Decoding the origins and sources of clay minerals in the Upper Cretaceous Tununk Shale of south-central Utah: Implications for the pursuit of climate and burial histories

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Abstract
Clay minerals in fine-grained marine sedimentary successions are most commonly considered to be detrital in origin and have been used extensively by geologists as indicators of palaeoclimate conditions in the hinterland. Most of these previous studies, however, were not designed to address in depth the potential effects of mixing clay minerals from multiple sources and the formation of authigenic clay minerals during early diagenesis on the ultimately observed clay mineral assemblages of fine-grained marine sedimentary successions. Herein, clay minerals in shales and bentonites of the Tununk Shale Member in south-central Utah were examined through integrated X-ray diffraction and petrographic (scanning electron microscopy) analysis, to evaluate the various origins of clay minerals in this offshore mudstone succession. Clays in Tununk Bentonites contain dominantly smectite (>80%) and a minor amount of kaolinite. Clays in Tununk shale samples consist dominantly of mixed-layer illite/smectite with up to 45% illite-like layers, small amounts of kaolinite and mica, and in places trace amounts of chlorite. Clay minerals in Tununk shale samples occur in the following three forms: (a) in clay-dominated aggregates (i.e. smectite-dominated altered volcanic rock fragments and illite/smectite-dominated shale lithics); (b) in the fine-grained matrix (mostly illite/smectite and minor amounts of mica and kaolinite); and (c) in intergranular and intragranular pore spaces (authigenic smectite, kaolinite and chlorite). Possible sources for the mixed-layer illite/smectite in shales include (a) erosion of older smectite-bearing mudstone successions and (b) weathering of volcanic rocks or volcanic debris that had been deposited on land. Most of the kaolinite and chlorite in the Tununk Shale were precipitated as pore-filling cements, rather than having a terrigenous source (as weathering products). A comprehensive understanding of the multiple origins of clay minerals (e.g. terrigenous input, volcanic input, recycled sediments and diagenesis) in marine mudstone successions is critical when attempting to use clay mineral data for reconstructions of palaeoclimate and burial and thermal histories.
1 | INTRODUCTION

Clay minerals dominate the clay-sized fraction (< 4 µm) of many fine-grained sedimentary (mudstone/shale) successions (Chamley, 1989; Velde, 1995; Potter et al., 2005; Lazar et al., 2015). Particularly in marine strata, clay mineral assemblages have been widely used as palaeoclimate proxies (Singer, 1980, 1984; Curtis, 1990; Robert and Kennett, 1997; Tamburini et al., 2003; Colin et al., 2014; Wendler et al., 2016). This is because clay minerals in marine sediments were generally assumed to be inherited from landmasses via weathering of bedrock, which is ultimately controlled by climate (Biscaye, 1965; Chamley, 1989; Hallam et al., 2000). That clay minerals can transform of specific clay types (e.g. smectite to illite) during deep burial has been studied extensively (Burst, 1957; Nadeau and Reynolds, 1981a; Sucha et al., 1993; Hillier et al., 1995; Anjos et al., 2003). Early diagenetic processes in mudstones are largely driven by redox reactions, in which sedimentary organic matter is metabolized by microbes (Froelich et al., 1979; Wilson et al., 1985; Canfield et al., 1993; Chester, 2009). Key products of these reactions include bicarbonate ions, reduced manganese and iron, and sulphide, all of which can be incorporated into early diagenetic minerals such as calcite, pyrite, siderite and dolomite (Berner, 1984; Raiswell, 1988; Macaulay et al., 1993; Raiswell and Fisher, 2000; Taylor et al., 2000). That clay minerals can also form during this shallow burial stage has been known for some time (Odin, 1990; Mackenzie and Kump, 1995; Michalopolous and Aller, 1995) but still is not considered of significant importance in many published studies of mudstones. Over the past decade, an increasing number of case studies have suggested that significant amounts of kaolinite in some ancient mudstone successions is of early diagenetic origin (Aplin and Macquaker, 2012; Macquaker et al., 2014; Taylor and Macquaker, 2014; Schieber et al., 2016), instead of being introduced to sedimentary basins by rivers that had intensely weathered soils in their catchment areas (Thiry, 2000; Tamburini et al., 2005; Wendler et al., 2016).

Presumed Milankovitch-scale climate cycles during the Late Cretaceous have been widely reported (Barron et al., 1985; Sageman et al., 1997; Floegel et al., 2005; Eldrett et al., 2015; Wendler et al., 2016). Many of these studies focusing on Late Cretaceous strata in the Western Interior Seaway (WIS) of North America interpreted variations in the relative amount of clay minerals of presumed detrital origin as high-frequency wet versus dry cycles (Pratt, 1984; Eicher and Diner, 1989; Sethi and Leithold, 1994). In the pelagic deposits of the Bridge Creek Limestone Member that accumulated in the central area of the WIS (at Pueblo, Colorado; Figure 1A), discrete illite and mixed-layer illite/smectite (I/S) are considered of detrital and volcanic origin, respectively (Pratt, 1984; Eicher and Diner, 1989). However, analysis of the clay mineralogy in proximal time-equivalents of the Bridge Creek Limestone, the Tropic Shale and Tununk Shale in southern Utah along the western margin of the WIS (Figure 1A), shows a much more complex assemblage of presumed detrital clays, including kaolinite, discrete illite, chlorite, and mixed-layer I/S (Sethi and Leithold, 1994). The discrepancies in the presumed detrital clay mineral assemblages suggested by these studies highlight the inherent difficulties in resolving the complex origins of clay minerals in marine sedimentary successions. These issues severely limit our ability to untangle clay mineral signals that reflect climate changes from those...
due to changes in depositional environment, provenance and diagenesis.

Most previous palaeoclimate interpretations in mudstones were based on the clay mineral assemblages determined via X-ray diffraction (XRD) methods (Moore and Reynolds, 1997). Yet, without detailed petrographic (scanning electron microscope) examination of the morphology, texture and distribution of clay minerals in fine-grained sedimentary successions, the exact origin of clay minerals and the impact of early diagenesis cannot be evaluated with confidence. This study focusses on the Upper Cretaceous Tununk Shale Member of the Mancos Shale in south-central Utah because the presumption is that it should carry a clearer and stronger detrital clay signal due to its proximal setting within the WIS. The Tununk Shale in the study area has undergone negligible burial metamorphism (Nadeau and Reynolds, 1981a), making it an ideal natural laboratory for constraining detrital clay inputs to the WIS. Based on integrated X-ray diffraction and petrographic (scanning electron microscopy) analysis, clay minerals in the Tununk Shale can be traced to multiple sources (e.g. wind-borne volcanic ash, recycled sediments and diagenesis) in addition to continental weathering. It is therefore critical to obtain a comprehensive understanding of the sources and origins of clay minerals in marine mudstone successions before using clay mineral assemblages as proxies for palaeoclimate conditions and burial and thermal histories.
2 | GEOLOGICAL BACKGROUND

During the Late Jurassic and Cretaceous, the geology of western North America was largely controlled by the subduction of the Farallon plate beneath the North American plate (Livaccari, 1991; DeCelles, 2004). The compressive forces associated with plate convergence, combined with conductive heating initiated by subduction, led to crustal thickening in orogenic belts such as the Sevier fold-thrust belt (Livaccari, 1991; DeCelles, 2004). Subsidence due to flexural loading of the crust formed an asymmetric foreland basin, bounded to the west by the rising Sevier orogenic belt and to the east by the stable North American craton (Kauffman, 1977, 1985; Kauffman and Caldwell, 1993). During the Late Cretaceous, the foreland basin was subject to active thrusting, subsidence, uplift (forebulge uplift) and extensive volcanism (DeCelles, 2004; DeCelles and Coogan, 2006; Yonkee and Weil, 2015). A magmatic arc, comprising a belt of calc-alkaline intrusive and volcanic rocks, was located along the western margin of the North American continent (Figure 1A).

The combined effects of load-induced subsidence and eustatic highstand throughout the Late Cretaceous resulted in the development of an epicontinental seaway known as the WIS (Kauffman, 1977, 1985). During the Albian to Maastrichtian, the WIS was subject to five major transgressive-regressive marine cycles with durations of 2-10 Myr (Kauffman, 1985). The most extensive flooding of the seaway occurred during the so-called Greenhorn cycle, when more than one-third of North America was inundated. The Greenhorn cycle began in the late Albian and ended in the middle Turonian (Kauffman, 1985). During peak transgression in the early Turonian, the WIS extended from the Arctic Ocean to the present-day location of the Gulf of Mexico and was characterized by a broad shelf (Figure 1A). In response to sediments supplied from the unroofing of the Sevier orogenic belt and volcanic highlands to the west, the WIS was characterized by high siliciclastic input and high sedimentation rates along its western margin (Kauffman, 1977, 1985).

The Tununk Shale accumulated along the western margin of the WIS during the late Cenomanian to middle Turonian. In outcrop it consists mainly of dark grey calcareous to non-calcareous mudstones with numerous thin silt and sand-rich beds (Peterson et al., 1980; Zelt, 1985). A series of regionally traceable bentonite beds has been used to correlate the Tununk Shale in south-central Utah (Henry Mountains Region) with its lateral equivalent, the Tropic Shale in southern Utah (Kaiparowits Plateau) (Zelt, 1985; Leithold, 1994; Figure 1). Driven by the Greenhorn transgressive-regressive sea-level cycle, the offshore mudstones of the Tununk Shale overlie the coarser non-marine and paralic deposits of the Dakota Sandstone and grade upward into the shallow marine and deltaic strata of the Ferron Sandstone Member (Figure 1B). The palaeoshoreline during deposition of the Tununk Shale generally trended north-east–south-west (Figure 1A; Zelt, 1985; Leithold, 1994). Although the dominant direction of sediment progradation was eastwards, nearshore sediments deposited along the western margin of the WIS were deflected southwards by coastal currents generated by a large-scale counterclockwise gyre (Figure 1A; Barron, 1989; Ericksen and Slingerland, 1990; Slingerland and Keen, 1999; Li et al., 2015).

2.1 | Depositional framework of the Tununk Shale

Characteristics of sedimentary facies and depositional framework of the Tununk Shale are discussed in detail in a previous study (Li and Schieber, 2018a), and are only briefly summarized here. The Tununk Shale in the study area is interpreted as an offshore mud belt on a storm-dominated shelf and consists of four shale lithofacies associations. In detail these are: (a) carbonate-bearing (<30% carbonate), silty and sandy mudstone (CSSM); (b) silt-bearing, calcareous mudstone (SCM, >30% carbonate); (c) carbonate-bearing, silty mudstone to muddy siltstone (CMS); and (d) non-calcareous, silty and sandy mudstone (SSM) (Figure 2), based mainly on the relative proportion of carbonate particles derived from carbonate productivity versus siliciclastic particles of detrital origin. A series of bentonite beds are present throughout the Tununk Shale (Figures 1B and 2). Upsection, the relative amount of the carbonate component derived from carbonate productivity gradually increases from the CSSM to SCM, then decreases to the CMS, and was not observed in the SSM lithofacies association. Vertical variations in lithofacies association and sedimentary facies characteristics indicate that the depositional environment of the Tununk Shale shifted laterally from distal middle-shelf to outer-shelf
(CSSM to SCM), then from outer-shelf to inner-shelf environment (SCM to CMS, and to SSM) during the second-order Greenhorn sea-level cycle (Figure 2). Storm-induced, shore-parallel geostrophic flow and offshore-directed flows probably were the dominant processes that governed the transport and deposition of mud along and across a storm-dominated shelf (Li and Schieber, 2018a).

3 | METHODS

Samples (shales and bentonites) for this study were collected from three detailed stratigraphic sections of the Tununk Shale near Hanksville, Utah (Figures 1C and 2). By comparing the composition and characteristics of clay minerals of bentonites with those of shale samples, the sources (other than volcanic ash) of clay minerals in Tununk shale samples can be determined. Forty-two representative shale samples and five bentonite samples from all lithofacies associations throughout the Tununk Shale were selected to make polished thin sections and ion-milled samples (up to 12 mm in diameter) for subsequent scanning electron microscopy (SEM) analysis. Detailed petrographic examination was conducted to determine the overall composition, as well as the type, morphology, texture and distribution of clay minerals present in both shales and bentonites using an FEI Quanta 400 scanning electron microscope, operating at 15 kV and a working distance of 10 mm. Energy-dispersive X-ray spectroscopy (EDS) was used to aid determination of mineralogy.

Five samples of bentonites and 13 samples of shales (including those next to and farther away from bentonites) throughout measured section NCa (Figure 1C) were selected for determination of clay mineral assemblages in the <2 μm fractions by XRD analysis. The stratigraphic locations of these samples are indicated in Figure 2. Samples were crushed and dispersed in distilled water using an ultrasonic probe. For each sample, the <2 μm fraction was separated by gravity settling in water, concentrated by centrifugation and prepared for XRD analysis by sedimentation onto a quartz plate (‘zero-background’ plate). The XRD patterns were acquired using a Bruker D8 Advance X-ray diffractometer equipped with incident and diffracted-beam Soller slits, a Sol-X solid-state Si (Li) energy-dispersive detector and CuKα radiation (45 kV, 35 mA). The <2 μm fraction was scanned from 2 to 36° 2θ in the air-dried state and after saturation with ethylene glycol. Estimates of the percentage of illite-type layers (collapsed layers) in the mixed-layer I/S were made based on the peak positions of glycolated mixed-layer I/S from about 26° to 27° 2θ and from 15.4° to 17.7° 2θ, using the method described by Środoń (1980). The accuracy of the determination of the percentage of expandable layers was ±5% for most mixed-layer I/S samples (Środoń, 1980).

4 | CLAY MINERALOGY OF THE TUNUNK SHALE

4.1 | Clay minerals in bentonites

Bentonite beds in the Tununk Shale range from 1 to 40 cm in thickness (Figure 2). On polished slabs, some bentonite beds are slightly to moderately bioturbated (Figure 3A and B) and show navichnia traces (sediment-swimmer traces sensu Lobza and Schieber, 1999). On polished slabs, bentonites can be structureless or show a variety of sedimentary structures such as ripple-scale cross lamination, stacked normally graded beds, and erosional bases (Figure 4). The <2 μm fractions of bentonites consist almost exclusively of smectite or mixed-layer I/S with up to 10% illite-type layers (Figures 2 and 4). In a few bentonite samples, a small peak near 7.0 Å indicates the presence of a small amount of kaolinite (Figure 4).

Under the SEM, un-reworked ash-fall bentonites (those showing no features indicating reworking by bioturbation or bottom currents) in the Tununk Shale contain common silt to sand-sized phenocrysts, including biotite, plagioclase, potassic-feldspar and small amounts of apatite and quartz (Figure 5A and B). The clay mineral-dominated matrix of un-reworked bentonites consists exclusively of smectite, characterized by a highly crenulated and honeycombed morphology (Figure 5C). The small amount of kaolinite present in some bentonites is mostly associated with biotite (Figure 5B and D).

4.2 | Clay minerals in shales

The <2 μm fractions of shale samples show a more complex composition. The sharp peaks at 26.6° and 29.4° are due to the presence of micron-sized quartz and calcite (as coccolith debris), respectively (Figure 6). The relative intensity of the quartz and calcite peaks is consistent with different lithofacies associations having varying carbonate content in the Tununk Shale. The dominant type of clay mineral in the shale samples is mixed-layer I/S with varying proportions of illite-type layers. Compared with the bentonites, the <2 μm fractions of shales contain greater amounts of mica and kaolinite (Figure 6B), indicated by peaks at ~10.0 Å and 7.0 Å, respectively. The partial resolution of two peaks at ~7.0 Å and 3.5 Å in a few samples indicates the presence of trace amounts of chlorite (Figure 6B). The mixed-layer I/S in shales close to bentonites contains 5%-10% illite-type mixed layers (Figures 6A and 7). In shales far (>1 m) away from bentonite beds, the 001 peak of the mixed-layer I/S shows significant broadening (Figure 6B). In these shale samples, a distinctly higher amount (20%-45%) of illite-type layers is present in the mixed-layer I/S (Figures 2 and 6B).

Detailed SEM examinations indicate that clay minerals in shale samples are present in three forms: (a) as clay-dominated composite particles (or aggregates) (Figure 8); (b) in
the fine-grained matrix (Figure 9); and (c) in pore spaces (both intergranular and intragranular) (Figure 10). This paper uses the term 'fine-grained matrix' to refer to the fine-grained groundmass (clay mineral-dominated, with clay to fine-silt-sized particles) in shale samples of the Tununk Shale. Framework grains (medium-silt to sand-sized grains/aggregates) in shale samples usually have different compositions from their surrounding fine-grained matrix.

The clay-dominated composite particles comprise two types, namely altered volcanic rock fragments and shale lithics. Altered volcanic rock fragments were the most commonly observed type in the Tununk Shale (Figure 8A). Volcanic rock fragments contain dominantly smectite characterized by a highly crenulated morphology, the same as observed in bentonites (Figure 8B). Some volcanic rock fragments contain micron-sized (up to 15 µm) mineral grains such as potassium-feldspar, plagioclase, biotite, quartz, apatite and ilmenite, which are also common phenocrysts in bentonites (Figures 5 and 8B). The other important type of clay-dominated composite particles is shale lithics. Shale lithics contain silt-sized minerals (mostly quartz) scattered in a clay mineral-dominated matrix (Figure 8C,D). The clays in shale lithics are characterized by a more wavy-planar fabric (Figure 8D), compared with the highly crenulated morphology of

**FIGURE 3** Scanned and processed images of polished slabs showing the sedimentary and biogenic features in different bentonite beds in the Tununk Shale. A) Part of bentonite T3 is commonly bioturbated and shows navichnia traces (black arrows). B) Part of bentonite T4 is slightly to commonly bioturbated and shows navichnia traces (black arrows). The abundant black specks revealed on the surface are biotite. C) Part of bentonite T9 shows unidirectionally dipping foresets (dashed lines) under a rather symmetrical crest, indicating influence by storm-induced bottom currents during its deposition. The upper part of this sample is slightly bioturbated (black arrow). D) Part of bentonite T10 shows stacked normal grading (stacked triangles). Although bentonite T9 in C) show sedimentary features indicating reworking by bottom currents, the smectite in this bentonite does not contain any illite-type layers, similar to the smectite in T10 in D)
smectite in bentonites and volcanic rock fragments. Because shale lithics show more distinct potassium peaks in SEM-EDS analyses when compared to smectite in bentonites, the clays in shale lithics are identified as mixed-layer I/S. Shale lithics show resistance to compaction and have textural characteristics of fully compacted mudstones, such as preferred orientation of clays and scattered fine silt grains (Figure 8C).

Clays in the fine-grained matrix of the Tununk Shale consist dominantly of mixed-layer I/S, as suggested by XRD data and SEM images (Figures 6B and 9). Under the SEM, the clay mineral-dominated matrix in shales close to bentonites resembles more the pure smectite of the bentonites (Figure 7), probably a reflection of the comparatively small amounts of illite-type layers in the mixed-layer I/S (Figure 6A). Small amounts of kaolinite are locally present in the clay mineral-dominated matrix of shales (Figure 9D).

The intragranular pore space within foraminifera chambers in the CSSM and SCM lithofacies associations can be filled with smectite and kaolinite (Figure 10A and C), in addition to

**FIGURE 4** Representative X-ray diffraction patterns of the <2 μm fractions in bentonite samples. All are ethylene glycol solvated. See Figure 4 for stratigraphic locations of different bentonite beds. S: smectite, K: kaolinite

**FIGURE 5** Characteristic features of un-reworked bentonites (altered volcanic ash) in the Tununk Shale under the SEM. A) and B) Overview of the mineral composition and texture of the bentonite in the Tununk Shale (both are backscatter electron images). Phenocrysts present in the bentonite include common potassium-feldspar (K-spar), biotite (Bio), and small amounts of quartz (Q) and apatite (P). Most kaolinite (Kao) occurs along cleavages of biotite in the bentonite. The clay-dominated matrix is mostly smectite (Smec). C) A closer view showing the highly crenulated morphology of smectite in bentonites. D) A closer view showing kaolinite forming along the cleavages of biotite flakes (relatively lighter-coloured). All are backscatter electron images
commonly being filled with early diagenetic calcite (Figure 10A). Kaolinite in the CMS and SSM lithofacies associations more commonly fills the intergranular pore space of silty/sandy laminae (Figure 10E and F). Locally, small amounts of chlorite occur associated with kaolinite, filling particularly large intergranular pore spaces. Kaolinite can also be present along cleavages of biotite flakes in shales (Figures 9B and 10D), similar to what is observed in bentonites (Figure 5B and D).

5 | DISCUSSION

5.1 | Volcanic contributions to the Tununk Shale

Bentonites are the product of early diagenetic alteration to volcanic ash in marine environments (Fisher and Schmincke, 1984; Bohor and Triplehorn, 1993; De Ros et al., 1997). The original transport of these ashes to the marine environment was probably by wind and episodic (Slaughter and Hamil, 1970; Nadeau and Reynolds, 1981b). Most bentonite beds in the Tununk Shale show reworking by bioturbation or bottom currents (Figure 3). The navichnia traces observed in some bentonite samples imply that the incoming volcanic ash was mixed with muds that represent the background sedimentation (muds deposited by suspension settling or bottom currents) (Figure 3A and B). Ripple-scale cross lamination and basal erosional scours associated with some bentonite beds attest to bottom currents and relatively rapid sedimentation (Figure 3C). The stacked normally graded beds in bentonite T10 (Figure 3D) are interpreted to reflect multiple pulses of ash settling during a single volcanic event (Fisher and Schmincke, 1984), in the context of its lateral continuity and similar appearance across the study area.
The preserved stacked normally graded beds in bentonite T10 are suggestive of passive settling and rapid burial (Figure 3D; Bentley and Nittrouer, 2003; Wheatcroft and Drake, 2003), and imply that it has not been reworked by bottom currents or bioturbation. The absence of illite-type layers in the <2 μm fraction from this bentonite indicates that the smectite has not undergone post-burial transformation to mixed-layer I/S, consistent with the relatively low vitrinite reflectance ($R_o < 0.6\%$; Schamel, 2008) and inferred shallow burial history of the Tununk Shale in the study area (Nadeau and Reynolds, 1981a; Sethi and Leithold, 1994). Because of this, the combined presence of navichnia traces and mixed-layer I/S with only small amounts of illite-type interlayer suggested by the XRD analysis. The mixing of volcanic ash and background sediment (source of mixed-layered I/S) is indicated by the presence of coccolith debris (red arrows). Both B) and C) are backscatter electron images. Ca: calcite, Py: pyrite, I/S: mixed-layer I/S

FIGURE 7 Sedimentologic and petrographic facies characteristics of shale samples directly above bentonites. A) Scanned and processed images of the polished slab made of shale sample 2, which lies directly above bentonite T3 (see Figure 4 for stratigraphic locations). This sample is commonly disrupted by navichnia traces (black arrows). The lighter-coloured zones on the polished surface are probably due to mixing of volcanic ashes by benthos. B) An overview of the shale sample shown in A) under the SEM showing the mixture of volcanic ash (smectite-dominated matrix) and background sediments (foraminifera tests and fossil fragments). The potassium-feldspar (K-spar) and biotite (Bio) and related kaolinite (Kao) can be derived from reworking of bentonite T3, lying directly below this sample. C) A closer view of the fine-grained matrix in B). The clays very much resemble the smectite in un-reworked bentonites (high crenulated morphology), which is consistent with the mixed-layer I/S with only 10% illite-type interlayer suggested by the XRD analysis. The mixing of volcanic ash and background sediment (source of mixed-layered I/S) is indicated by the presence of coccolith debris (red arrows). Both B) and C) are backscatter electron images. Ca: calcite, Py: pyrite, I/S: mixed-layer I/S

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Bentonite beds in shale successions can be viewed as ‘internal standards’ for evaluating the degree of burial metamorphism in shales because their source is distinct from the shale they are found in. The smectite in un-reworked bentonites in the Tununk is characterized by a lack of illitization, indicating that the associated shales should as well have undergone negligible illitization after deposition (Schultz, 1978).
Therefore, the randomly interstratified I/S in shale samples must be detrital and existed prior to deposition in the Tununk system. Similarly, the relatively low amount of illite-type layers in the mixed-layer I/S from shales adjacent to bentonites is probably indicative of a volcanic ash – background sediment mixing effect that diminishes away from the ash bed. This interpretation is consistent with the bioturbated nature and the highly similar smectite-dominated texture of these shale samples (Figure 7).

5.2 Origins of other clay minerals in the Tununk Shale

Shales located at some distance (>1 m) above bentonite beds contain dominantly mixed-layer I/S with a distinctly higher proportion of illite-type layers than shale samples close to bentonites. This probably reflects the volumetric dominance of detrital-terrigenous matrix I/S over smectite-dominated altered volcanic rock fragments (Figure 9). The mixed-layer I/S in shales has multiple possible sources (Figure 11). It can for example be derived from various older shale/mudstone successions in the hinterland, an interpretation that is consistent with the presence of shale lithics in the Tununk Shale. Other possible sources for the mixed-layer I/S are soil profiles developed on older volcanic rocks and weathering of volcanic debris that fell on land areas and underwent repeated wetting and drying episodes (Hein and Scholl, 1978; Schultz, 1978; Eberl et al., 1987; Kemp et al., 2016).

In XRD patterns from the Tununk shale samples, the symmetrical peak at ~10 Å indicates the presence of mica instead of discrete illite in the <2 μm fractions (Środóń, 1981). The SEM analysis shows that biotite is the dominant type of mica in the Tununk Shale. Because biotite is a common phenocryst in bentonites (Figures 3B and 5B), the biotite in shales may be derived from resuspension and redeposition of volcanic ash in the marine environment as well as from terrigenous sources (also probably supplying some muscovite).
Most (>90%) kaolinite recognized under the SEM in the Tununk shale samples occurs as pore‐filling cements and therefore formed during early diagenesis before significant compaction (Figures 10 and 11). Possible sources of silica for kaolinite authigenesis in the Tununk Shale are the silicic present as dissolved (probably present as Si(OH)$_4$) and particulate (a wide variety of silicate and aluminosilicate minerals) forms in sea water (Chester, 2009), as well as alteration in volcanic ash particles. The transformation from volcanic glass to smectite is generally thought to occur during early diagenesis and can release silicon (Fisher and Schmincke, 1984). Due to the extremely low solubility of aluminium in sea water, aluminium is primarily present in particulate form in aluminosilicate minerals and hydrous aluminium oxides (Michalopoulos and Aller, 1995). The breakdown of organic carbon by bacterial sulphate reduction (indicated by the presence of pyrite) and methanogenesis is typically associated with high concentrations of organic acids in the sediment interstitial waters (Barcelona, 1980; Gunnarsson and Rönnow, 1982; Surdam et al., 1989; Manning et al., 1994; Macquaker et al., 2014). These acids can promote the dissolution of unstable aluminosilicates (e.g. biotite, feldspars, unstable clays and volcanic detritus), volcanic glass and hydrous aluminium oxides and thus result in localized availability of aluminium (Moncure et al., 1984; Surdam and Crossey, 1985; Acker and Bricker, 1992; Fein, 1994). Kaolinite can then precipitate from interstitial solutions in intergranular and intragranular pore spaces (Figure 10B,C,E), along the cleavages of biotite flakes (Figure 10D), and as replacement of unstable detrital grains (Figure 10F). In the Tununk Shale, the precipitation of kaolinite was not accompanied by significant carbonate dissolution (e.g. dissolution of foraminifera tests), suggesting that kaolinite authigenesis, rather than carbonate dissolution, was the main acid buffer mechanism in its sediment interstitial waters (Macquaker et al., 2014).

Only a small amount of chlorite is present as pore‐filling cements in relatively few shale samples. The precipitation of chlorite in the Tununk Shale probably was limited by the availability of iron and magnesium in interstitial waters. Because pyrite forms very early in diagenesis, the common presence of pyrite in the Tununk Shale (Figure 10B and D) indicates that reduced iron in interstitial waters was largely removed before favourable conditions for chlorite precipitation.
were reached. Furthermore, the authigenesis of smectite from alteration in volcanic glass or unstable volcanic debris, which occurred prior to kaolinite formation (Figure 10A through C), requires significant incorporation of magnesium (Fisher and Schmincke, 1984). It is therefore probable that chlorite was only precipitated at places where silicon, aluminium, iron and magnesium derived from dissolution of unstable volcanic materials (e.g. mafic minerals) were sufficiently retained at a local scale (Figure 10F; Anjos et al., 2003).

5.3 Implications for using clay mineral assemblages as proxies for palaeoclimate and burial history

Clay mineralogy has been widely used as a proxy for quantifying the intensity of continental weathering and reconstructing palaeoclimate (Singer, 1984; Dera et al., 2009; Kemp et al., 2016; Wendler et al., 2016). The most important underlying assumption for this approach is that the clay minerals in a given sedimentary succession are derived mainly from coeval soils and thus directly reflect climate parameters (weathering conditions) in the hinterland (Biscaye, 1965; Singer, 1984; Chamley, 1989; Thiry, 2000; Junttila et al., 2005; Wendler et al., 2016). A previous study of clay minerals in the Tropic Shale and Tununk Shale in southern Utah (south of the study area) reported that detrital clays include kaolinite, discrete illite (mica), chlorite and mixed-layer I/S (Sethi and Leithold, 1994). Results from this study, however, based on comparisons between clay mineral assemblages of bentonites and shales, suggest that the dominant land-derived detrital clay mineral in the Tununk Shale is mixer-layer I/S, whereas kaolinite and chlorite are mostly authigenic and mica (dominantly biotite) is mainly associated with volcanic ash falls. More importantly, only the mixed-layer I/S derived from weathering of volcanic rocks and volcanic ash deposited on land may have a bearing on weathering conditions in the hinterland.

**Figure 10** A) A foraminifera test in the SCM lithofacies is filled with authigenic smectite and calcite. B) A foraminifera test in the SCM lithofacies is filled with authigenic smectite, kaolinite and pyrite. C) Closer view of the dashed area in C). Authigenic smectite formed prior to authigenic kaolinite. D) Closer view in the CSM lithofacies showing authigenic kaolinite and pyrite forming along the cleavages within biotite. E) Presence of common authigenic kaolinite (within red-dashed lines) and a minor amount of chlorite in the pore space within a silty/sandy lamina in the SSM lithofacies. F) Closer view of the area within the red square in E). Note booklet-like kaolinite and plate-like chlorite (white arrows). This pore is larger than others surrounding it, and both the pore and its kaolinite and chlorite fill may be the result of dissolution of a feldspar grain or a volcanic detritus. Ca: calcite; Smec: smectite; Kao: kaolinite; Bio: biotite; Py: pyrite; K-spars: potassium-feldspar; Plag: plagioclase; Q: quartz.
In contrast, the mixed-layer I/S derived from recycled mudstones that experienced burial metamorphism, signified by shale lithics seen in SEM petrography, has no relation to the weathering regime at the time of deposition. In the context of palaeogeographic reconstructions (Figure 1), shale lithics in the Tununk Shale have two possible sources, including disintegration of exposed older mudstone successions in the hinterland and the unconformity located updrift of the study area (then dispersed by southward-directed longshore currents). A significant amount of kaolinite (and trace amount of chlorite) was precipitated as early diagenetic cements in the pore spaces, instead of being introduced as weathered terrestrial detritus via river run-off.

As discussed above, micas (dominantly biotite) in the Tununk Shale can be derived from either wind-borne volcanic ash or terrigenous input. Significant amounts of kaolinite and chlorite were directly precipitated from pore waters as early diagenetic cements, instead of being introduced as weathered terrestrial detritus via river run-off. It is therefore difficult, if not impossible, to determine the detrital contribution of mixed-layer I/S, mica, kaolinite, and chlorite to the Tununk Shale and then to use relative proportions of these clays to infer palaeoclimate in the hinterland. The mixing of sediments derived from different sources, as well as early diagenetic contributions and overprint, significantly obscures the original detrital clay mineral assemblages of marine mudstone fragments cannot possibly be related to palaeoclimate conditions that prevailed during the deposition of the Tununk Shale.

FIGURE 11 Schematic drawing of the dispersal processes and origins of clay minerals in the Tununk system. The smectite in shales is largely derived from alteration in volcanic ash in the marine environment during early diagenesis. The mixed-layer I/S in shales can be derived from weathering of volcanic debris that had been deposited in the hinterland and from older mudstones/shales (as suggested by the presence of shale lithics). Shale lithics in the Tununk Shale have two possible sources, including disintegration of exposed older mudstone successions in the hinterland and the unconformity located updrift of the study area (then dispersed by southward-directed longshore currents). A significant amount of kaolinite (and trace amount of chlorite) was precipitated as early diagenetic cements in the pore spaces, instead of being introduced as weathered terrestrial detritus via river run-off.
mudstone successions. When attempting to interpret the palaeoclimate based on clay mineral assemblages in marine mudstone successions, one needs to focus only on clay minerals derived from coeval soils, factoring out clays derived from volcanic input, recycled sediments and early diagenesis.

In the pelagic deposits of the Bridge Creek Limestone Member of central Colorado, time-equivalent to the Tununk Shale and deposited in the central part of the WIS, discrete illite and mixed-layer I/S were considered as detrital and volcanic in origin, respectively, because bentonites in the Bridge Creek Limestone also contain mixed-layer I/S (Pratt, 1984). The mismatch between the detrital clay mineral assemblages in the distal Bridge Creek Limestone (discrete illite suggested by Pratt, 1984; Eicher and Diner, 1989) and the proximal Tununk Shale (abundant detrital-terrigenous matrix I/S) indicates that clay mineral assemblages in the Bridge Creek Limestone were probably modified after deposition. For example, the mixed-layer I/S in the commonly bioturbated bentonites from the Bridge Creek Limestone (Elder, 1985) could be due to mixture of volcanic ash with background sediments, similar to what was observed in the Tununk Shale. However, alteration during burial metamorphism may be a more plausible explanation for the observed mixed-layer I/S because analysis of the clay mineralogy in the Smoky Hill Shale Member of the Niobrara Formation (stratigraphically higher than the Bridge Creek Limestone) in the same area (Pueblo, Colorado) indicate burial temperatures between 70 and 100°C (Pollastro and Martinze, 1985). Being stratigraphically lower, the Bridge Creek Limestone should have experienced even higher burial temperatures, making it probable that the observed mixed-layer I/S compositions are not a detrital signature and thus of dubious value as a palaeoclimate proxy.

The progressive conversion of smectite into illite with increasing depth has been widely recognized (Burst, 1957; Perry and Hower, 1970; Schultz, 1978), and allows the degree of illitization in shales to be used as a geothermometer (Huang et al., 1993; Pollastro, 1993; Price and McDowell, 1993; Hillier et al., 1995; Uysal et al., 2000). Results from this study demonstrate that the mixing of sediments from multiple sources severely complicates deciphering the illitization history of sedimentary successions and its application to infer burial and thermal histories. Shales containing a high proportion of material derived from recycled sediments could have a ‘head start’ in the illitization process (Rettke, 1981), compared with those that are dominated by higher proportions of volcanic ash input, because the kinetics of the smectite-to-illite reaction appears to be more sensitive to the time available for the process than to the actual burial temperature (Eberl et al., 1987; Hillier et al., 1995). Many studies of ancient mudstone successions containing bentonites show that the mixed-layer I/S in shales usually contains a greater amount of illite-type layers compared with mixed-layer I/S or smectite in associated bentonites (Schulz, 1978; Sucha et al., 1993; Hillier et al., 1995). On one hand, some researchers have attributed this difference to delayed illitization of smectite in the bentonite (Li et al., 1997). On the other hand, in the Pierre Shale of the northern Great Plains region, comparison of the clay minerals in the bentonite and host shales that had different burial histories shows no appreciable tendency for bentonite to lag behind shales during illitization (Schulz, 1978). The difference in the amount of illite-type layers in the mixed-layer I/S between bentonites and associated shales is therefore more probably the result of higher proportions of recycled sediments in shales (Sucha et al., 1993). Therefore, if one wants to use mixed-layer I/S compositions in a given sedimentary succession to infer thermal regimes, the complex sources of clay minerals have to be taken into consideration. Careful petrographic examination and selection of samples without shale lithics should help elucidate the clay mineral signals that reflect burial history from those influenced by recycled sediments.

6 | CONCLUSIONS

The origin of clay minerals in the Upper Cretaceous Tununk Shale Member of the Mancos Shale in south-central Utah is complex. Through integration of sedimentologic, petrographic and clay mineralogical analysis, however, this complexity can be evaluated and clay mineral origins can be decoded. The main conclusions from this study are:

1. In the Tununk Shale, the phyllosilicate content of bentonites consists of abundant smectite (>80%) and minor amounts of biotite and kaolinite. The smectite in the bentonites reflects alteration in volcanic glass in the marine environment. Bentonites can be reworked and mixed with background sediment by benthic bioturbation and bottom currents. The pure nature of smectite (containing no illite-type layers) in un-reworked bentonites indicates a lack of illitization during burial, and by extension, that burial illitization of smectite in associated shales should be negligible.

2. Clays in Tununk Shale beds consist dominantly of mixed-layer illite/smectite (I/S) with up to 45% illite-type layers, small amounts of kaolinite and mica, and sometimes trace amounts of chlorite. The SEM examination indicates that the clay minerals in shales occur in three forms: (a) within clay-dominated composite particles (or aggregates); (b) in the fine-grained matrix; and (c) within pore spaces (both intergranular and intragranular).

3. The most abundant types of clay-rich composite particles in the Tununk Shale are altered volcanic rock fragments (dominated by smectite) and shale lithics (dominated by mixed-layer I/S). Altered volcanic rock fragments in shales were most probably derived from re-suspended
and re-deposited volcanic ash particles (volcanic glass), judging from their compositional and textural similarity to bentonites. Shale lithics can plausibly be derived from (a) erosion of older mudstone successions exposed in the hinterland and (b) submarine erosion at the unconformity located updip of the study area.

4. Integrating XRD data with petrographic observations suggests that mixed-layer I/S, the dominant clay mineral in these shales, can be plausibly derived from older mudstones/shales (as suggested by the presence of shale lithics) and from soil profiles developed on older volcanic rocks and weathering of volcanic debris that had been deposited in the hinterland. The biotite in shales can be attributed to reworking and redistribution of volcanic ashes in the marine environment as well as to terrigenous input (also supplying some muscovite). A significant amount of kaolinite and chlorite was precipitated as early diagenetic cements in the pore spaces.

5. Clay minerals in the Tununk Shale were derived from multiple sources. These include terrigenous input (weathering products and recycled sediments), wind-borne volcanic ash, intrabasinal erosion, and early diagenesis. The example of the Tununk Shale serves to illustrate that complex origins of clay minerals in marine mudstone successions place significant limitations on the use of clay mineral assemblages to infer palaeoclimate and for the reconstruction of burial and thermal histories (based on the progressive conversion from smectite to illite). For best results, clay mineralogical analysis must be integrated with careful studies of sedimentologic and petrographic characteristics, so that a comprehensive understanding of the origin of clay minerals in fine-grained sedimentary successions may be obtained.

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CONFLICT OF INTEREST

The authors have no conflict of interest to declare.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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