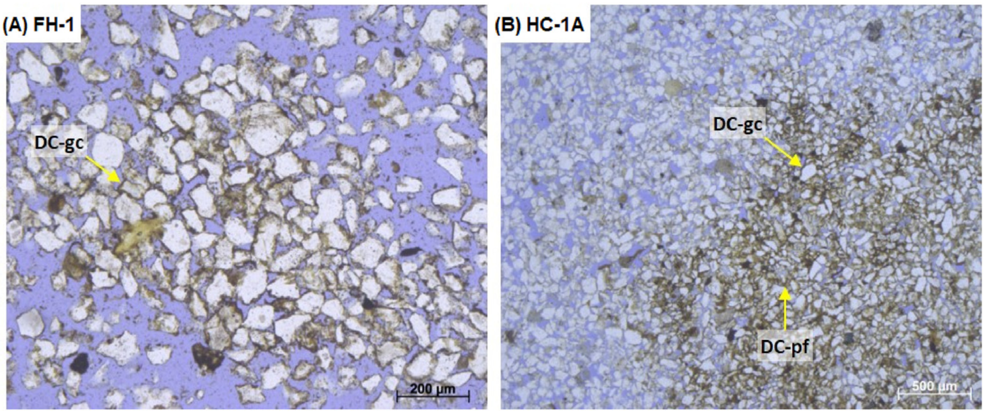

Fig. 4 Thin-section photomicrographs showing the distribution of detrital clay minerals in Hastings Beds sandstones: **A)** Evenly distributed grain-coating detrital clays (DC-gc) and **B)** A local clay-rich region displaying grain-coating and pore-filling (DC-pf) detrital clays. Sampling locations: FH — Freckley Hollows; HC — Haddock's Cottages.

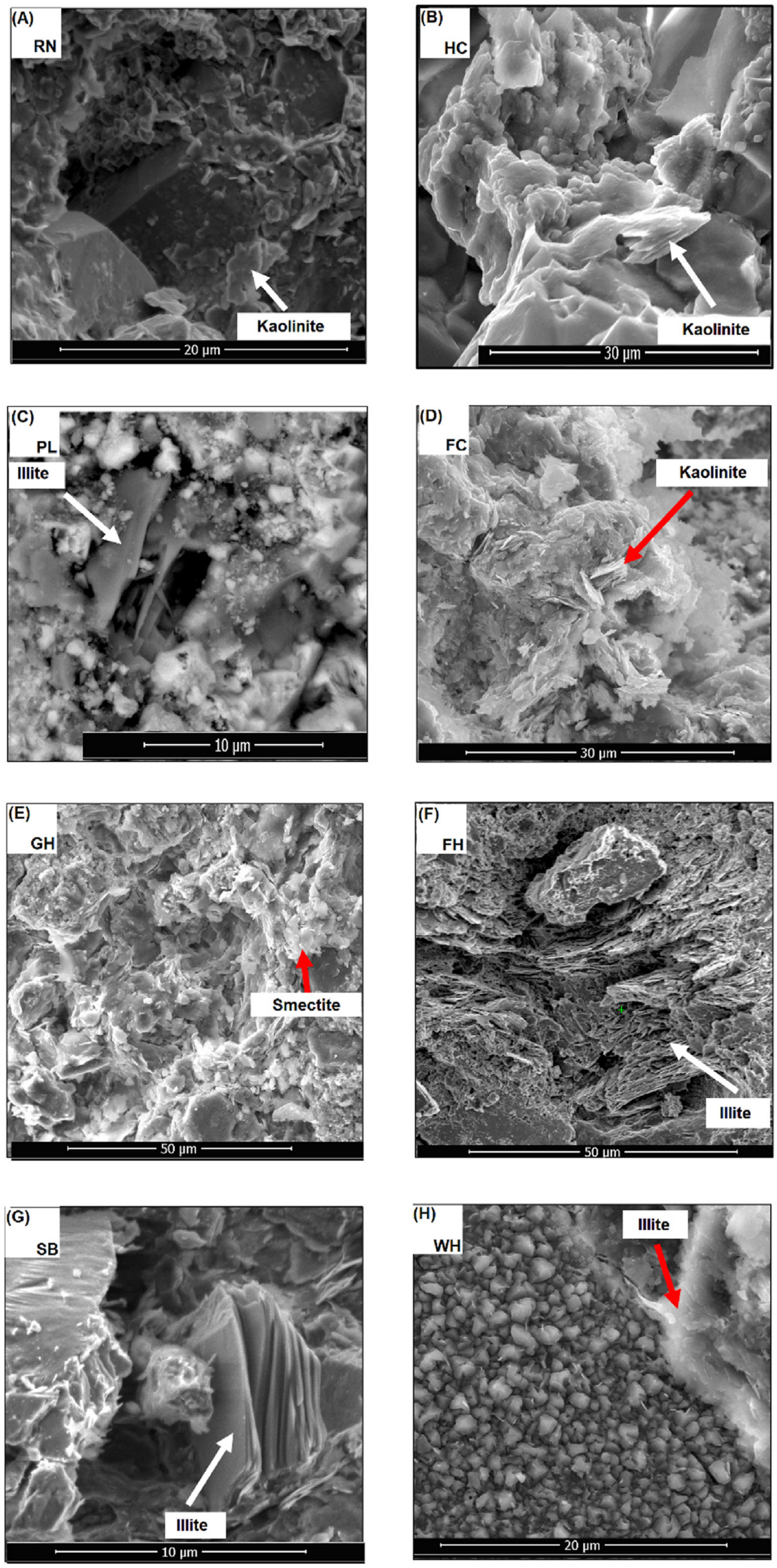

Fig. 5 SEM images of clay minerals selected from the Ashdown Formation: **A)** Kaolinite sampled in Rock-A-Nore (RN); **B)** Kaolinite sampled in Haddock's Cottages (HC); Wadhurst Clay Formation: **C)** Illite sampled in Pett Level (PL); Tunbridge Wells Sand Formation: **D)** Kaolinite sampled in Founthill Cutting (FC); **E)** Smectite sampled in Galley Hill (GH); **F)** Illite sampled in Freckley Hollows (FH); Weald Clay Formation: **G)** Illite sampled in Smokejack/Ewhurst Brickworks (SB); **H)** Illite sampled in Warnham Brickworks (WH).

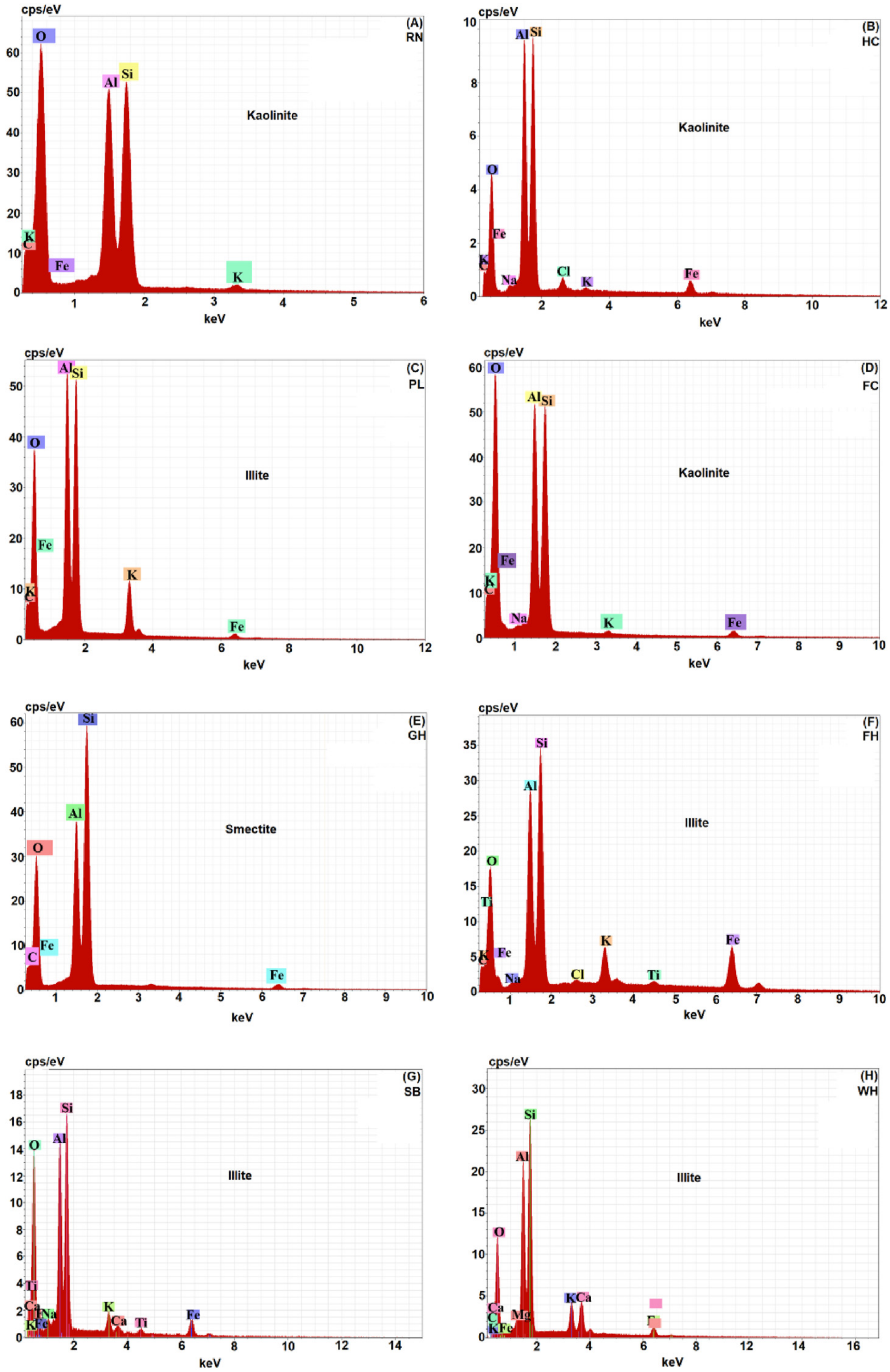

Fig. 6 EDS graphs showing the elemental constituents of selected rocks sampled in Ashdown Formation: **A)** Rock-A-Nore (RN); **B)** Haddock's Cottages (HC); in Wadhurst Clay Formation: **C)** Pett Level (PL); in Tunbridge Wells Sand Formation: **D)** Founthill Cutting (FC); **E)** Galley Hill (GH); **F)** Freckley Hollows (FH); in Weald Clay Formation: **G)** Smokejack/Ewhurst Brickworks (SB); **H)** Warnham Brickworks (WH). Al — Aluminium; Si — Silicon; O — Oxygen; C — Carbon; K — Potassium; Na — Sodium; Fe — Iron; Ca — Calcium; Mg — Magnesium; Ti — Titanium; Cl — Chlorine.

samples at Harrison's Rock and kaolinite becomes the dominant clay mineral (ca. 49%). Illite also increases to ca. 33%, while chlorite reduces to about 14%. Kaolinite remains dominant in the middle part of the Ardingly Sandstone Member at Founthill Cutting, constituting about 46% of the total clay minerals (Fig. 9). Chlorite is observed to surpass illite, increasing to ca. 34%, while illite reduces to about 18% of the total clay minerals.

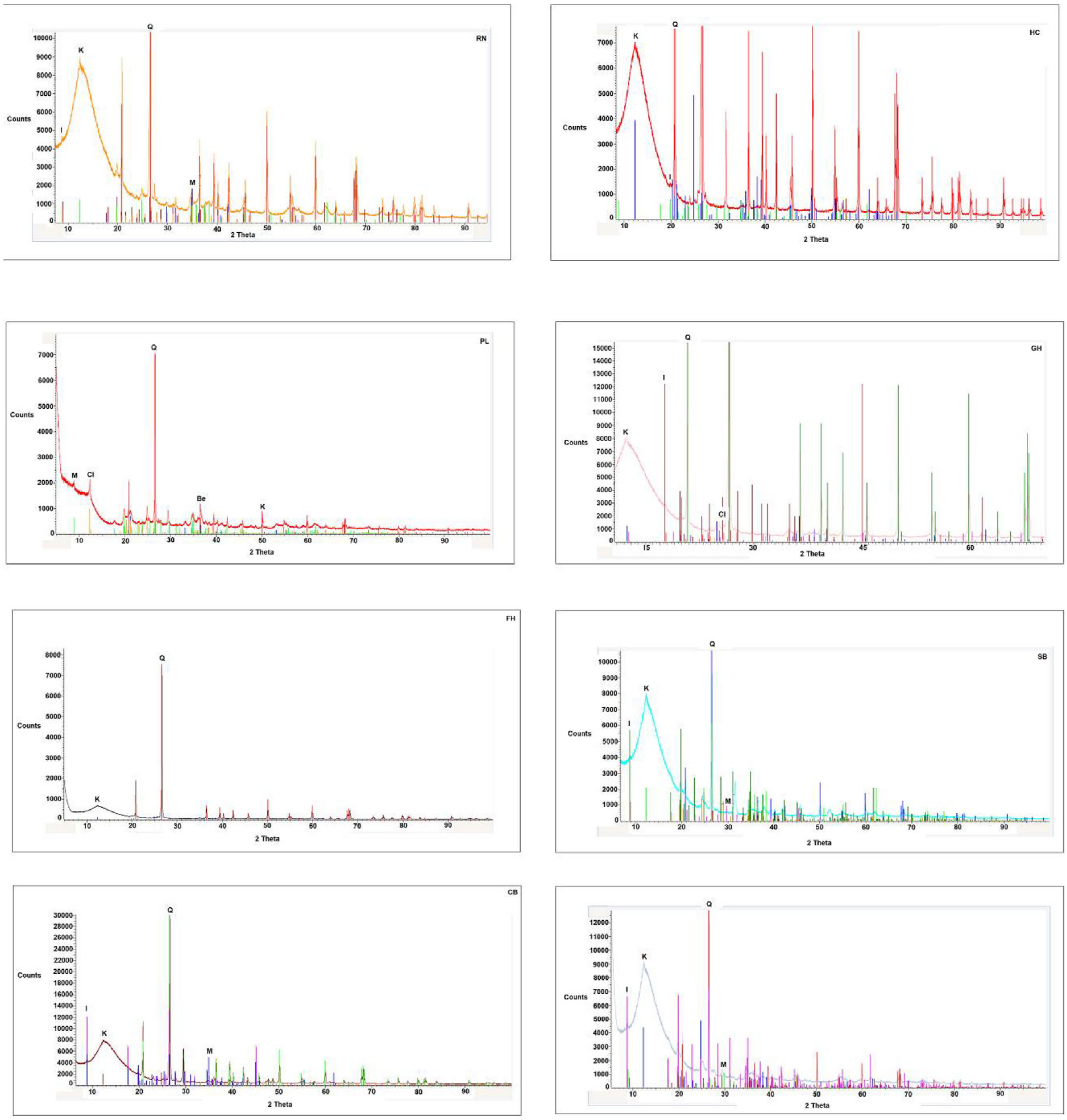

Fig. 7 Selected XRD phase diagrams showing the clay minerals from the Weald Basin. RN — Rock-A-Nore; HC — Haddock's Cottages; PL — Pett Level; GH — Galley Hill; FH — Freckley Hollows; SB — Smokejack/Ewhurst Brickworks; CB — Clock House Brickworks; WH — Warnham Brickworks. K — Kaolinite; I — Illite; M– Muscovite; Q — Quartz; Cl — Clinochlore; Be — Berthierine.

Illite slightly surpasses kaolinite in the upper part of the Ardingly Sandstone Member at Lake Wood, Uckfield (ca. 44.5% and 43.5% respectively), with chlorite reducing to ca. 10.8% of the total clay minerals (Table 1). The uppermost part of the Ardingly Sandstone Member at Philpots Quarry is characterized by an increase in chlorite (ca. 69%), and reductions in kaolinite and illite concentrations (14.9% and 15.7% respectively). A return to kaolinite dominance is observed in the upper part of the Tunbridge Wells Sand Formation. Kaolinite makes up ca. 49% of the total clay minerals at sampling location Galley Hill (Table 1). Illite and chlorite constitute 33.8% and 16.8% respectively of the total clay minerals (Table 1). At Freckley Hollows, kaolinite further increases to 61.8%, while illite and chlorite further decrease to 24.6% and 12.9% respectively (Table 1).

Illite (52.5%—76.2%) typically forms the dominant clay mineral phase in the Weald Clay Formation, with some intervals containing similar quantities of illite and kaolinite while chlorite (4.9%—12.6%) is the least abundant of the three main clay minerals (Table 1). Samples from the Weald Clay Formation at Clock House Brickworks show dominant illite concentration (ca.

Table 1 Summary of relative proportions of clay minerals from QEMSCAN® analysis.

Formation	Sampling locations	Kaolinite (%)	Illite (%)	Chlorite (%)	Smectite (%)	Glauconite (%)
Weald Clay	Smokejack/Ewhurst Brickworks, Ockley (SB)	40.9	45.7	12.6	0.7	0.1
	Clock House Brickworks, Capel (CB)	17.4	76.2	4.9	1.3	0.3
	Warnham Brickworks, Horsham (WH)	36.6	52.5	9.5	1.3	0.1
Tunbridge Wells Sand	Freckley Hollows, Battle (FH)	61.8	24.6	12.9	0.3	0.4
	Galley Hill, Bexhill (GH)	49.0	33.8	16.8	0.3	0.1
	Philpots Quarry, West Hoathly (PQ)	14.9	15.7	69.0	0.3	0.1
	Lake Wood, Uckfield (LW)	43.5	44.5	10.8	0.8	0.4
	Founthill Cutting, Newick (FC)	46.2	18.5	34.5	0.3	0.5
	Harrison's Rock, Mott Hill, (HR)	49.3	33.6	13.1	2.3	1.7
	The Hermitage, High Hurstwood (TH)	34.3	14.3	50.9	0.3	0.1
Wadhurst Clay	Pett Level, Fairlight (PL)	35.2	41.3	22.1	0.9	0.4
Ashdown	Haddock's Cottages, Fairlight (HC)	52.1	37.4	8.6	1.6	0.3
	Rock-A-Nore, Hastings (RN)	54.9	41.4	3.0	0.6	0.2

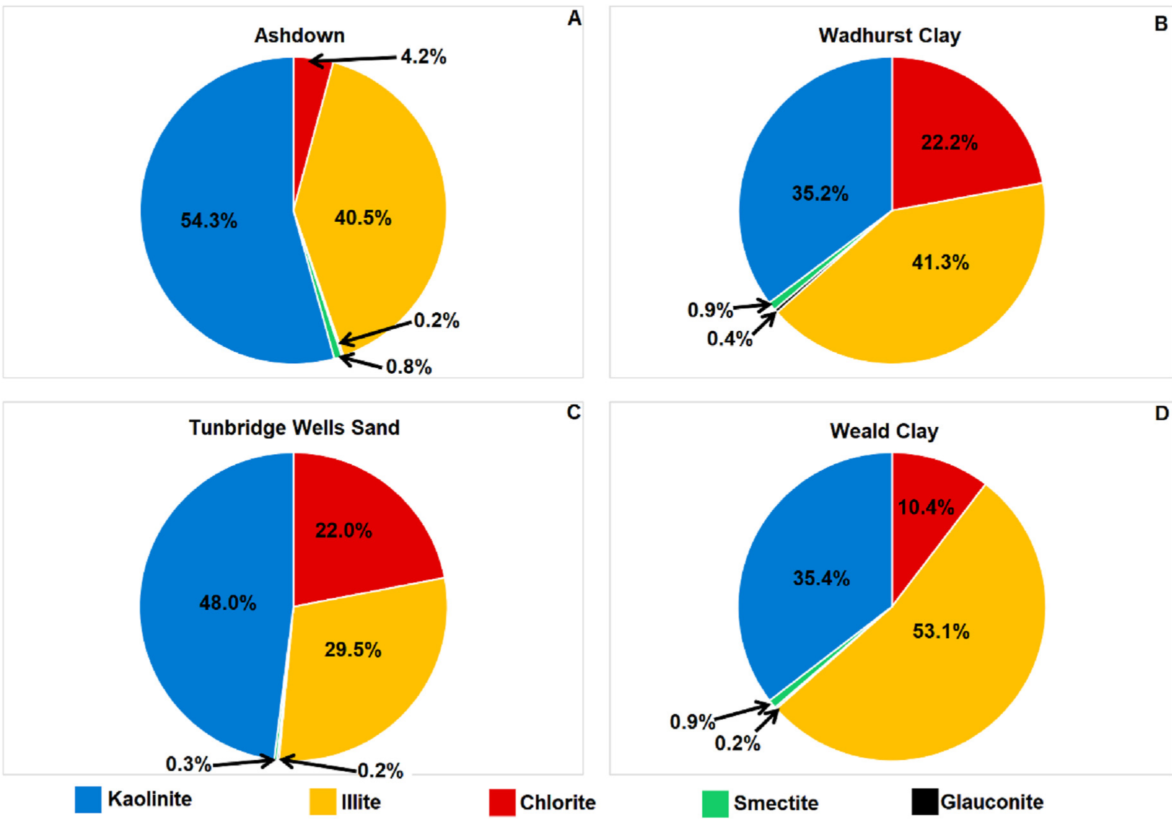

Fig. 8 Clay mineral concentrations within the Wealden rocks from the **A)** Ashdown, **B)** Wadhurst Clay, **C)** Tunbridge Wells Sand, and **D)** Weald Clay formations.

76.2%) with decreased kaolinite and chlorite (17.4% and 4.9% respectively). At Smokejack/Ewhurst Brickworks (Fig. 9), illite dominates (ca. 45%), however, kaolinite and chlorite increase to ca. 40% and ca. 12% respectively. Illite continues to dominate in the Weald Clay Formation at Warnham Brickworks (ca. 52%), while kaolinite and chlorite decrease to ca. 36% and ca. 9% respectively (Fig. 9). Black shales and ironstones form an exception to this, which either contain more kaolinite than illite, or similar quantities of the two. In the black shales, chlorite is consistently the least abundant clay mineral, but in the upper (youngest) three ironstone samples it is the most abundant (Table 2).

4.1.3. Trends

Observations from clay mineral trends in the Weald Basin (Fig. 9) revealed two major patterns of depositions: 1) the Ashdown and Tunbridge Wells Sand

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Table 2 Average QEMSCAN® datasets for the three main clay mineral types identified in the Hastings Beds and Weald Clay Formation showing the variation between different stratigraphic units and lithologies. Data are presented as (a) volumetric percentages (vol. %), and (b) relative abundance of total clays (rel. %).

Stratigraphic unit	Mudstone and sandstone			Ironstone and Fe-cemented mudstone			Black shale		
	Kaolinite	Illite	Chlorite	Kaolinite	Illite	Chlorite	Kaolinite	Illite	Chlorite
(a) Clay abundances (vol. %)									
Weald Clay Formation	13.59	24.17	3.95	5.62	3.97	6.9	32.86	28.4	4.19
Tunbridge Wells Sand Formation	9.36	6.08	2.15	1.26	0.95	5.78	n/a	n/a	n/a
Ardingly Sandstone Member	1.64	0.71	1.85	0.45	0.21	1.08	n/a	n/a	n/a
Ashdown and Wadhurst Clay formations	10.98	9.64	1.42	3.37	4.32	17.02	n/a	n/a	n/a
(b) Clay abundances (rel. %)									
Weald Clay Formation	32.21	57.29	9.37	33.6	23.75	41.27	49.86	43.08	6.35
Tunbridge Wells Sand Formation	52.97	34.41	12.17	15.69	11.78	71.84	n/a	n/a	n/a
Ardingly Sandstone Member	38.77	16.78	43.81	25.90	12.13	61.74	n/a	n/a	n/a
Ashdown and Wadhurst Clay formations	49.21	43.2	6.37	13.58	17.41	68.55	n/a	n/a	n/a

formations are characterized by kaolinite-dominated deposition. However, kaolinite production reduced in the Tunbridge Wells Sand Formation, hence giving room for increased chlorite concentration; 2) an illite-dominated deposition characterizes the Wadhurst Clay and Weald Clay formations. Illite concentration in the Weald Clay Formation surpasses that of the Wadhurst Clay Formation, hence reducing chlorite and causing

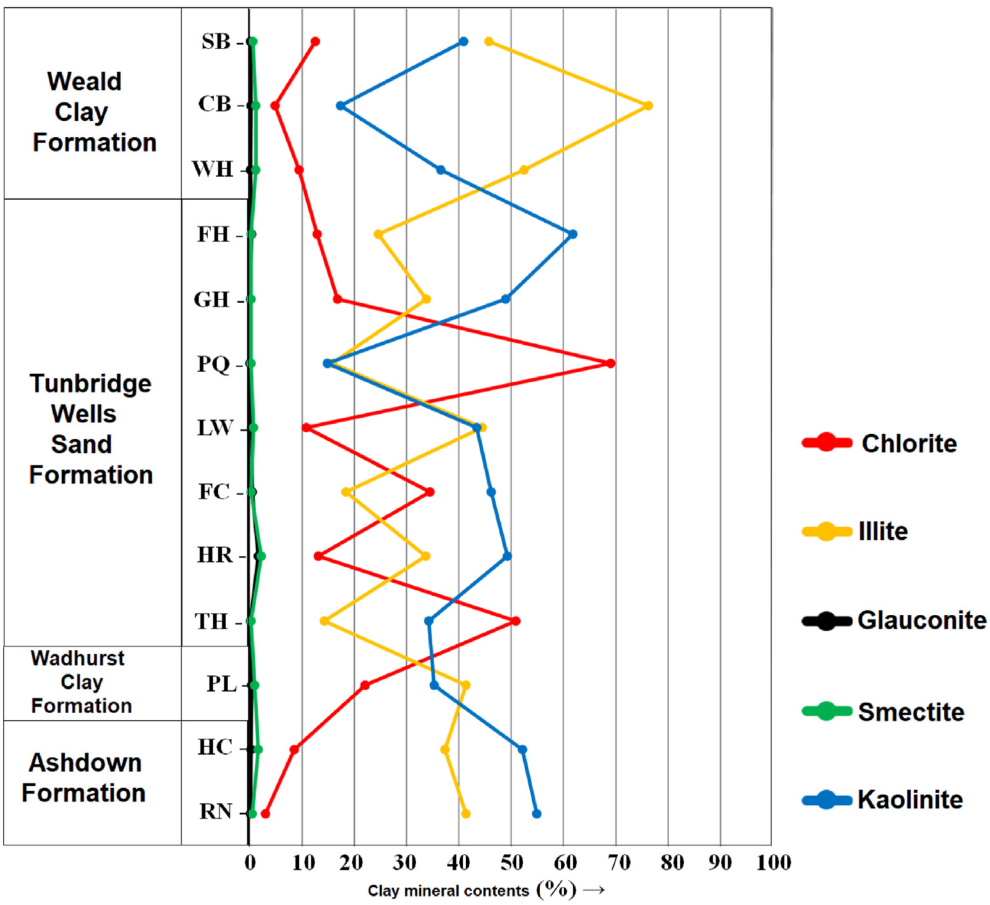

Fig. 9 Clay mineral trend within the Wealden rocks from the Ashdown, Wadhurst Clay, Tunbridge Wells Sand, and Weald Clay formations. Ashdown Formation: RN — Rock-A-Nore; HC — Haddock's Cottages; Wadhurst Clay Formation: PL — Pett Level; Tunbridge Wells Sand Formation: TH — The Hermitage; HR — Harrison's Rock; FC — Founthill Cutting; LW — Lake Wood; PQ — Philpots Quarry; GH — Galley Hill; FH — Freckley Hollows; Weald Clay Formation: CB — Clock House Brickworks; SB — Smokejack/Ewhurst Brickworks; WH — Warnham Brickworks.

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illite to increase to more than 50% of the total clay minerals in the Weald Clay Formation (Figs. 8 and 9). On a basin scale, clay minerals within the Hastings Beds are dominated by kaolinite, which constitutes from 14% to 61% of the total clay minerals. The Weald Clay Formation contains <41% kaolinite and is instead made up predominantly of illite, which constitutes between 45% and 76% of the total clay minerals.

Kaolinite to illite + chlorite ratios (K/[I + C]) can be used to identify changes in humidity with low value of this ratio indicating aridity while high value indicates humid conditions (Hesselbo *et al.*, 2009; Bougeault *et al.*, 2017; Zuo *et al.*, 2019). K/[I + C] ratios range from 0.18 to 1.65 (Fig. 10). The Ashdown and Tunbridge Wells Sand formations which have high kaolinite have K/[I + C] ratios which are greater than 0.8 (Fig. 10). The uppermost part of Tunbridge Wells Sand Formation (at Freckley Hollow) records the highest K/[I + C] ratio of 1.65 due to a peak in kaolinite production (ca. 61% of the total clay minerals) (Figs. 9 and 10). K/[I + C] ratios less than 0.8 were observed in samples where illite or chlorite surpasses kaolinite as the dominant mineral in localised areas underlain by the Tunbridge Wells Sand Formation (The Hermitage, Lake Wood and Philpots Quarry). K/[I + C] ratios lower or equal to 0.7 were noted in areas with high concentration of illite. For instance, the highest concentration of illite in the Weald Clay Formation at Clockhouse Brickworks (ca. 76% of the total clay minerals; Fig. 9) is observed to yield a K/[I + C] value of ca. 0.21 (Fig. 10). Glauconite and smectite are the least dominant clay minerals, however they have similar trend showing a marked increase within the Tunbridge Wells Sand Formation (Harrison's Rock) and a corresponding sharp decrease in chlorite (Fig. 10). The reverse is also the case in areas with increase concentration of chlorite having lower concentration of both smectite and glauconite.

4.2. *Authigenic clay minerals*

4.2.1. *Occurrence*

Compared to detrital clays, authigenic clays are less significant in the Weald Basin, which were only observed within the Hastings Beds and were not identified in the Weald Clay Formation. The characteristics of the dominant authigenic clay minerals are summarised here. Three forms of kaolinite observed from the Hastings Beds were identified based on textural habits. Mica-replacive kaolinite occurs as grain-coating to pore-filling clay with

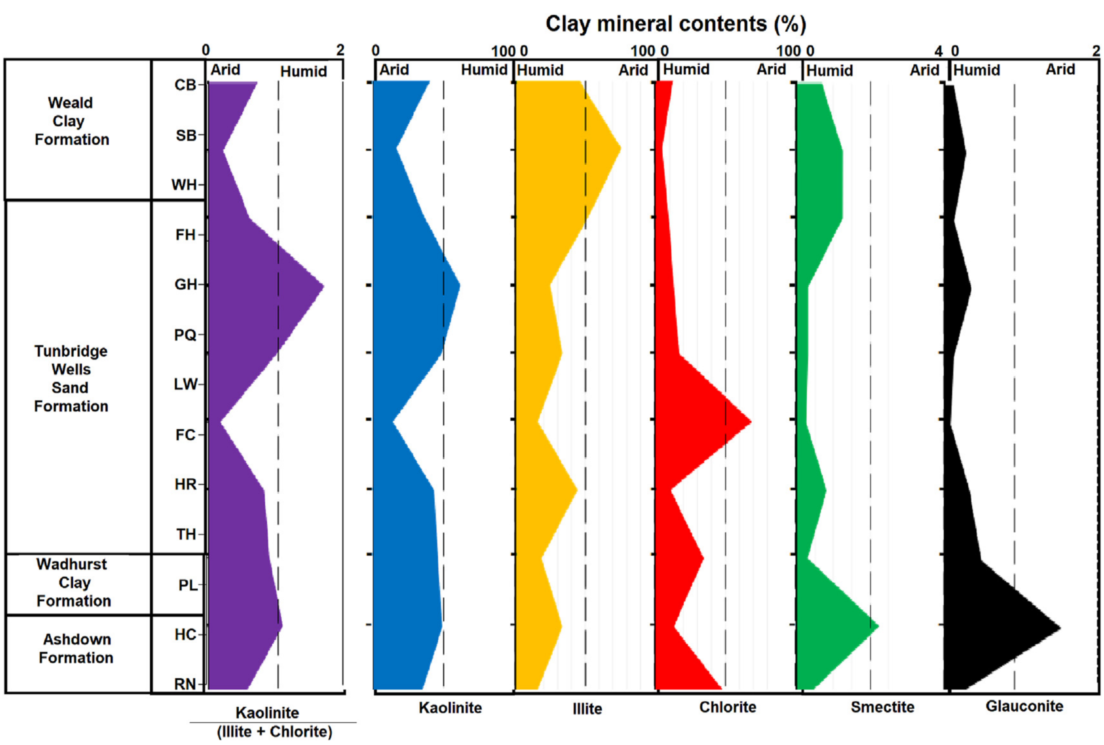

Fig. 10 Fluctuations in kaolinite to illite + chlorite ratio (K/[I + C]) values and individual clay mineral trend within the Wealden rocks from the Ashdown Formation: RN — Rock-A-Nore; HC — Haddock's Cottages; from the Wadhurst Clay Formation: PL — Pett Level; from the Tunbridge Wells Sand Formation: TH — The Hermitage; HR — Harrison's Rock; FC — Founthill Cutting; LW — Lake Wood; PQ — Philpots Quarry; GH — Galley Hill; FH — Freckley Hollows; and, from the Weald Clay Formation: CB — Clock House Brickworks; SB — Smokejack/Ewhurst Brickworks; WH — Warnham Brickworks.

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variable textural habits consisting of isolated plates, platy aggregates or squashed platy texture (Fig. 11) with EDX spectrums showing Al and Si as the primary constituent elements (Fig. 6). The crystals are 0.6 μm–5 μm (average of 2.4 μm) in diameter. The crystal plates are randomly distributed in pores and on detrital framework minerals and engulfed by quartz overgrowth in places. Vermiform kaolinite shows a similar EDX spectrum and occurrence (grain-coating to pore-filling clay) as the mica-replacive type. However, the vermiform kaolinite differs by having a distinct book- or worm-like (vermicular) habit and is distinctly engulfed by iron oxide (Fig. 11F). The thickness of each crystal plate is often less than 1.0 μm. Blocky kaolinite shows a similar EDX spectrum as the mica-replacive and vermiform kaolinite but occurs mainly as pore-filling clay. This kaolinite clay consists of thin, blocky crystal aggregates (1.0–5.6 μm) and interleaves with a few kaolinite verms (<1.0 μm). This kaolinite is distinctly associated with or engulfed by euhedral quartz overgrowth and authigenic illite (Fig. 11B, D). Authigenic illite clay occurs as fibrous crystals (about 2.5 μm thick) with curled or filamentous edges coating grains or thin hair-like strands (about 1.0 μm long) extending tangentially or perpendicular from the grain and to bridge pores (Fig. 11D). The fibrous form appears to be more dominant than the hair-like

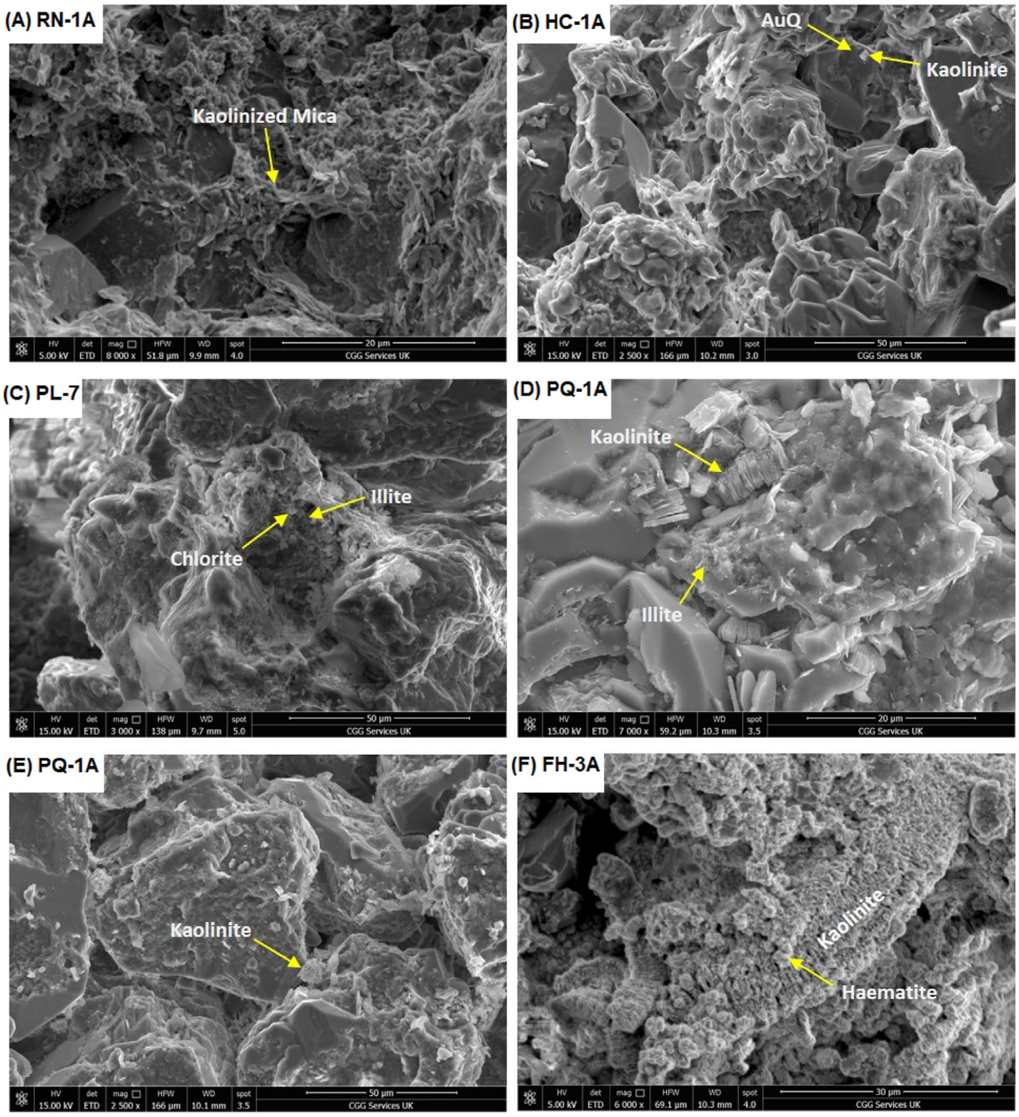

Fig. 11 SEM images of authigenic clays identified in the Hastings Beds sandstones, Weald Basin: **A)** Highly altered and kaolinized mica; **B)** A small, rare kaolinite verm partially enclosed in an authigenic quartz overgrowth (AuQ); **C)** A chloritic/chloritized grain which also contains less abundant authigenic illite; **D)** Vermiform kaolinite and rare fibrous illite; **E)** A pore-throat plugging kaolinite verm; **F)** Variably sized kaolinite verms partially covered in haematite cement. Sampling locations: RN – Rock-A-Nore; HC – Haddock's Cottages; FH – Freckley Hollows; PL – Pett Level; PQ – Philpots Quarry.

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texture. However, both are found in association with blocky kaolinite and enveloped by quartz overgrowths. This study has presented the evidence for the presence of authigenic clay minerals in the Weald Basin.

4.2.2. Trends

Within the Ashdown Formation, two distinct forms of authigenic kaolinite were identified by SEM and EDS methods. The first is disordered kaolinite (crystals up to 2 μm in length) replacive of highly expanded and altered (kaolinized) mica (Fig. 11A); while the second occurs in form of long stacks of kaolinite sheets — generally small, isolated, up to 4 μm and partially enclosed by quartz overgrowths (Fig. 11B). Authigenic clays within the Wadhurst Clay Formation were found on a chloritic/chloritized grain within brown sandstone at Pett Level (Fairlight; Fig. 11C). This grain contains randomly orientated, subhedral to euhedral, chlorite blades (up to ~2 μm in length) mixed with possible, less abundant, finer crystalline illite fibres, which is supported by a K signal in the EDS spectrum. This grain also contains probable authigenic iron-oxide crystals. Authigenic clay minerals were identified at the top of the unit at Philpots Quarry (West Hoathly) within the Ardingly Sandstone Member. The brown sandstone contains authigenic pore-lining and pore-filling vermiform kaolinite, with individual verms up to 10.5 μm long (Fig. 11D and E). Less abundant pore-lining fibrous authigenic illite up to 0.9 μm in length is also present, which locally overlies kaolinite verms (Fig. 11D). Both clays are locally partially enveloped by authigenic quartz overgrowths. Within the upper part of the Tunbridge Wells Sand Formation (Freckley Hollows, Battle), authigenic kaolinite occurs in brown sandstone as well-developed, pore-filling vermiform kaolinite (Fig. 11F). Here, it is notably coarser crystalline than seen anywhere else in the Hastings Beds, with individual verms up to about 50 μm long. Some verms are partially coated by haematite cement.

5. Discussion

5.1. Provenance of detrital clay minerals

It is well documented that rocks within the Weald Basin were sourced and recycled from multiple sources including Londinian, Amorican, Cornubian massifs and Boreal Sea (Allen, 1975; Sladen and Batten, 1984; Allen and Wimbledon, 1991) meaning that most of the clays are detrital in nature. This study presents some lines of evidence to support this conclusion. SEM and EDS results from the current study show the presence of detrital clay minerals (Figs. 5 and 6). Previous studies have shown that the Wealden rocks experienced relatively shallow burial depth of about 2 km (Sladen and Batten, 1984). The occurrence and dominance of kaolinite observed in the current study throughout the Weald Basin (Fig. 8) supports shallow burial depth as kaolinite may be destroyed by high temperatures due to deeper burial (Tucker, 2001). In addition, optical microscopy from this study shows that the sands are characterized with line contacts with adjacent grains, which may imply loose grain packing and translate to lesser compaction probably due to shallow burial depth (Sladen, 1983, 1987; Sladen and Batten, 1984; Jeans, 2006).

5.2. Detrital clay minerals and the English Wealden palaeoenvironments

The production of kaolinite is favourable in warm and moist environments with intense chemical weathering and leaching profiles (Chamley, 1989; Proust et al., 1995; Thiry, 2000; Song et al., 2018). The Ashdown Formation contains abundant kaolinite, with a relatively lesser amount of illite and chlorite. This indicates that the palaeoclimate of the formation was a generally warm temperate-subtropical one with high levels of humidity and rainfall patterns that allowed for severe chemical weathering and leaching. Illite and chlorite were formed through physical weathering of low-grade metamorphic rocks and minimal chemical weathering (Chamley, 1989; Alizai et al., 2012), hence demonstrating a limited leaching profile. The change from kaolinite to illite dominance, as well as increase in chlorite in the Wadhurst Clay Formation is suggestive of weathering under alkaline conditions in a cold and arid climate (Fig. 10), where leaching is limited. A resurgence of kaolinite in the Tunbridge Wells Sand Formation suggests warm and moist climatic conditions, similar to that which prevailed during the deposition of the Ashdown Formation. However, dominance of chlorite/illite at certain locations suggests localised cold and dry conditions. A spike in illite in the Weald Clay Formation indicates a return to cold and semi-arid/arid climatic conditions similar to when the Wadhurst Clay Formation was deposited.

The kaolinite to illite + chlorite (K/[I + C]) ratios have been used to identify changes in humidity (Hesselbo et al., 2009; Bougeault et al., 2017; Zuo et al., 2019). The K/[I + C] trend of the Wealden rocks shows fluctuations in humidity levels, although an overall decrease in humidity is noticeable (Fig. 10). This suggests that humidity levels during deposition in the Weald Basin generally tended towards being arid, with peak humidity levels being recorded during the deposition of the lower and uppermost parts of the Ashdown and Tunbridge Wells Sand formations respectively.

5.3. *Origin of detrital chlorite in the English Wealden ironstones*

Previous studies have identified kaolinite and illite as the most abundant clay mineral phases in the Hastings Beds and Weald Clay Formation (Sladen and Batten, 1984; Sladen, 1987; Akinlotan, 2017a, 2018) which is consistent with the data presented in this study (Tables 1 and 2). This study also reveals, however, that chlorite is also an important phase (Figs. 8 and 9), particularly in the ironstones where it is typically the dominant clay mineral (Table 2). Chlorite was present in the source region of the Weald Basin sediments (Sladen and Batten, 1984), and its presence at the point of deposition tends to indicate limited chemical weathering, due to low temperature or humidity or rapid transportation that resulted from steep gradients (Worden *et al.*, 2020). In the Weald Basin, the temperature was considered to be relatively stable during the Early Cretaceous (Smith *et al.*, 1973; Sladen and Batten, 1984), but notable variation in humidity and elevation of the source region has been identified (Sladen and Batten, 1984). Uplift in the source region led to increased annual rainfall rates, which resulted in soil and weathering profiles becoming more acidic and leached, leading to the increased formation of kaolinite. Conversely, with decreased elevation in the source region, the subsequent reduction in annual rainfall led to soils becoming less acidic, reducing kaolinite production and increasing illite formation (Sladen and Batten, 1984). Since there was an increased weathering intensity in the Weald Basin and the temperature remained stable during the Early Cretaceous period, the mostly possible explanation for the reduced weathering intensity of the source region required to increase deposition of detrital chlorite is a reduction in humidity, potentially from arid or semi-arid conditions (Fig. 10). Such conditions may have been generated from increased down-faulting in the source region lowering the elevation (Sladen and Batten, 1984).

Limited weathering in the source region not only leads to increased deposition of chlorite, but also of primary minerals: e.g. pyroxenes, garnets, amphiboles, etc. (Akinlotan *et al.*, 2021a) that are more readily broken down in high weathering environments (Worden *et al.*, 2020). Therefore, if the increased detrital chlorite observed in the ironstones is the result of reduced weathering in the source region, then such minerals should be more abundant too. QEMSCAN® datasets reveal that this is indeed the case, with ironstones having increased concentrations of, for example, garnet, pyroxene, amphibole and tourmaline (0.7%–2.9% of detrital components, rising to up to 47% if apatite is included), compared with the chlorite-poor mudstones and sandstones (typically <0.3% of detrital components) of the Hastings Beds and Weald Clay Formation. This supports the interpretation that the increased detrital chlorite compositions observed are the result of reduced weathering conditions in the source region, potentially due to a reduction in rainfall/humidity (Fig. 10).

5.4. *Clay mineral transformation*

5.4.1. *Kaolinization*

Although the majority of the Wealden clays are detrital in origin as described above (Sladen and Batten, 1984), there are strong indications to suggest that some of the detrital clays have been subjected to post-depositional and diagenetic transformation. The controls on clay transformation in the Weald Basin are described here. In normal circumstances, there are three main types of authigenic kaolinite that form in sandstones during eodiagenesis: Blocky kaolinite, vermiform kaolinite, and kaolinite replacing detrital mica (Lanson *et al.*, 2002). These types of kaolinite have been identified in the Hastings Beds sandstones (Fig. 11). In this current study, kaolinite formation from replacement of detrital mica is clear as kaolinite observed growing within the expanded mica flakes. Vermiform and blocky kaolinite crystallise at the expense of feldspars (Lanson *et al.*, 2002; Worden and Morad, 2003):

$$2KAlSi_3O_3[Feldspar]+2H^++9H_2O \rightarrow Al_2Si_2O_5(OH)_4[Kaolinite]+4H_4SiO_4+2K^+ \quad (1)$$

The distinct saw-like outlines of crystal plates of mica-replacive kaolinite (Fig. 11A) suggest variable effects of dissolution and alteration from a possible micaceous precursor source (Lanson *et al.*, 2002; Worden and Morad, 2003). Feldspar dissolution and subsequent kaolinite precipitation typically occur at shallow burial depths in warm and humid climate conditions from flushing of meteoric fluids during early diagenesis, and an open system is required to form substantial quantities of kaolinite (Lanson *et al.*, 2002; Worden and Morad, 2003). The distinct book- or worm-like (vermicular) habit of kaolinite (Fig. 11F), suggests that it was formed in a near-surface eogenetic/telogenetic environment. Thus, the presence of authigenic kaolinite in the Hastings Beds is consistent with the widely held interpretation that they formed in a subtropical to warm temperate environment (Sladen and Batten, 1984; Allen, 1998; Akinlotan, 2017a, 2018). The distinct association of blocky kaolinite with euhedral quartz overgrowth and authigenic illite (Fig. 11B, D) suggests that it was formed in a higher temperature-mesogenetic environment deeper than those of the mica-replacive and vermiform types.

Additionally, grain dissolution and kaolinization can result from CO_2-rich or organic acid-rich fluids from shales and coal beds adjacent to sandstones (Lanson *et al.*, 2002). Organic-rich shale interbeds are present within the Hastings Beds (Lake and Shephard-Thorn, 1987; Akinlotan, 2015, 2018), and so CO_2-rich or organic acid-rich fluids also contributed to the formation of authigenic kaolinite in the Hastings Beds sandstones. Given that the environmental and fluid chemistry conditions required for kaolinization were met in the Weald Basin, a higher abundance of diagenetic kaolinite might be expected. However, the dissolution of feldspar is an important precursor to provide the necessary components for kaolinite precipitation (Equation (1)). Thus, the low feldspar concentration (average 1.5%) within the Hastings Beds sandstones possibly played a key role in limiting the availability of kaolinite building blocks, resulting in the low authigenic kaolinite abundance observed.

5.4.2. *Illitization*

As burial depth and temperature increase into mesodiagenesis phase, kaolinite is transformed into the dickite polymorph/polytype of kaolin. This replacement typically occurs around 2500—5000 m, beginning with the transformation of ordered kaolinite into disordered dickite (Beaufort *et al.*, 1998; Lanson *et al.*, 2002). In this current study, XRD only identifies kaolinite in the Hastings Beds sandstones, but not dickite (Fig. 11). This suggests that the Hastings Beds have only undergone shallow diagenesis with a burial depth of ~2500 m.

Authigenic illite, however, typically forms during mesodiagenesis at depths of >3500 m with temperature of >120 °C, from illitization of kaolinite via the reaction (Lanson *et al.*, 2002; Worden and Morad, 2003):

$$\mathrm{KAlSi_3O_8[K\text{-}feldspar]+Al_2Si_2O_5(OH)_4[Kaolinite]} \rightarrow \mathrm{KAl_3Si_3O_{10}(OH)_2[Illite]+2SiO_{2(aq)}[Quartz]+H_2O} \quad (2)$$

Under these same conditions kaolinite undergoes a transition to dickite (Beaufort *et al.*, 1998; Lanson *et al.*, 2002), yet no dickite has been identified in any Hastings Beds sandstones, including in those containing illite. Alternatively, illite can form in temperature conditions as low as 70 °C (Worden and Morad, 2003) if the kinetic barrier to illite crystallisation is overcome. This is achieved through a high K^+/H^+ activity ratio, which requires a fluid oversaturated with respect to illite and K-feldspar. These conditions cannot occur in a closed system where K-feldspar solubility controls the activation energy; instead, it requires an external source of K^+ or increase in pH to reach the critical K^+/H^+ activity ratio. If this critical K^+/H^+ activity ratio is reached, illitization occurs through this reaction (Berger *et al.*, 1995; Lanson *et al.*, 2002):

$$\mathrm{3Al_2Si_2O_5(OH)_4[Kaolinite]+2K^+} \rightarrow \mathrm{2KAl_3Si_3O_{10}(OH)_2[Illite]+2H^++3H_2O} \quad (3)$$

A high K^+/H^+ activity ratio may be induced by either a tectonic event such as invasion of K^+-rich liquids, or by gradual removal of initially acidic pore-fluids by reactions of minerals (e.g. K-feldspar and carbonate dissolution) (Lanson *et al.*, 2002).

The authigenic illite observed in the Hastings Beds sandstones possibly formed from the reaction (3), allowing illitization to occur under shallower burial depths before kaolinite re-precipitates as dickite. This would also account for the limited occurrence of illite, because if a burial depth was reached that allowed illitization to occur through reaction (2), then illitization of kaolinite should be consistent/prevalent throughout the Hastings Beds sandstones. However, as authigenic illite is only observed in one location, this suggests that the critical K^+/H^+ activity ratio was only achieved locally within the Ardingly Sandstone Member and not on a basin-wide scale.

In the Rotliegendes sandstones, offshore Netherlands, Lanson *et al.* (1996) observed that the texture of authigenic illite is a function of temperature/depth. The shallowest illite occurs at 3000 m burial depth where crystals are elongated and filamentous, with a maximum width of 0.5 μm. As burial depth increases, illite crystals display more rigid lath morphology, with isometric pseudo-hexagonal-shape crystals also occurring below depth of 4000 m. This is accompanied by a concomitant increase in maximum crystal thickness up to 5 μm (Lanson *et al.*, 2002). The illite observed in the Ardingly Sandstone Member has a thin (<0.5 μm) filamentous appearance similar to the shallowest illite in the Rotliegendes sandstones (Fig. 11D). This suggests that the illite in the Ardingly Sandstone Member formed at the shallow end of illite formation depths.

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Collectively, the absence of dickite in the presence of kaolinite, the localised illitization under reduced burial depths due to reaching a critical K^+/H^+ activity ratio, and illite morphology suggest that the Hastings Beds reached an estimated maximum burial depth of ca. 2500–3000 m which is at least ca. 1000 m deeper than previously suggested (Sladen and Batten, 1984). Authigenesis of quartz cements is considered to occur at temperatures above 70 °C (Worden and Morad, 2000; Bjørlykke, 2014), which is compatible with the low end of illite formation conditions.

Authigenic illite generally develops by three mechanisms involving the illitization of kaolinite, the replacement of K-feldspar, and the transformation of mixed illite–smectite clay (Worden and Burley, 2003). Of these three processes, the authigenic illite in this study appears to be formed primarily from the illitization of kaolinite as indicated by the textural relationships between illite and kaolinite (Fig. 11). This diagenetic alteration seems to be controlled by the presence and amount of K-feldspar, burial depths (>2 km) and temperatures in the range of 120–140 °C (Bjørlykke and Aagaard, 1992; Worden and Morad, 2003; Bjørlykke, 2014).

5.5. Possible control on clay transformation: Weald Clay Formation

Given the presence of authigenic clays in the Hastings Beds (Fig. 11), and that the environmental and diagenetic conditions appear to have been conducive to authigenic clay formation (section 5.4 and 5.5), the question arises as to why no authigenic clays are present in the mudstones of the Weald Clay Formation. Possible control is briefly explored here. In the porous arkosic sandstones of Rio de Peixe Basin, Brazil (Maciel *et al.*, 2018), authigenic clay minerals (smectite, illite and kaolinite) formed during shallow diagenetic processes involving meteoric fluids. It was observed, however, that clay minerals are absent from, or present in significantly reduced quantities in the sandstones within fault zones. The faulting reduced the porosity and permeability of the sandstones in the fault zones by the destruction of space needed for formation and by the provision of tight water-resistant fabrics that inhibit fluid flow, thus hindering authigenic clay formation (Maciel *et al.*, 2018). A review of the clay mineral diagenesis in interbedded sandstones and mudstones of the Vienna Basin, Austria, found that the mudstones have significantly lower quantities of authigenic clay minerals than the sandstones (Gier *et al.*, 2018). This difference was attributed to the lower permeability of the mudstones that hindered fluid flow and reduced the rate of alteration of feldspar in the mudstones compared to the sandstones (Gier *et al.*, 2018). Similarly, when authigenic minerals in interbedded sandstones, siltstones, mudstones and shales in the Hartford Basin, USA were compared (Ahmed, 2007), it was revealed that the finer-grained deposits (i.e., mudstones and shales) contained significantly lower quantities of all authigenic phases (including clay minerals), due to the lower permeability of these rocks. While no quantitative porosity or permeability data exists for the Weald Clay Formation mudstones, SEM can provide qualitative analysis of the pore networks. The Weald Clay Formation mudstones contain very tight pore systems devoid of macropores, and contain limited, poorly connected micropores, which form ineffective pore systems (Fig. 12A and B) in contrast to porous Hastings Beds sandstones (Fig. 12C and D). Thus, we suggest that when compared to the Hastings Beds, the limited porosity and permeability of the Weald Clay Formation mudstones coupled with the low feldspar content, may have inhibited the formation of authigenic clays. This may have occurred by hindering flow of meteoric fluids, limiting feldspar alteration, and limiting growth space, which are very essential for the development of diagenesis of clay minerals.

5.6. The Early Cretaceous climatic evolution in Southeast England

The clay minerals described here show that during the deposition time of the Ashdown and Tunbridge Wells Sand formations, climatic conditions were warm and moist (Fig. 10). The climate was characterized by temperatures of up to 25 °C and spells of rainfall, ranging between 1000 mm and 1500 mm per annum (Allen, 1998) which aided intense chemical weathering of the source areas. The high rainfall pattern encouraged river discharge into the basin after leaching the severely weathered profiles, thus creating an acidic environment that favoured kaolinite concentration. Depositional energy was relatively high, leading to the formation of sandy facies. The warm and humid climate associated with the Ashdown and Tunbridge Wells Sand formations is suggested to be regional but not global as the rainfall patterns which were also recorded in other parts of Northwest Europe, were not reported in other places where climatic condition was arid (Sladen, 1983). The high rainfall pattern is attributed to a relatively higher elevation of the source massifs, indicating an upfaulting of the surrounding massifs. The warm and humid climate with high precipitation is also suggested to have favoured rich and diverse vegetation over the source areas and the basin. Palaeosalinity within the Weald Basin during

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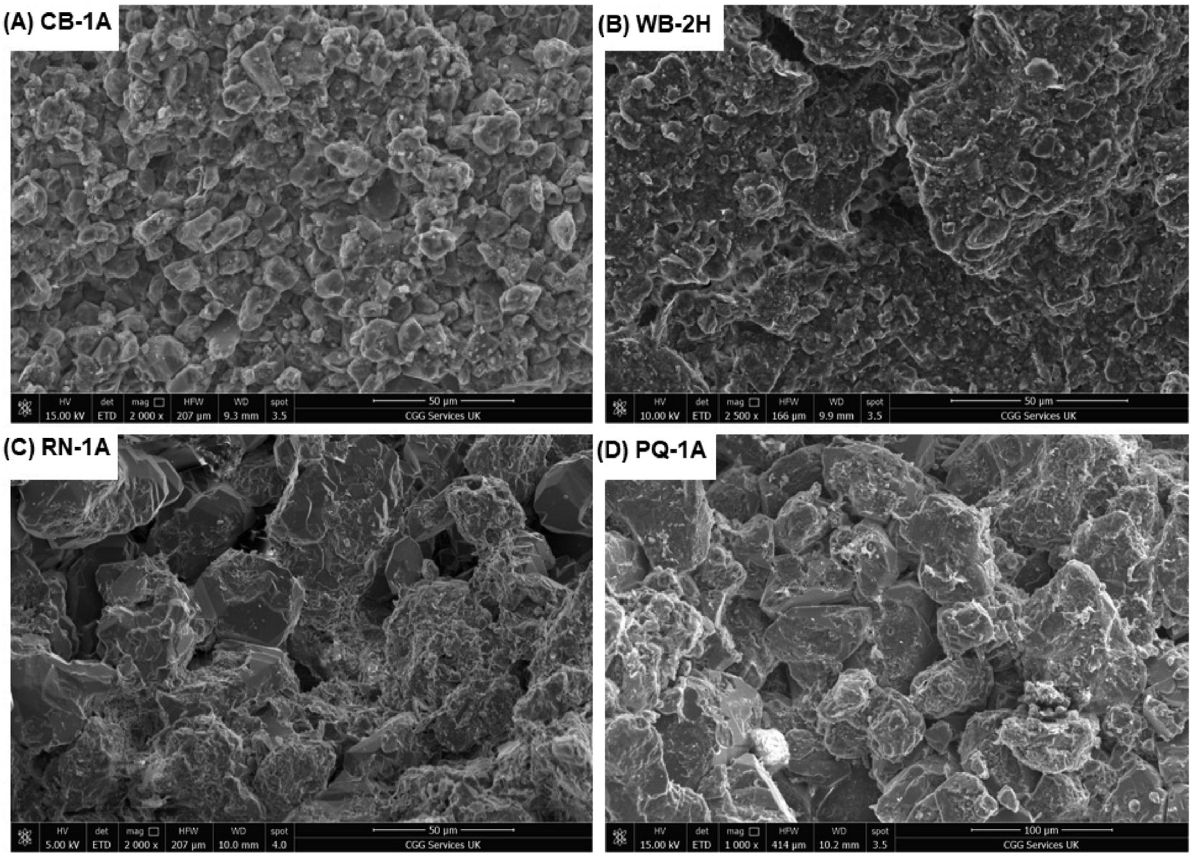

Fig. 12 SEM images providing an overview of the pore systems of representative Weald Clay Formation mudstones at **A)** Clock House Brickworks (CB) and **B)** Warnham Brickworks (WH), showing the tight, ineffective nature of the pore systems. SEM images of representative Hastings Beds sandstones at **C)** Rock-A-Nore (RN) and **D)** Philpots Quarry (PQ) are shown for comparison, displaying their more open, macroporous pore systems.

the deposition time of the Ashdown and Tunbridge Wells Sand formations appeared to be low due to high precipitation (Sladen, 1983).

The deposition of Wadhurst Clay and Weald Clay formations marks periods of aridity (Fig. 10) and relatively low rainfall. Less precipitation means less chemical weathering and minimal leaching of the source areas, causing the environment to be less acidic and encouraging concentrations of illite and chlorite. The reduced rainfall led to lesser run-off and lower energy of the depositing medium which favoured the deposition of mudstones in quiet lacustrine waters. The dry conditions and low precipitation are also suggested to have increased salinity levels in the basin during the deposition of the clay formations. The arid climatic conditions are suggested to be restricted to north-western Europe as similar trends have not been reported in other parts of Europe for the same time interval (Sladen, 1983). This means that the source areas were downfaulted to relatively lower elevations compared to the time when the Ashdown and Tunbridge Wells Sand formations were deposited. On a wider scale, palaeoenvironmental conditions during the deposition of the Hastings Beds was characterized by warmer climatic conditions, higher precipitation, and lower salinity levels, compared to that of the Weald Clay Formation (Fig. 10).

The interpretations based on clay minerals presented in this study correlate with interpretations based on other proxies such as sedimentology (Allen, 1981, 1989; Stewart, 1981; Lake and Shephard-Thorn, 1987), micropalaeontology (Anderson, 1975), palynology (Batten, 1982; Batten *et al.*, 1996), elemental geochemistry (Akinlotan, 2017b), siderite geochemistry (Taylor, 1992; Akinlotan, 2019), heavy minerals (Akinlotan and Rogers, 2021) and petrography (Akinlotan *et al.*, 2021b).

6. Wider implications

This study has shed more light on the formation of authigenic clay minerals in sedimentary basins, especially ones that are considered to have experienced shallow burial. The understanding of a complex burial/diagenetic history as it affects clay mineral transformation in the Weald Basin would also play a useful role in understanding basin evolution and clay diagenesis in other sedimentary basins. Interpreting palaeoenvironmental conditions of sedimentary basins (provenance, tectonics, palaeoclimate, palaeosalinity, palaeoweathering, etc) is usually done using detrital clay minerals. This study has demonstrated the importance of investigating both detrital and authigenic clay mineral assemblages when assessing the palaeoenvironments and palaeoclimate of both the source regions and site of deposition, to gain a more complete understanding of source and depositional systems. This is especially important if other data, such as palynology and geochemistry are unavailable.

The use of QEMSCAN® technique to supplement XRD and optical microscopy datasets is a significant importance of this study which can be applied to other basins when studying clay minerals. The high precision of QEMSCAN® technique resulted in high quality quantitative clay mineral data which allowed for more precise interpretations. For instance, optical microscopy revealed the presence of a few visible and large-sized clay minerals, but the technique was however limited due to the fine-grained size of most of the clay minerals. SEM method helped to study the clay minerals at a higher resolution. Suspected clay minerals which were subjected to EDS showed elemental compositions similar to kaolinite, illite and chlorite. However, due to the ability to analyse only a single portion of a whole sample at a time, SEM/EDS did not give a comprehensive result of the clay mineralogy of the selected samples. Although further analysis using XRD method confirmed the presence of kaolinite, illite and chlorite, the quantities in which these clay minerals occurred remained vague. It was the introduction of the QEMSCAN® technique that solved this problem by quantifying the clay minerals and revealing the presence of two more clay minerals, smectite and glauconite. This reinforces the importance of using of relevant multiple methods to facilitate a robust and balanced understanding of clay mineral formation and diagenetic transformation in sedimentary basins.

Understanding the formation of authigenic clays can also reveal insights into the factors that control their abundance (and lack thereof) and distribution (feldspar content, and porosity and permeability), as shown by this study and this can be effectively applied when studying other sedimentary basins. Authigenic clay mineral growth controls reservoir quality in hydrocarbon systems, so understanding the processes that control authigenic clay mineral growth can be vital to understanding reservoir quality and hydrocarbon distribution. Finally, clay minerals would be extremely useful either as complementary or alternative data to field and fossil datasets when investigating palaeoenvironmental conditions of sedimentary rocks.

7. Conclusions

XRD, SEM, EDS, optical microscopy and QEMSCAN® analyses show that most clay minerals in the Hastings Beds are detrital in origin and all clay minerals within the Weald Clay Formation appear to be detrital in origin. This multi-proxy study shows that kaolinite, illite, and to a lesser extent chlorite, are the most common detrital clay minerals present, with chlorite being more abundant than previously recognized. Chlorite abundance is particularly high in ironstone samples. The kaolinite enrichment in the Ashdown and Tunbridge Wells Sand formations indicates a warm and humid climate during their deposition time with high precipitation that encouraged chemical weathering and leaching. The high concentration of illite within the Wadhurst Clay and Weald Clay formations indicates colder and more arid conditions with lesser rainfall, lesser leaching and more of a physical weathering process. The lower rainfall favoured a quiet energy lacustrine environment in which the clay formations were deposited. The abundance of chlorite is thought to result from reduced weathering intensity in the source region during periods of down-faulting. This also led to increased deposition of Fe-silicate minerals from the source region, such as pyroxene and garnet. These interpretations indicate lower salinity levels and luxuriant vegetation types during the deposition time of Ashdown and Tunbridge Wells Sand formations, which are in contrast to the higher salinity levels and xeric vegetation types that existed during the deposition time of the Wadhurst Clay and Weald Clay formations.

Three forms of authigenic kaolinite were observed within the Hastings Beds: Mica-replacive kaolinite, vermiform kaolinite, and blocky kaolinite, although vermiform and mica-replacive kaolinite are the most common authigenic clay minerals. The presence of authigenic kaolinite is consistent with the prevailing model that the Weald Basin sediments were deposited in humid, subtropical to warm temperate climates. The absence of dickite and the fibrous morphology of the illite provide an estimated maximum burial depth of ca. 2500—3000 m for the Hastings Beds, which is ca. 1000 m deeper than inferred from previous studies. The absence of authigenic clays in the Weald Clay

Formation is suggested to be the result of restricted porosity and permeability of the mudstones.

This study is significant in two ways: 1) reinforcing the using of multi-proxy methods to facilitate a robust and balanced understanding of clay mineral formation and diagenetic transformation in sedimentary basins; 2) reiterating the investigation of both detrital and authigenic clay mineral assemblages when assessing the palaeoenvironments and palaeoclimate of both the source regions and deposition sites, to gain a broad understanding of source and depositional systems.

Authors' contributions

All the authors have actively participated in the preparation of this manuscript.

All authors have read and approved the final manuscript.

Availability of data and materials

Supplementary data are available as Appendix 1. The corresponding author can be contacted for queries or clarifications.

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Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary data

Appendix 1: Clay mineral data from QEMSCAN® analysis.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jop.2022.04.002.

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