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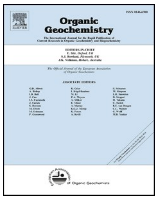

Review Article

Organic matter enrichment in black shales: How important are clay minerals?

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ABSTRACT

Clay minerals possess large surface areas and have long been thought to contribute to organic matter (OM) enrichment in black shales through the adsorption of OM in clay mineral interlayers. In this study, we review OM types and their modes of occurrence in fresh muddy sediments and their lithified counterparts, black shales, as well as the potential role of clay-OM interactions during black shale deposition. Solid OM (tens of nanometer- to hundreds of micrometer-scale particles) is the dominant form of OM in fresh muddy sediments and black shales and is too large to be accommodated in the interlayer region (<5 nm) of smectite and interstratified illite/smectite. For this reason alone, it is implausible that an interlayer adsorption mechanism would be responsible for bulk organic carbon preservation in black shales. OM and clay minerals in nature are instead physically associated via the formation of floccules and aggregates, which serve to retard the microbial degradation of OM. The association of OM and clay minerals argues for a reevaluation of the impact of clay minerals on organic carbon preservation in the sedimentary systems and the global carbon cycle.

1. Introduction

Organic-rich sedimentary rocks are not only important petroleum source rocks (Tissot and Welte, 1984) but also play an important role in the global carbon cycle (Berner, 2003; Murray and Jagoutz, 2024). Primary factors influencing organic matter (OM) enrichment in these rocks include paleoproductivity (Pedersen and Calvert, 1990), bottom-water redox conditions (Demaison and Moore, 1980), sedimentation rate (Tyson, 2005), and the combined effect of these factors as modulated by relative sea-level fluctuations (Sageman et al., 2003; Arthur and Sageman, 2005; Bohacs et al., 2005; Tyson, 2005; Dong et al., 2018; Liu et al., 2019).

Black shales are fine-grained sedimentary rocks with an enrichment of OM (derived from once-living organisms) and varying contents of clay minerals (Ulmer-Scholle et al., 2014). Research conducted over the past three decades posits that clay minerals, specifically smectite and interstratified illite/smectite, are closely linked to the enrichment of OM in black shales (Keil and Hedges, 1993; Mayer, 1994a, b; Kennedy et al., 2002, 2014; Kennedy and Wagner, 2011; Blattmann et al., 2019; Zhu et al., 2020; Zhao et al., 2023), with the basic assumption being that smectite and interstratified illite/smectite adsorb large amounts of OM within their interlayers (Kennedy et al., 2002, 2014; Kennedy and Wagner, 2011; Cai et al., 2023). Smectite has a specific surface area (SSA) of up to 800 m^2/g as measured via ethylene glycol monomethyl ether (EGME) adsorption (Środoń and McCarty, 2008; Kennedy et al., 2014; Bu et al., 2019), and multiple investigations have reported a positive correlation between total organic carbon (TOC) and the SSA of black shales in support of a mineral-surface control mechanism resulting in OM enrichment in black shales (Kennedy et al., 2002, 2014; Kennedy and Wagner, 2011; Zhu et al., 2018, 2020; Cai et al., 2023).

The objective of this study is to review the relationship between clay minerals and OM in fresh muddy sediments and black shales, with a focus on primary OM (kerogen). Secondary OM generated from kerogen thermal transformation (bitumen and solid bitumen) and its relationships with clays are not part of this discussion. Kerogen is equivalent to primary macerals (e.g., alginite, bituminite, vitrinite, inertinite) in thermally immature shales in organic petrology nomenclature (Mastalerz et al., 2018; Liu et al., 2022). The types and occurrence modes of OM in fresh muddy sediments and black shales, as well as the structure and properties of clay minerals, are reviewed in the context of a presumed mineral-surface control mechanism on OM enrichment. Clay mineral-OM interactions are discussed to elucidate the relationships

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between clay minerals and OM accumulation.

The data discussed in this paper mostly come from the literature, but they are also supplemented by unpublished data from our samples. These additional samples come from the large collection of black shales in the Center for Energy at the Indiana Geological and Water Survey and the Indiana University Shale Research Lab. The samples include the Devonian New Albany Shale of the Illinois Basin, USA, and the Ordovician-Silurian Wufeng-Longmaxi Shale of the Sichuan Basin, China.

2. Organic matter in fresh muddy sediments and black shales

2.1. Organic matter in fresh muddy sediments

The first step to understand the interactions between clay minerals and OM in black shales is to understand the types of OM and their modes of occurrence at the time of deposition, i.e., in what form the OM occurs and where it is located in fresh organic-rich muddy sediments. In freshly deposited sediments, OM occurs as a variety of solid components (including slimy amorphous OM) and as dissolved OM (Boucsein and Stein, 2000; Hansell et al., 2009; Bainbridge et al., 2018; Kleber et al., 2021).

Algae and terrestrial higher plants (post-Silurian) are the main primary producers contributing to sedimentary OM (Taylor et al., 1998; Burdige, 2007; Vandenbroucke and Largeau, 2007; Kleber et al., 2021; Zhou et al., 2024). Ransom et al. (1997) examined OM and clay mineral relationships in sediments and marine snow (macroscopic organic aggregates; Turner, 2015) from the northern California continental slope and Eckernförde Bay in the Baltic Sea via TEM. They found that OM mainly occurs as 1) discrete, discontinuous particles; 2) bacterial cells and associated muco-polysaccharide networks; and 3) localized smears generally associated with clay minerals and domain junctions in clay-rich flocs (Fig. 1). All three of these types of occurrences are within the size range of tens to hundreds of nm. Miao et al. (2018) identified eight organic-walled microfossil and particle morphologies (e.g., spores, algae, fungi, and zooclasts) in the Neogene sediments from the South China Sea at IODP Site 1433. Oboh-Ikuenobe et al. (1999) identified 12 types of dispersed OM (e.g., marine palynomorphs, nonmarine algae, wood, and spores) in the Oligocene to Early Miocene sediments at ODP Site 959.

Oceanic waters contain large quantities of dissolved OM in the form of organic molecules (Arístegui et al., 2002; Ogawa and Tanoue, 2003; Hansell et al., 2009; Dittmar and Stubbins, 2014; Arrieta et al., 2015), and dissolved organic carbon (global inventory ~662 × 10^{15} g) may well constitute >90 % of the total organic carbon in the ocean (Ogawa and Tanoue, 2003; Hansell et al., 2009; Dittmar and Stubbins, 2014). Yet, because dissolved organic carbon in the ocean exists at extremely low concentrations (34–~80 μmol kg^{-1}; Hansell et al., 2009), the pore waters of freshly deposited muds can only contribute negligible amounts of OM, and solid organic matter (algae, zooplankton, plant fragments, and bacterial biomass; Boucsein and Stein, 2000; Wakeham and Canuel, 2006; Milliken et al., 2014; Bainbridge et al., 2018) contributes the bulk of OM to accumulating muddy sediments. If one were to assume that dissolved OM is the only OM that exists in a fresh mud (~85 vol% water content, Fig. 2) and that all of it can be converted to sedimentary OM, pore waters with 80 μmol kg^{-1} dissolved organic carbon would result in a (consolidated) sediment with a miniscule TOC of 0.0002 wt%, four

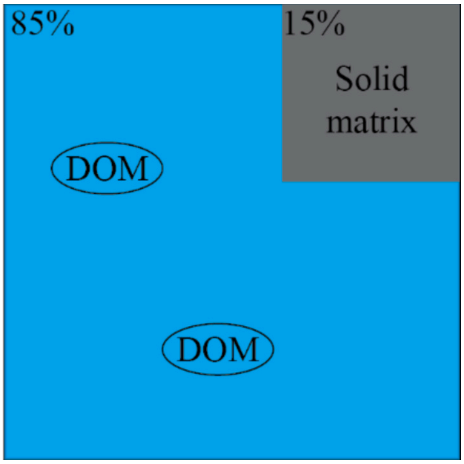

Fig. 2. Graphic that illustrates the volume percent of solid matrix (~15 %) and pore water (~85 %) in fresh organic-rich muddy sediments. Solid matrix is composed of clay minerals, silt grains, and organic matter (algae, terrestrial higher plant fragments (post-Silurian), zooclasts, etc.), whereas pore waters contain dissolved salts and small amounts of dissolved organic matter. DOM = dissolved organic matter.

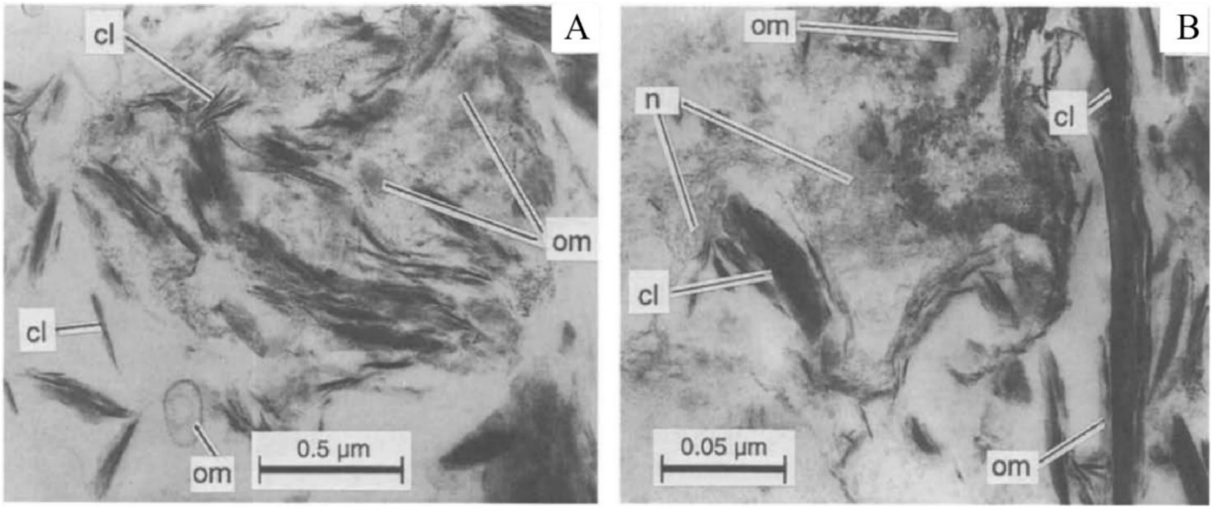

Fig. 1. TEM images showing the textural relations between OM and mineral grains in a typical seawater-sediment interface from Eckernförde Bay in the Baltic Sea (Ransom et al., 1997). TEM sections were stained with osmium tetroxide, lead citrate, and uranyl acetate to make organic matter visible under the electron beam. (A) Aggregate of clay minerals (cl) and organic matter particles (om) from Eckernförde Bay from a burial depth of 1 mm. (B) Clay minerals (cl) and diffuse organic matter (om) from Eckernförde Bay that is caught up in a muco-polysaccharide network (n) in the sediment 1 mm below the sediment–water interface. Note that abundant organic matter is visibly mixed with clay particles at the 10′s to 100′s nm scale.

orders of magnitude less than what is considered necessary for a generic petroleum source rock (TOC > 1 wt% for good petroleum potential; Peters and Cassa, 1994). As most pore waters (including their dissolved OM) are expelled to the overlying seawater early in the compaction history, even the 0.0002 wt% TOC value is excessively optimistic. In addition, the adsorption capacity of natural smectite for organic molecules such as benzene and trichloroethylene is <2 mg/g, a concentration that translates into a TOC content of 0.2 % at an equilibrium concentration at 100 mg/L (Boyd et al., 1988), which is still high for dissolved organic carbon concentration in the ocean (80 μmol kg^{-1} translates into 6.24 mg/L in the case of benzene). Thus, the OM budget of accumulating muds is essentially controlled by solid OM (Fig. 3).

2.2. Organic matter in black shales

Black shales are consolidated organic-rich muddy sediments. OM in black shales consists of solid components (kerogen) and solvent-soluble OM (bitumen and oil). Solid OM in black shales can be identified under the microscope using well-established organic petrographic techniques (Suárez-Ruiz et al., 2012; Hackley and Cardott, 2016; Flores and Suárez-Ruiz, 2017). Solid OM in black shales occurs as micrometer-size particles and can be described as different macerals based on their reflectance, morphology, texture, and fluorescence properties (Hackley and Cardott, 2016; Flores and Suárez-Ruiz, 2017; Mastalerz et al., 2018; Liu et al., 2022). Common macerals in black shales include primary components such as alginite (algal material), bituminite (algal/bacterial biomass), vitrinite (woody material), inertinite (burnt or oxidized biomass), zooclasts (animal fragments), and secondary products (solid bitumen) (Fig. 4). Except for solid bitumen that, as a product of kerogen transformation, is abundant in thermally mature shales, most or all of the other macerals are present in muddy sediments during deposition and in most cases in a form similar to that observed in black shales, except for the compaction effect that could reduce the particle size and possibly modify morphology. Some difference in the mode of occurrence between fresh muddy sediments and black shales could be expected in bituminite, which is likely a product of the bacterial degradation of algal material early in diagenesis (Kus et al., 2017).

The microscopically identifiable solid OM content (expressed as volume %) generally matches the TOC content (in weight %) (Zhou and Sanei, 2025), with volume % approximately twice the weight % due to a lower density (1.1–1.5 g/cm^3) of OM (Mastalerz et al., 2012; Craddock et al., 2020) when compared with minerals (average ~2.7 g/cm^3). Zhou and Sanei (2025) showed a strong correlation between calculated TOC volume percent (weight percent measured by pyrolysis) and maceral content based on point counting in the Vaca Muerta Formation, Argentina, and in the Lark Formation, eastern North Sea Basin (R^2 = 0.80 and 0.98, respectively). This correspondence between maceral content quantified under the microscope and TOC content provides a strong argument that solid OM is the main contributor to the TOC of black shales. OM in black shales (mostly hydrocarbons) that can be extracted with non-polar solvents such as chloroform exists in large quantities in the oil window (Jarvie, 2012; Hu et al., 2021) and declines via expulsion of hydrocarbons into migration pathways. As such, it should not be considered in evaluations of clay mineral-OM interactions during or shortly after deposition.

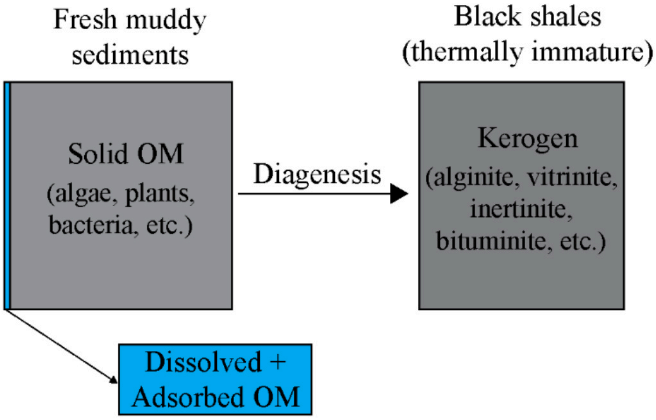

Fig. 3. Graphic that illustrates the evolution of organic matter (OM) from fresh muddy sediments to thermally immature black shales that would be considered petroleum source rocks. Dissolved and adsorbed OM contributes to kerogen formation in black shales, but in a trivial amount. Solid OM contributes to the bulk of all kerogen in black shales. The volume percent of dissolved and solid OM in fresh muddy sediments is not to scale.

3. Clay mineral structure and properties

Clay minerals are a group of hydrated layered aluminosilicates composed of silicon-oxygen tetrahedral and aluminum-oxygen octahedral sheets (Grim, 1953; Brindley and Brown, 1980) and are ubiquitous components of fine-grained sediments such as mudstones and shales (Ulmer-Scholle et al., 2014). As a result of isomorphous substitutions within the tetrahedral and the octahedral sheets, the common clay minerals smectite and illite have negatively charged layers, with charge-balancing cations located in the interlayer region. These so-called interlayer cations can be accompanied by H_2O molecules and a variety of organic molecules. The illite interlayer region is typically of well-defined thickness and occupied by K^+ ions with few or no H_2O molecules, whereas the smectite interlayer region is of variable thickness and can be occupied by alkali and alkaline earth cations (e.g., Na^+ and Ca^{2+}) and other molecules such as H_2O and polar organic molecules (Fig. 5; Rotenberg et al., 2007; Johnston, 2010; Bu et al., 2019).

The interlayer spacing or layer-layer repeat distance of these different clay minerals is variable (Table 1). For example, the layer-layer repeat (d_{001}) distance of smectite ranges typically from 1.2 to 1.7 nm, but may reach up to 5 nm, depending on factors such as tetrahedral-octahedral-tetrahedral (TOT) layer charge, activity of H_2O, and the presence or absence of organic molecules. The large interlayer spacing of smectites, and the fact that individual layers may disaggregate fully in some liquids, result in their large SSA values. For example, the SSA of smectite measured by EGME adsorption can be up to 800 m^2/g (Środoń and McCarty, 2008; Kennedy et al., 2014; Bu et al., 2019) because EGME is thought to enter the interlayer region of clay minerals (Michot and Villieras, 2006).

The types of detrital clay minerals transported to oceans or lakes depend on the interplay of terrestrial bedrock types and weathering conditions (Hallam et al., 1991; Potter et al., 2005). Because smectite and interstratified illite/smectite are the most common clay minerals in fresh sediments and immature shales (Perry and Hower, 1970; Awwiller, 1993; Li et al., 2019) and also have favorable SSAs and interlayer properties to permit the adsorption of organic molecules, studies have suggested that they play a major role in the OM enrichment in black shales (Kennedy et al., 2002, 2014; Kennedy and Wagner, 2011; Zhu et al., 2018, 2020; Cai et al., 2023). They will, therefore, be the focus of the following discussion about OM-clay interactions.

4. Clay mineral-organic matter interactions

4.1. Interlayer adsorption mechanism for organic matter enrichment in black shales

An interlayer adsorption mechanism has been proposed as the key control on the accumulation of OM in black shales (e.g., Mayer, 1994a, b; Kennedy et al., 2002, 2014; Kennedy and Wagner, 2011; Zhu et al., 2020; Blattmann et al., 2019; Cai et al., 2023; Zhao et al., 2023). This hypothesis posits that the large SSAs of smectite and interstratified illite/smectite, reflective of the EGME-accessible interlayer region, facilitate OM preservation and burial (Kennedy et al., 2002, 2014;

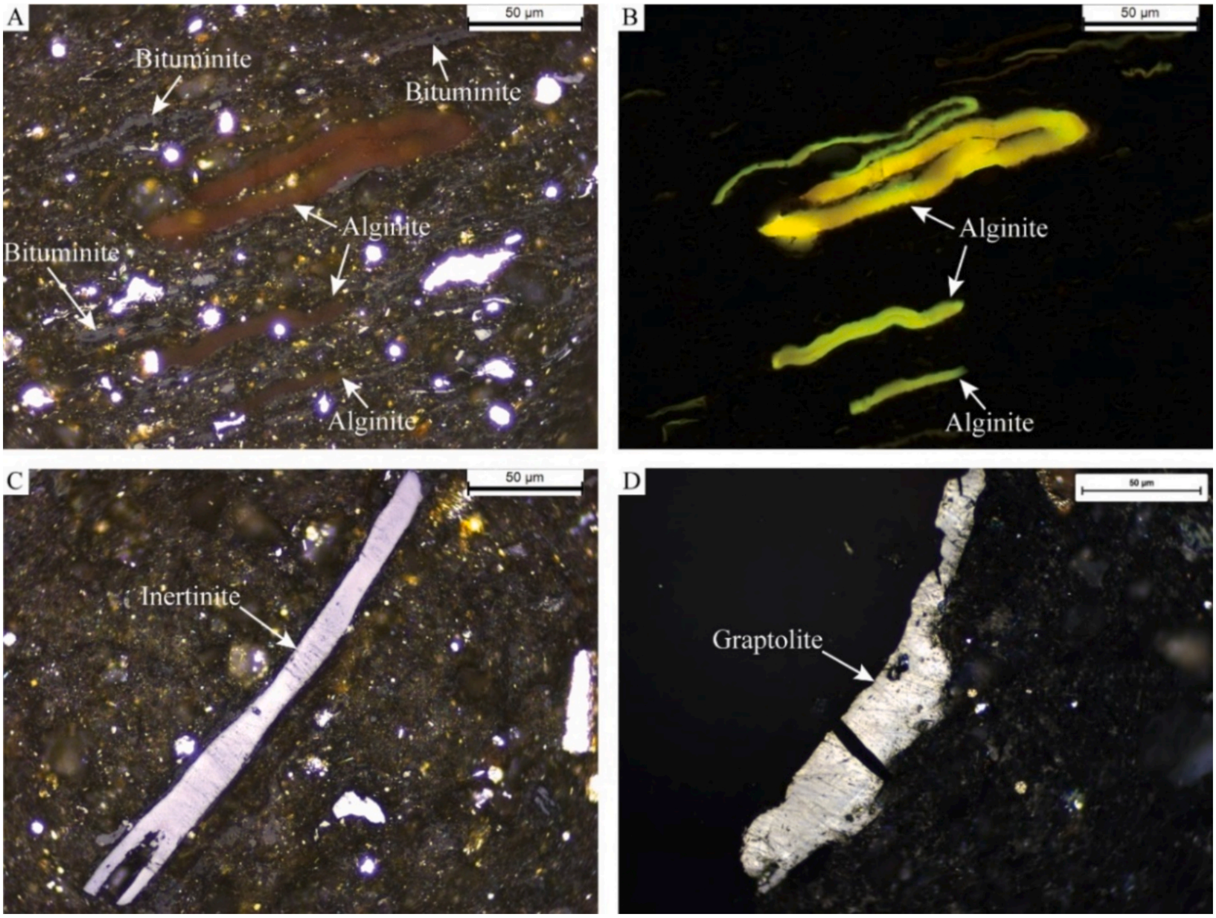

Fig. 4. Photomicrographs of common OM components in black shales. (A, C, D) Reflected white light and oil immersion; (B) Fluorescence mode. (A, B) Alginite and bituminite (the two most common macerals in black shales) in the New Albany Shale, Illinois Basin, USA. Alginite derived from *Tasmanites* cysts shows greenish-yellow fluorescence. (C) Inertinite (derived from terrestrial higher plants) in the New Albany Shale, Illinois Basin. (D) Graptolite fragment in the Wufeng-Longmaxi Shale, Sichuan Basin, China. (A-C) Core sample of the New Albany Shale, Daviess County, Indiana, USA (vitrinite reflectance VR_o 0.55% and TOC 10.77 wt%). (D) Core sample of the Wufeng-Longmaxi Shale from Chongqing, China (equivalent VR_o 2.23% and TOC 5.92 wt%). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

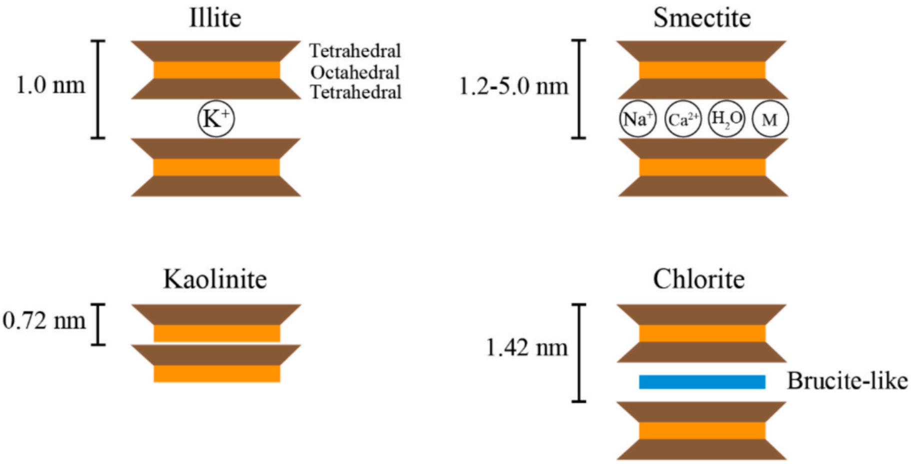

Fig. 5. Schematic of layering styles of common clay minerals. M = organic molecules. The tetrahedral-octahedral-tetrahedral layer thickness and interlayer spacing are not to scale.

Kennedy and Wagner, 2011; Zhu et al., 2018, 2020; Cai et al., 2023). Specifically, these studies suggest that a critical element of OM preservation is protection from microbial degradation in clay mineral interlayer locations. However, this clay mineral surface control hypothesis does not fully explain the enrichment of OM in black shales.

First of all, excellent carbonate and siliceous source rocks, as well as coals, contain scant quantities of clay minerals but have significant TOC (Katz et al., 2000; Ko et al., 2017; Xia et al., 2019), which makes clay

Table 1
Layer-layer repeat distance, clay type, and major bond type of common clay minerals in shales.

Clay mineral	Layer-layer repeat distance (d_{001}) (nm)	Clay type	Major interlayer bonding
Smectite	0.96–4.96[a]	2:1	van der Waals and ionic bonds
Illite	1.0[b]	2:1	van der Waals and ionic bonds
Chlorite	1.42[b]	2:1:1	Ionic and hydrogen bonds
Kaolinite	0.72[b]	1:1	Hydrogen bond

[a] Johnston, 2010.
[b] Moore and Reynolds, 1989.

mineral surface control on OM enrichment in marine and lacustrine mudstones far less certain than presumed by previous studies (Kennedy et al., 2002, 2014; Kennedy and Wagner, 2011; Zhu et al., 2018, 2020; Cai et al., 2023). For example, the Lower Cretaceous Sunniland carbonate source rocks in the South Florida Basin have up to 7.41 wt% TOC with 79 % calcite (Palacas et al., 1984), and the Lower Cambrian Niutitang Formation siliceous shales in South China have 4.6 wt% clay minerals and 20.1 wt% TOC (Tan et al., 2021). Other excellent source rocks with low clay content include the Utica Shale and Point Pleasant Formation of the Appalachian Basin (Sheets et al., 2021) and the Eagle Ford Formation (Ko et al., 2017).

Second, the clay-mineral-surface control mechanism implies that most OM in black shales is in dissolved form at the time of deposition (Kennedy et al., 2002). However, as discussed in Section 2, OM in freshly accumulated marine or lacustrine muds is invariably dominated by solid OM and is composed of various macerals (Taylor et al., 1998; Boucsein and Stein, 2000; Bainbridge et al., 2018). OM derived from terrestrial higher plants (vitrinite and inertinite), algae (e.g., *Tasmanites*, *Botryococcus*, and *Leiosphaeridia*), and zooclasts (e.g., graptolites and chitinozoans) occurs as discrete particles in marine and lacustrine sediments, and these particles are generally larger than 1 μm. For example, *Tasmanites* algae, a type of unicellular green algae (Tappan, 1980), is widely

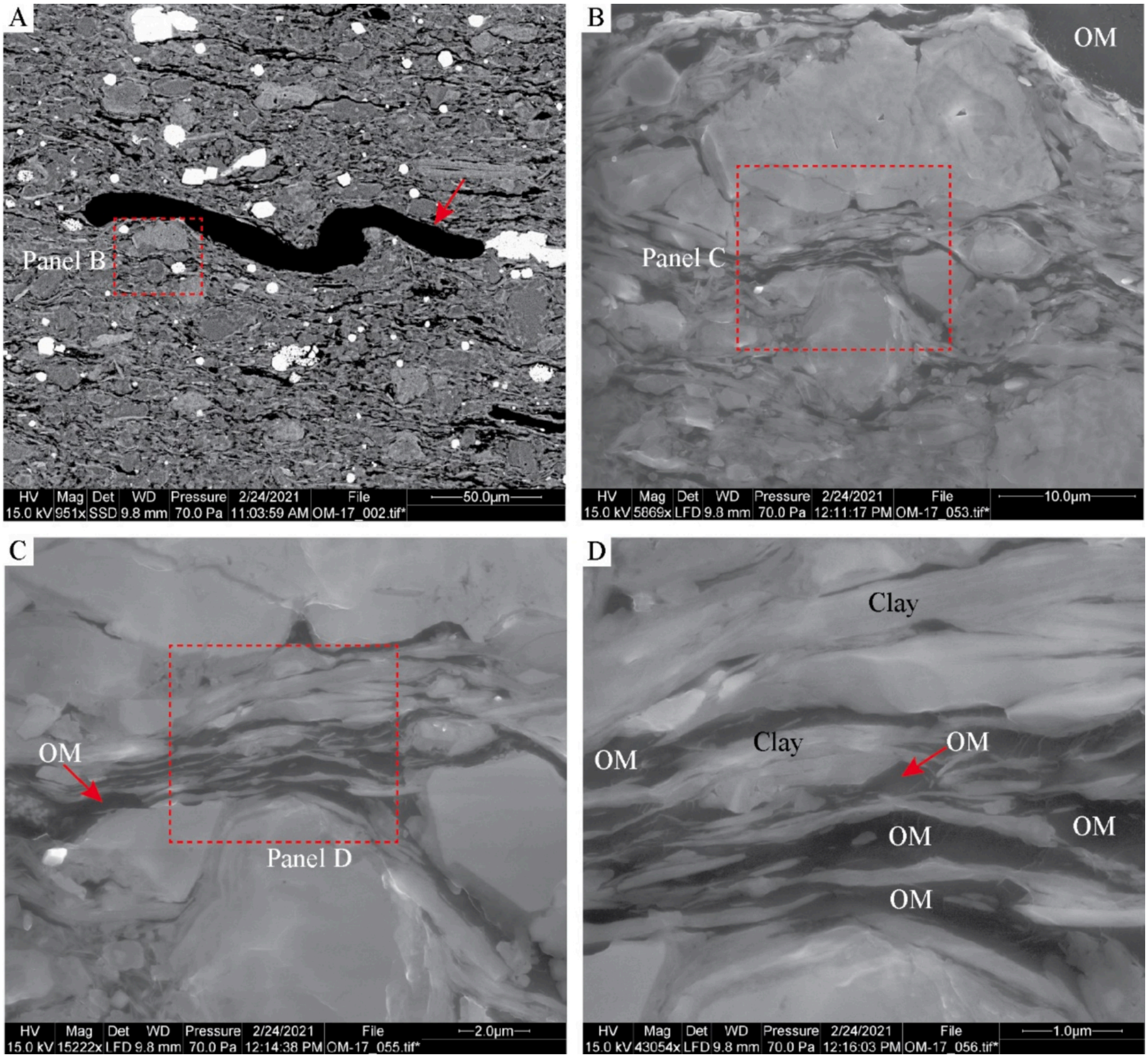

Fig. 6. SEM images of the New Albany Shale, Illinois Basin. (A) Backscattered-electron mode; (B-D) Secondary-electron mode. Solid organic matter (OM) (marked by red arrows) appears black under the SEM and constitutes the bulk of OM in the sample. Red dashed squares in panels (A), (B), and (C) represent enlarged area shown in panels (B), (C), and (D), respectively. Red arrow in panel A marks a *Tasmanites* cyst. OM in panels B-D is bituminite, one of the most common macerals in the New Albany Shale. Bituminite was identified based on its streaky nature and association with clay minerals. Core sample of the New Albany Shale, Daviess County, Indiana, USA (VR_o 0.55% and TOC 7.29 wt%). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

present in the Tasmanite oil shale from Tasmania, Australia (Revill et al., 1994) and the Devonian black shales (e.g., New Albany Shale, Ohio Shale, Antrim Shale, Chattanooga Shale, and Bakken Shale) of the North America (Hackley et al., 2017; Liu et al., 2017, 2019) and can contribute 80 % or more of the total OM in these black shales. Given that *Tasmanites* cysts range in size from 100 to 500 μm, they obviously cannot fit into the nanometer-scale interlayers (<4 nm) of smectite and interstratified illite/smectite. Fig. 6 shows a typical texture of black shales under the SEM, which demonstrates the dominance of solid OM. Further examination by TEM imaging at a higher magnification also shows a dominance of solid OM in black shales (Fig. 7). As discussed in Section 2, ocean water contains dissolved OM that can be adsorbed by clay minerals (Arístegui et al., 2002; Lagaly et al., 2006; Hansell et al., 2009; Arrieta et al., 2015; Kleber et al., 2021) and then converted to kerogen via geopolymerization during diagenesis (Burdige, 2007; Moore et al., 2023). However, involving dissolved OM clearly cannot be the dominant mechanism for kerogen formation in marine and lacustrine sediments. One might argue that the amorphous kerogen in Figs. 6 and 7 could have started as organic molecules adsorbed in the interlayers and then converted to kerogen during diagenesis. This process might be responsible for a small portion of nanometer-scale amorphous kerogen (Fig. 3). However, if the bulk of amorphous kerogen formed through this process, it would be more homogeneous instead of showing variations in fluorescence, texture, and reflectance (Hackley and Cardott, 2016; Flores and Suárez-Ruiz, 2017; Kus et al., 2017; Mastalerz et al., 2018; Liu et al., 2022; Gonçalves et al., 2024).

Third, if the surface area of clay minerals indeed played a dominant role (first-order control) in OM accumulation, a positive correlation between TOC and clay mineral content of black shales should be expected. However, after analyzing the TOC vs total clay mineral content relationship reported from more than 1000 shale samples (including some carbonaceous chert and carbonates) of different ages and

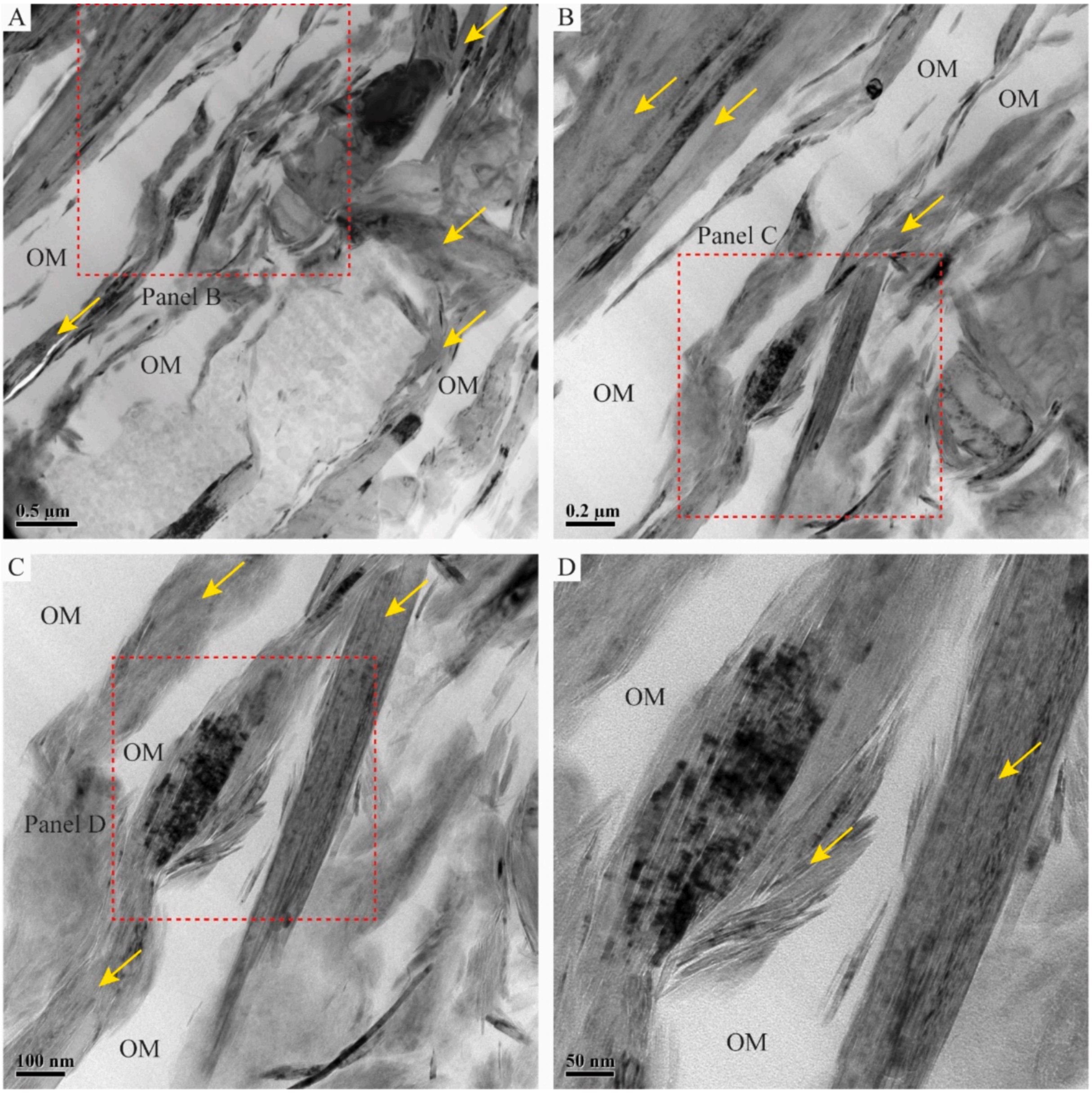

Fig. 7. TEM images of the New Albany Shale, Illinois Basin. Solid organic matter (OM) appears white and constitutes the bulk of OM in the sample, and clay minerals are medium to dark gray (marked by yellow arrows). OM in the TEM images could be solid bitumen considering the oil-window maturity of this sample. Solid bitumen could be converted from bituminite based on its streaky nature and association with clay minerals. Red dashed squares in panels (A), (B), and (C) represent enlarged area shown in panels (B), (C), and (D), respectively. Core sample of the New Albany Shale, Posey County, Indiana, USA (VR$_o$ 0.83% and TOC 12.25 wt%). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

compositions, a weak parabolic trend, instead of a positive correlation, emerges (Fig. 8). TOC content is mostly below 10 wt% when total clay mineral content is <10 wt% and is below 5 wt% when total clay mineral content is >60 wt%. High TOC values (>10 wt%) usually occur when total clay mineral content is in the range of 10–50 wt%. The first-stage positive correlation between TOC and total clay contents results because 1) detrital materials provide nutrients (N, K, P, Fe, etc.) to phytoplanktons and enhance primary productivity in surface waters (Killops and Killops, 2005); and 2) the formation of composite floccules with clay particles acts to protect OM from microbial decay or enzymatic attack (Bennett et al., 2012a, b; Zhao et al., 2023). The negative correlation with high total clay content is due to dilution by terrigenous clastic materials (Bohacs et al., 2005; Liu et al., 2019). A basin-scale study of Devonian black shales in the Appalachian Basin demonstrated an inverse relationship between dilution and OM enrichment (Smith et al., 2019).

Clay minerals play an important role in preserving OM in soils (Kleber et al., 2021). A conceptual model of mineral-OM interactions in soils, supported by NanoSIMS imaging, suggested that the intercalation of OM into phyllosilicate interlayers is not the major interaction between OM and clay minerals in soils (Schweizer, 2022), a finding analogous to those of this study.

4.2. *Clay mineral-organic matter associations*

Black shales have long been interpreted to be deposited in quiescent anoxic conditions, presumed to be governed by settling from suspension. However, flume studies and sedimentary features observed in ancient shales demonstrate that mud can be deposited by swiftly moving bottom currents that are also powerful enough to transport and deposit sand (Schieber et al., 2007, 2023; Schieber and Southard, 2009; Macquaker et al., 2010; Schieber, 2011; Yawar and Schieber, 2017; Tribovillard et al., 2019). In these flume experiments, once flow velocity was lowered to near bed velocities of 25 cm/s or a bottom shear stress of 0.25 Pa, bedload floccules formed, giving rise to migrating floccule ripples, and accretion of flow-deposited mud layers (Schieber et al., 2007, 2023; Schieber, 2011).

OM is a common constituent of muddy sediments, and how it interacts with clay minerals during the bedload transport of organic-rich muddy sediments is an issue that requires further research. Nonetheless, the formation of clay aggregates physically traps OM during sediment transport and deposition (Schieber et al., 2011, 2012, 2018; Bennett et al., 2012a, 2012b). Electrostatic attraction between OM and clay surfaces contributes to the intimate mixing of OM and clay particles, leading to the formation of OM-clay floccules and associated aggregates (Fig. 9; Ransom et al., 1997; Bennett et al., 2012a, 2012b; Cai et al., 2023).

Sinking quickly through the water column, marine snow aggregates are considered the main vehicle for gravitational (settling) transport of OM to the seafloor (Kiørboe, 2001; Macquaker et al., 2010; Turner, 2015). These aggregates are composed of phytoplankton, zooplankton, microbial and fecal components, as well as minerals and terrigenous organic matter, and they form in the upper water column (Turner, 2015). After arriving at the seafloor, they can be reworked by bottom currents (Macquaker et al., 2010). Flume experiments and the rock record show that bedload transport of seafloor muds composed of various types of aggregates is common during mudstone deposition (Schieber et al., 2007, 2019; Schieber and Southard, 2009; Macquaker et al., 2010; Schieber, 2011; Hart et al., 2013; Li and Schieber, 2018; Li et al., 2021). It is also plausible that marine snow aggregates disintegrate and reconstitute as secondary organomineralic aggregates during bedload transport, an assertion that is supported by observation of clay-OM aggregates in flume experiments (Fig. 10; Schieber et al., 2011, 2012, 2018; Bennett et al., 2012b). Organomineralic aggregates can form in the water column, at the sediment–water interface, or during bedload transport, and can undergo multiple cycles of formation and disintegration before final deposition. Where examined (Figs. 1 and 10), OM is abundantly visible between clay particles. Because of its significantly greater density than water-rich OM and muco-polysaccharides, mineral matter accelerates the settling of organomineralic aggregates. In

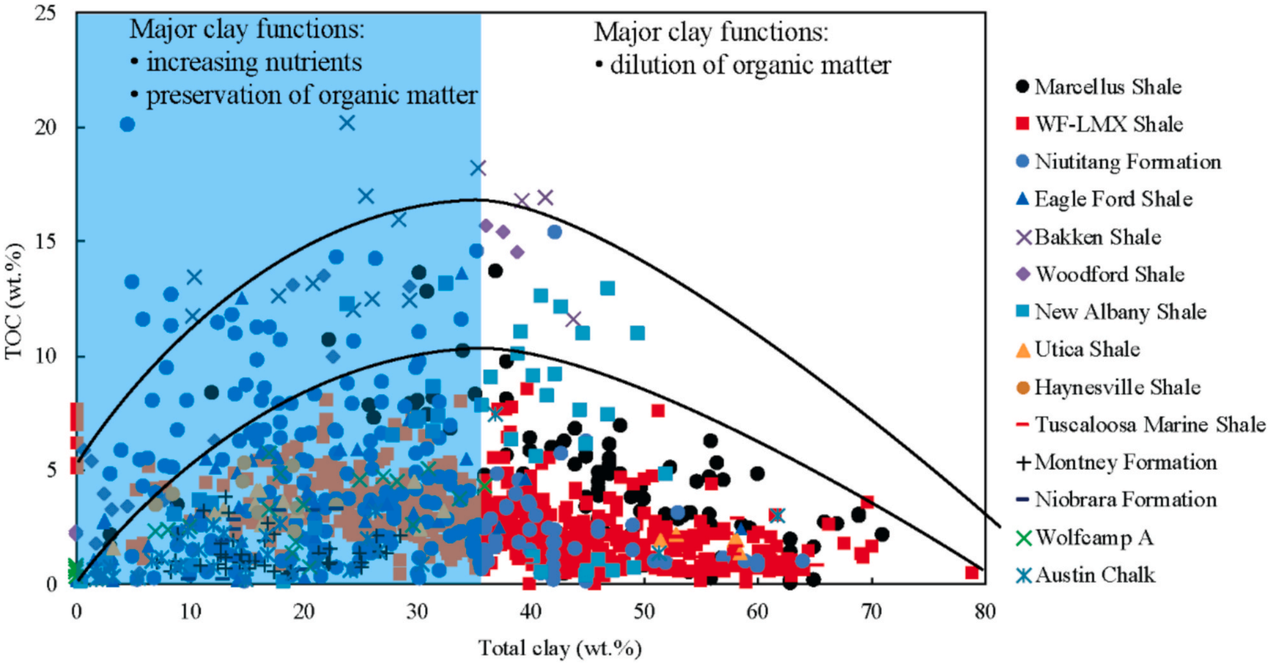

Fig. 8. Plot showing the relationship between total organic carbon (TOC) and total clay mineral content for shales (including some carbonaceous cherts and carbonates) of different ages and compositions. 1161 data points from 14 shale formations worldwide. WF-LMX = Wufeng-Longmaxi Shale of the Sichuan Basin, China. Although the clay minerals in these shales are now mostly illite and interstratified illite/smectite because of illitization during diagenesis, their precursors were most likely smectite and interstratified illite/smectite (e.g., Abercrombie et al., 1994; Bjørlykke, 1998). The citations for the plotted data are available in the Supplementary Material.

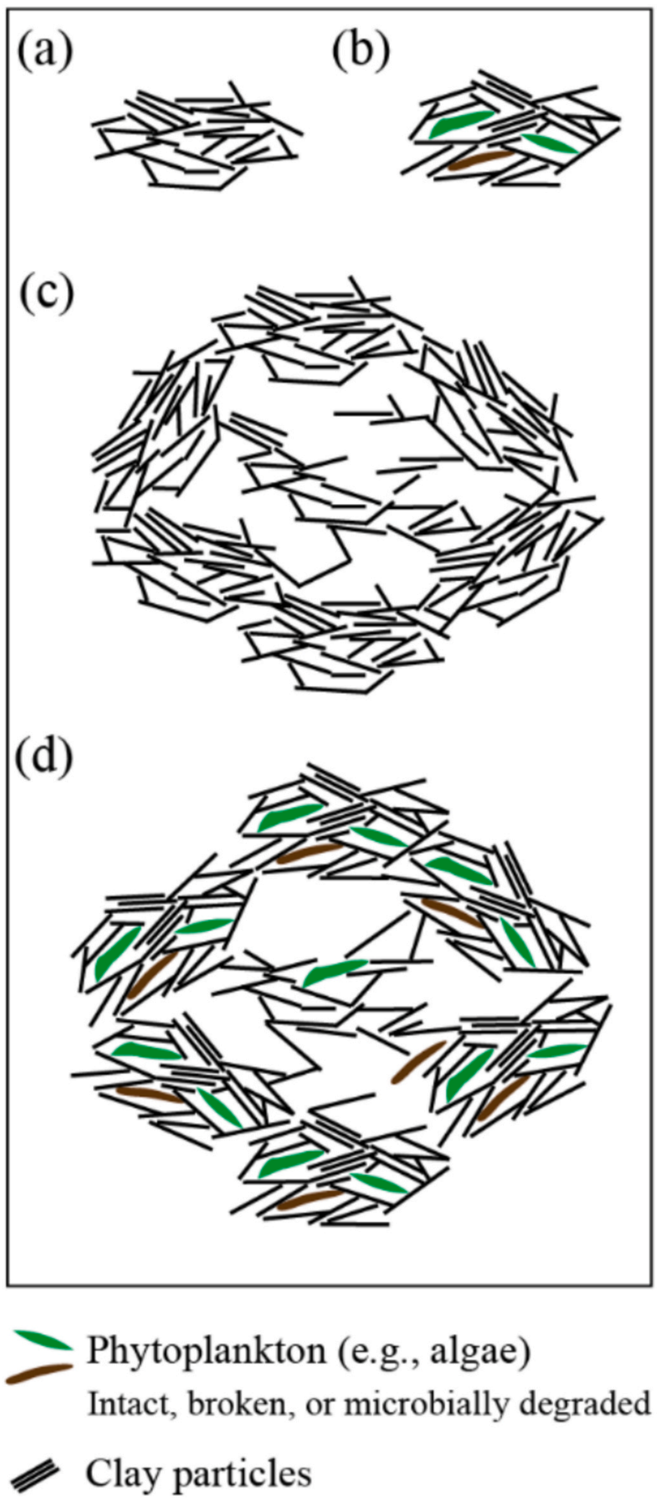

Fig. 9. Schematic illustration of clay mineral-OM floccules/aggregates. (a) Floccules composed of clay minerals only; (b) floccules composed of clay minerals and OM; (c) aggregates composed of clay floccules only; (d) aggregates composed of clay-OM floccules. OM can participate in building the floccules/aggregates instead of getting trapped by clay floccules/aggregates. Clay-OM aggregates that can reach hundreds of micrometers in size are composed of smaller floccules and micro-aggregates.

addition to clay minerals, fine-grained carbonate particles (phytoplankton sourced, such as coccoliths and foraminifera) can serve the same function and are known to form floccules that travel in bedload and deposit laminated sediments (Schieber et al., 2013).

Optical organic petrographic, SEM, and TEM observations of flume sediments (Fig. 10), fresh sediments from the ocean (Fig. 1), and the rock record (Figs. 4, 6, 7) collectively show that the bulk of OM in fine-grained sediments does not occur within the interlayer region of clay minerals. Instead, OM is intimately mixed, yet distinctly visible (Figs. 1 and 10), with clays and other mineral components as these materials settle and become organized into floccules and larger aggregates.

Once deposited, sedimentary OM in muddy sediments undergoes a series of physical and chemical changes to its molecular structure during transformation to kerogen (Durand, 1980; Vandenbroucke and Largeau, 2007). Discrete solid OM derived from algae can transform to alginite during diagenesis. A good example in the rock record would be *Tasmanites* cysts that are widely present in the Tasmanite oil shale from Tasmania, Australia (Revill et al., 1994), and the Devonian black shales of the United States (Hackley et al., 2017; Liu et al., 2017, 2019). *Tasmanites* cysts occur as elongated rods when viewed perpendicular to the bedding (Fig. 4), a result of mechanical compaction. Slimy amorphous OM and OM streaks, composed of mucus, bacteria, and degraded phytoplankton and zooplankton (Riemann and Schrage, 1978), likely transform to amorphous kerogen in black shales once solidified during diagenesis (Thompson and Dembicki, 1986; Largeau et al., 1990), an inference that is supported by the observation that bituminite in black shales is always mixed with clay minerals and clay-sized particles (Kus et al., 2017; Liu et al., 2020; Teng et al., 2021). Inorganic components in organomineralic aggregates are implicated to enhance OM preservation by acting as 1) ballast materials (e.g., clays, opaline silica, and calcite) that can increase settling velocity and decrease the residence time in the water column (Francois et al., 2002; Armstrong et al., 2009); and 2) mineral coatings that can retard the diffusion of dissolved oxygen from the water column or pore water to the interstitial OM and thereby inhibit microbial degradation (Mayer, 1994a, b; Hedges and Keil, 1995; Bennett et al., 2012a, 2012b). Chemical interactions, such as sorption, oxidation, and catalysis, occur at the mineral–OM interface (Kleber et al., 2021).

Although clay mineral-OM floccules/aggregates are proposed in this study to enhance OM preservation, other factors such as sedimentation rate, bottom-water redox conditions, and surface water productivity are also important in OM enrichment in black shales at a large scale (Demaison and Moore, 1980; Bohacs et al., 2005; Tyson, 2005). These factors interact with each other in a given depositional setting.

It should be noted that original clay mineral-OM floccules/aggregates are not likely to be identified in high-maturity black shales because OM undergoes thermal maturation, migration, and redistribution during burial (Cardott et al., 2015; Hackley and Cardott, 2016; Mastalerz et al., 2018; Camp, 2019; Sanei, 2020; Liu et al., 2022; Luo et al., 2025). OM in high-maturity black shales is dominated by solid bitumen/pyrobitumen, and the distribution and occurrence modes of OM are significantly different compared with that in freshly deposited sediments and thermally immature shales. Molecular dynamics simulations (Cao et al., 2021) suggest that liquid hydrocarbons could potentially migrate into the interlayer region of clay minerals within the oil window and subsequently degrade into solid bitumen later during burial, but this process is not related to OM accumulation and fixation at the time of deposition.

Finally, this study does not deny the existence of intercalation of organic molecules into clay mineral interlayers. There is no doubt that this process operates in nature and contributes to OM in black shales (e.g., Kleber et al., 2021). However, its relative contribution to OM enrichment in black shales is insignificant. As such, OM enrichment in black shales based on an interlayer adsorption mechanism and its importance in the carbon cycle through the geological history is suspect and should be critically reevaluated.

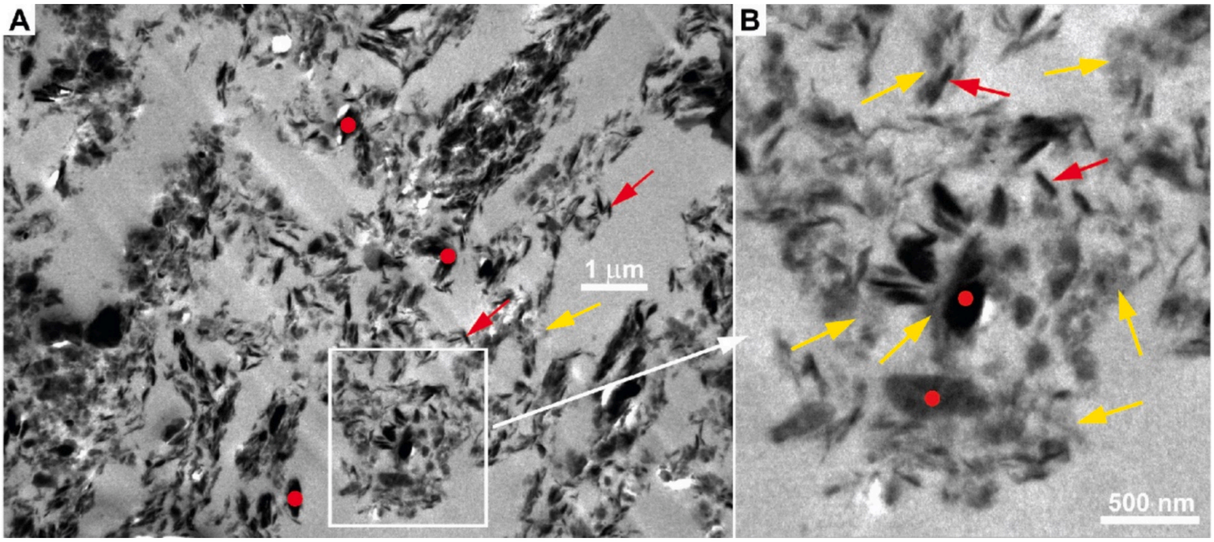

Fig. 10. TEM images of flocculated clay mineral-organic matter (OM) aggregates deposited in flume experiments (Schieber et al., 2012). (A) The interior fabric of OM-clay floccules is a network of smaller OM-clay masses with intervening pore water. (B) Detail view of OM-clay masses. Darker objects are largely clay mineral flakes (illite) in various orientations. Red dots mark “face views” of clay mineral flakes, and red arrows mark “edge views” of clay flakes. The yellow arrows point to OM that has been fixed with glutaraldehyde and stained for contrast with Pb/U. In the flume experiments, the sediment load consisted of illite and a marine snow simulant (ground and degraded kelp). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

5. Conclusions

Organic matter in thermally immature black shales and freshly deposited muddy sediments is dominated by solid constituents. Dissolved OM exists in freshly deposited muddy sediments and can be adsorbed by smectite and interstratified illite/smectite, but it is negligible compared with the mass of solid OM in a specific volume of muddy sediments. Clay minerals and OM are intimately mixed when examined under the SEM and TEM, and in some instances their contents may be positively correlated. From a process perspective, however, the association of clays and OM is not controlled by the adsorption of OM within the interlayer region of clay minerals but rather is a byproduct of the mechanical mixing of clays and OM during flocculation and aggregate formation. The intimate mixing of clay and other mineral matter retards microbial degradation of interstitial (between mineral particles) OM and enhances the potential for carbon burial. The interlayer region of smectite and interstratified illite/smectite is too narrow to accommodate the bulk of OM (solid OM) in muddy sediments, and other clay minerals that potentially occur in shales (kaolinite, illite, and chlorite) have limited or no ability to interact with organic molecules. Further study of modern organic-rich sediments (both marine and lacustrine) and flume experiments are needed to provide a better understanding of the roles of transport-driven flocculation and aggregation in the formation of organic-rich sediments and the potential for organic carbon burial. The size, density, architecture, and formation conditions of clay-OM floccules/aggregates will provide important insights into the deposition of organic-rich sediments.

CRediT authorship contribution statement

Bei Liu: Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Maria Mastalerz:** Writing – review & editing, Writing – original draft, Conceptualization. **Juergen Schieber:** Writing – review & editing, Writing – original draft, Conceptualization. **David Bish:** Writing – review & editing, Writing – original draft.

Declaration of competing 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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.orggeochem.2025.105058.

Data availability

Data will be made available on request.

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