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Depositional Heterogeneities and Brittleness of Mudstone Lithofacies in the Marcellus Subgroup, Appalachian Basin, New York, U.S.A.

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Abstract: Organic-rich rocks of the Marcellus subgroup in the study area consist of a diverse suite of mudstone lithofacies that were deposited in distinct facies belts. Lithofacies in the succession range in composition from argillaceous to siliceous, calcareous, and carbonaceous mudstone. Heterogeneities in the succession occurs in the form of varying mineralogical composition, slightly bioturbated to highly bioturbated chaotic matrix, organic-rich and organic-lean laminae, scattered fossil shells in the matrix, and fossils acting as lamination planes. Lithofacies were deposited in three facies belts from the proximal to the distal zone of the depositional system. Bedded siliceous mudstone (BSM) facies occur in the proximal facies belt and consists of a high quartz content in addition to clay minerals and pyrite. In the medial part of the facies belt lies the laminated argillaceous mudstone (LAM), bedded calcareous mudstone (BCaM), and bedded carbonaceous mudstone (BCM). The size of detrital mineral grains in the lithofacies of the medial facies belt is larger than bedded argillaceous mudstone (BAM) of the distal facies belt, characterized by clay-rich matrix with occasional fossil shells and horizontally aligned fossils. Two types of horizontal traces and one type of fecal string characterize the proximal mud-stone facies, whereas only single horizontal trace fossil is found in the mudstones of the medial and distal facies belt. Parallel alignment of fossil shells and fossil lags in lithofacies indicate that bed-load transport was active periodically from the proximal source of the depositional system. Bioturbation has heavily affected all of the lithofacies and presence of mottled burrows as well as Devonian fauna indicate that oxic to dysoxic conditions prevailed during deposition. The deposition of this organic-rich mudstone succession through dynamic processes in an overall oxic to dysoxic environment is different from conventional anoxic depositional models interpreted for most of the organic rich black shales worldwide. Total organic content (TOC) varies from top to bottom in the succession and is highest in BCM facies. The brittleness index, calculated on the basis of mineralogy, allowed classification of the lithofacies into three distinct zones, i.e., a brittle zone, a less brittle zone, and a ductile zone with a general proximal to distal decrease in the brittle behavior due to a decrease in the size of the sediments.

Keywords: mineral composition; organic-rich lithofacies; ichnofossils; brittleness index; marine environment

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Citation: Haq, I.U.; Padmanabhan, E.; Iqbal, O. Depositional Heterogeneities and Brittleness of Mudstone Lithofacies in the Marcellus Subgroup, Appalachian Basin, New York, U.S.A. *Energies* **2021**, *14*, 6620. https://doi.org/10.3390/en14206620

Academic Editor: Pål Østebø Andersen

Received: 26 June 2021
Accepted: 16 September 2021
Published: 14 October 2021

1. Introduction

The exploration and development of hydrocarbons from unconventional shale reservoirs has made tremendous progress in the last decade. Unlike conventional ones, unconventional reservoirs are more expensive to develop and require detailed investigation of the rocks for the consistent production of hydrocarbons. With the advent of new technologies, horizontal drilling and hydraulic fracturing operations have been improved. However, there are still challenges in the effective stimulation of the hydrocarbon reservoirs owing to the heterogeneous and complex nature of the shales [1–5]. Deposition of shales usually takes place in marine, fluvial, or lake environments under low-energy conditions, however, geologically these rocks are not uniform in their vertical and lateral extent. This is because local fluctuations in the energy power of the bottom waters, climatic conditions, sediment types and input rates affect the sediment deposition in various environments. As a result, the fine-grained rocks develop millimeter-scale intervals with varying content of brittle and ductile minerals, organic-poor and organic-rich laminae, fossil lamination/lags, in addition to chaotic fabrics resulting from bioturbation. Physical processes leading to the development of these variations in the fabric of shales have only recently received attention [6–9]. Considerable research has been done on the classification of sandstone and carbonate rock lithofacies, described as a subdivision of a stratigraphic unit that can be mapped and distinguished from their adjacent subdivisions on the basis of lithology. On the other hand, classification of mudstone/shale lithofacies using core and outcrop data is in the initial stages, and various classification schemes have been proposed by different researchers depending on their research objectives [10–13]. Some recent case studies on facies interpretation of shale gas reservoirs, including the Mississippian Barnett shale [12] and upper shale member of the Upper Devonian-Lower Mississippian Bakken Formation [9], provide new and interesting insights into the origin of organic-rich black shales. These rocks, which were initially thought to represent a calm and persistently oxygen-poor (anoxic) environment, may have been deposited in an environment having very low-oxygen level (dysoxic) and moderate current activity [7,9,14]. Depositional environment and physiochemical conditions during the deposition of Devonian Marcellus black shales as discussed by various authors is shown in Table 1.

Table 1. Depositional environment and physiochemical conditions during the deposition of Marcellus black shales, Appalachian Basin, reported in literature.

Reference	Strata	Methods	Depositional Environment/Features	Oxygenation Level of Bottom Waters
Smith et al. [15]	Devonian Marcellus shales, Appalachian Basin	correlation of wireline logs	shallow water conditions that supported benthic life	oxic
Straeten et al. [16]	Devonian Marcellus shales, Appalachian Basin	sedimentological, geochemical, and paleontological context	represent the deepest end members of cycles, on the order of 5–150 m deep	
Chen and Sharma, [17]	Devonian Marcellus shale, Appalachian Basin	geochemical analysis		sub-oxic to anoxic
Emmanuel et al. [18]	Union Spring Formation of Marcellus subgroup, Appalachian Basin	wireline logs, petrographic and geochemical analysis		anoxic to euxinic bottom-water conditions
Kohl et al. [19]	Devonian Marcellus shales, Appalachian Basin	petrographic and mineralogical analysis	deposition in a distal, bathymetrically subdued marine environment	
Smith [20]	Marcellus mudstones, Dunkirk shales, and Utica shale of New York state, Appalachian Basin	correlation of wireline logs	deposition in shallow marine water, no more than a few tens of meters deep	not anoxic

Table 1. *Cont.*

Reference	Strata	Methods	Depositional Environment/Features	Oxygenation Level of Bottom Waters
Boyer and Droser, [21]	Devonian black shales of New York state, Appalachian Basin	paleontological analysis		bottom water oxygen levels were likely a major factor controlling shallow marine ecosystems

Depositional heterogeneities in shale rocks also affect growth of the hydraulic fractures, and production relies primarily on enhanced permeability of the stimulated-reservoir volume (SRV) [22,23]. The permeability and complex network of the hydraulic fractures is more prominent in brittle zones as compared to the ductile zones in a mudstone succession [3,24]. High concentration of brittle minerals such as quartz, feldspar, dolomite, and pyrite enhance the brittle behavior of mudstones as opposed to phyllosilicate minerals that makes a rock to behave in a ductile manner [25]. The design of a hydraulic fracturing stimulation job relies heavily on the extent of brittle behavior and stiffness of the targeted layers in order to stimulate large reservoir volumes in an effective manner. Lithofacies, as a product of a certain depositional and diagenetic realm, offers a reliable framework to investigate the correlation between mineralogy and brittle/ductile behavior of rocks. Thus, investigation of sedimentary processes responsible for small-scale mineralogical and fabric variations are important for a thorough understanding of the depositional conditions and heterogeneities in organic-rich mudstones, given their importance in the petroleum system as well as their growing significance as sites for subsurface CO_2 sequestration.

This study documents the sedimentary processes that were active during deposition of the Marcellus subgroup in western New York (the Oatka Creek Formation and topmost part of the Union Spring Formation) by analyzing mineralogy, organic richness, and depositional features of the mudstone lithofacies. Furthermore, this study also discusses fluctuations in energy power, the level of oxygenation of bottom waters, and variations in brittle and ductile behavior of lithofacies.

2. Geological Setting

The Appalachian Basin went through a series of orogenic events in the Paleozoic era as a result of tectonic collisions between the Laurentia (North American Plate) and the eastern oceanic crust. These tectonic events converted the region to a foreland basin attached to ocean by a narrow seaway, which was a passive margin during the Ordovician period [26]. The Late Silurian to Early Carboniferous Acadian Orogeny being the second of three Paleozoic-age mountain-building events in eastern North America resulted incontinent-continent type collisional tectonics [27,28]. From Middle to Late Devonian, the Appalachian foreland basin was bounded by the Acadian mountains in the east and south, the Cincinnati arch on the west, and the Old Red sandstone continent to the north. Moreover, it was connected to the Theic Ocean by a narrow and long seaway in the south-west, forming a nearly enclosed epicontinental sea [27,29]. Tectonic loading, stemming from the second orogenic event coupled with eustatic sea level rise, terminated shallow-shelf carbonate deposition during the Early Devonian and led to the accumulation of several organic-rich shale units in the basin [30,31]. The term "Marcellus subgroup" of the Hamilton group was used by the New York Devonian researchers [28,32], and includes the Union Spring Formation overlain by coeval Oatka Creek and Mount Marion formations. These units of the Marcellus subgroup cover most of the basin with an area of approximately 500,000 km^2 (Figure 1).

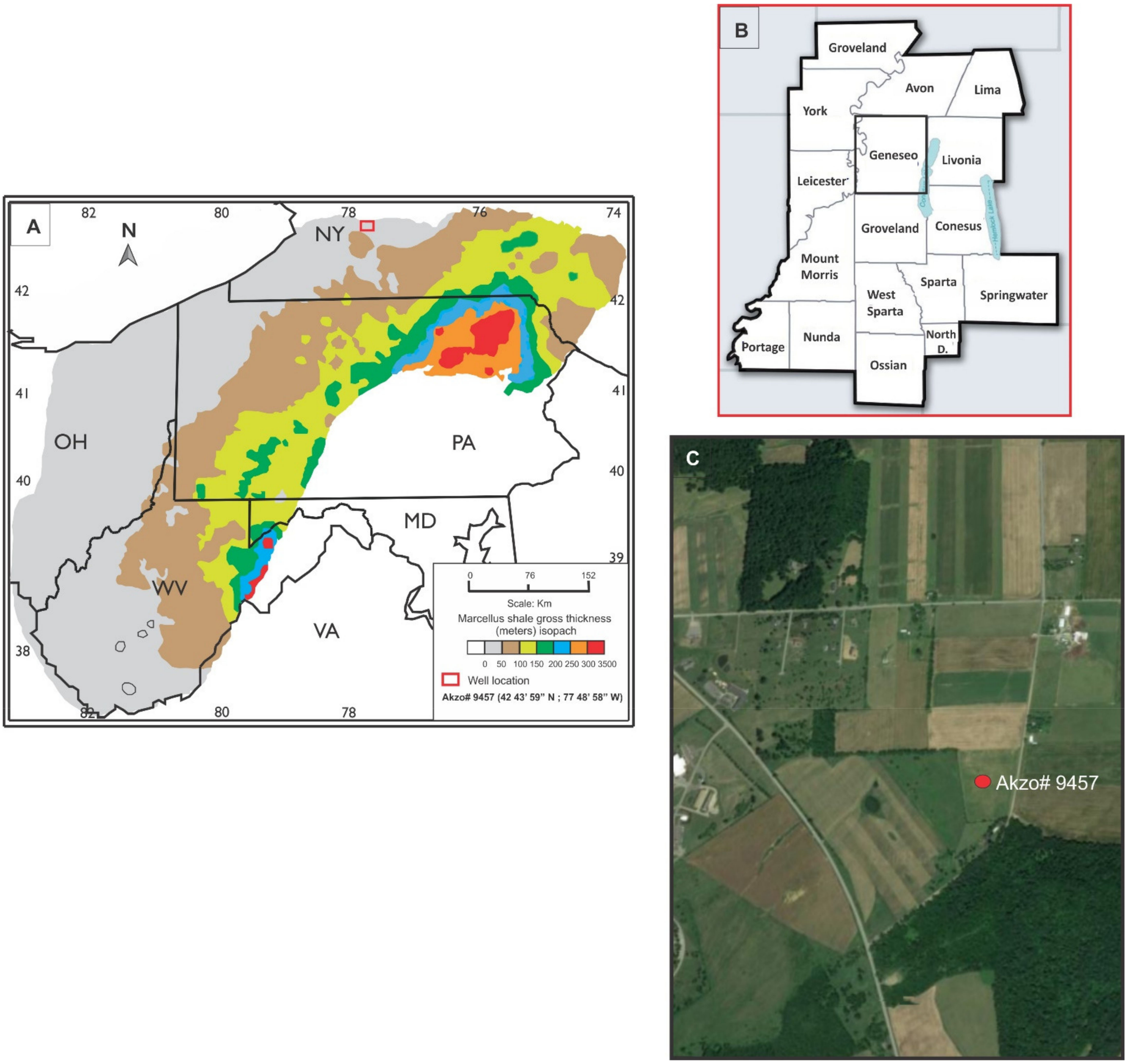


Figure 1. (**A**) Map of the Appalachian Basin showing the gross thickness (in meters) of the Marcellus subgroup strata in the subsurface. The rectangle in red shows the location of the examined well in New York State, USA [33]. (**B**) Livingston County map in New York showing the town of Groveland where the studied well was drilled. (**C**) Map showing detailed location of the Akzo# 9457 in the town of Groveland.

3. Methods

Characterization of the drill core samples from exploratory wells within the Marcellus subgroup was done by TOC estimation, mineral composition and lithofacies analysis for describing the depositional setting and rock brittleness.

3.1. Data and Analysis

Data for this study comprises drill core samples from the Akzo# 9457, drilled in western New York State, representing two formations of the Marcellus Subgroup, i.e., the Oatka Creek Formation and the Union Spring Formation. To document the sedimentological facies distribution, from base to top in ten meters interval, microscopic, mineralogical, and geochemical analyses were carried out for all the samples. After examining the core slabs under a binocular microscope, one half of the sample was used for making polished thin sections while on the other half for X-ray diffraction (XRD) and Total organic carbon (TOC) analysis were undertaken. Thin sections were studied in transmitted light and cross polar modes using a Leica petrographic microscope. The grades used to plot the intensity of the bioturbation in this study follows that proposed by Taylor and Golding [34]. These grades are expressed in numbers that range from 0 (no bioturbation) to 6 (complete bioturbation). For XRD analysis, the rocks were first crushed using a mortar and pestle, sieved, oven dried at 58 °C, and then a 1 g powdered sample was used to determine the weight percent of minerals present in the shale matrix. Prior to TOC analysis, powdered samples were treated with dilute hydrochloric acid (HCL) to remove the carbonate content and to avoid the effect of inorganic carbon on the results.

The brittleness index of lithofacies was calculated on the basis of mineralogy to determine the brittle/ductile behavior of individual lithofacies and delineate the brittle from ductile zones in the studied interval. Shales are usually deposited under low-energy conditions with local fluctuations and later during burial these rocks experience a series of unique diagenetic alteration reactions. As a result, the amount of ductile and brittle minerals varies in shale rocks. Therefore, the concept behind calculating the rock brittleness from mineralogy is still the same as was proposed by Jarvie [25]. In this study the equation proposed and used to calculate the brittleness index of the rocks is based on the Mohs scale of hardness of minerals by considering calcite (having a hardness of 3 on the Mohs hardness scale) half as brittle as quartz, feldspar and pyrite.

$$\text{Brittleness index (BI)} = \frac{\text{quartz} + \text{feldspar} + \text{carbonate}/2 + \text{pyrite}}{\text{quartz} + \text{feldspar} + \text{carbonate} + \text{clay} + \text{pyrite} + \text{TOC}} \quad (1)$$

3.2. Classification Criteria of Lithofacies

Lithofacies were defined as the sum of all the features linked to lithology such as mineral components, stratification, color, texture and grain size [35]. Lithofacies in general refer to the association of rocks that reflects deposition under certain sedimentary depositional environments. Lithofacies classification of sandstones and carbonate rocks are not challenging like shale rocks. Shales usually being deposited under relatively calm and low-energy conditions a have fine size of mineral grains, uniform texture, stratification, and are mostly rich in organic material. Numerous efforts have been made to classify the shale lithofacies which resulted in different classification schemes [9,11,36–39].

The evaluation of shales hydrocarbon potential by gas sorption mechanism in shale reservoirs largely depends on its mineralogy and organic richness. Secondly, in addition to variations in the pore pressure and subsurface stress profiles, fracture growth in the shale reservoir is also closely related to depositional heterogeneities in a succession, as these can strongly influence the stimulation process of the targeted interval. To evaluate these micro-scale quantitative aspects, lithofacies classification is important in successful completion of the fracking operation and in prolonging production from shale gas reservoirs. The shale lithofacies in this study were identified based on mineral composition, bedding thickness, and organic richness (Figure 2).

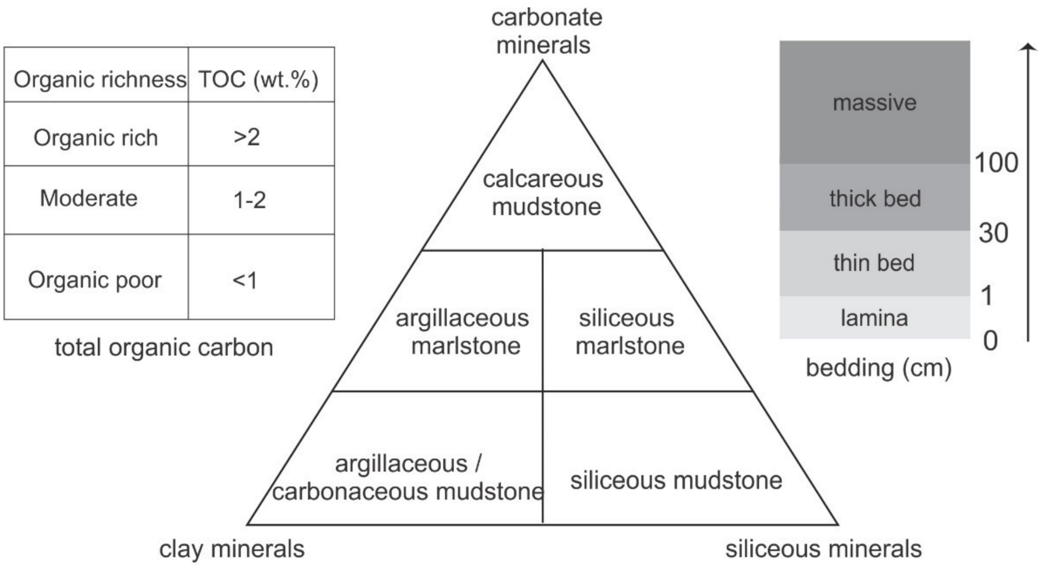


Figure 2. Classification scheme for shale lithofacies based on TOC [10], mineralogy, and bed thickness.

4. Results

4.1. Minerology, TOC and Facies Distribution

The data in this study is from Akzo# 9457, at a total depth from 255 m to 265 m and is presented in the log. The log is showing the distribution of lithofacies in both the formations of Marcellus subgroup together with observed macroscopic and microscopic sedimentary features (Figure 3). A detailed description of minerology, TOC and brittleness index is shown in Figure 4. The mineralogical quantification is using the X-ray diffraction method, while TOC is calculated by the combustion of treated powder using a TOC analyzer.

4.2. Ichnological Elements

Two types of burrows and one type of fecal strings were identified in the mudstones of the Oatka Creek Formation, while the mudstones of the Union Springs Formation show only one type of burrow. These ichnofossils are termed herein as facies elements because the detailed taxonomy is out of the scope of this study. Horizontal burrows are comprised of unbranched, lenticular structures readily distinguishable from the surrounding rock matrix on the basis of shape and color. These horizontal burrows are somewhat similar in appearance to Planolites [9,40,41]. The lens-shaped burrows, herein referred to as Planolites, are elliptical in shape having sharp margins and are 0.3 to 0.7 mm across, commonly found lying parallel to the bedding plane (Figure 5A). The filling material of the Planolites are similar to that of the host rock matrix, except that they lack organic matter and pyrite. The second type of burrows, herein referred to as mottled burrows, are randomly oriented with a highly mottled tube-like appearance (Y-shaped, I-shaped, U-shaped) indicating intense bioturbation (Figures 5A–F and 6B). The length of these mottled burrows varies from a few to several millimeters while their filling material is preferentially similar in composition to the matrix of host rock. The fecal strings are wavy and irregular in shape, 0.1 to 0.3 mm across, and lie parallel to the bedding plane in the horizontal position (Figure 5B). These fecal strings are herein referred to as Phycosiphon incertum [42], and they are dark brown in color and are noticeably restricted to the facie rich in siliceous sediments with low organic content.

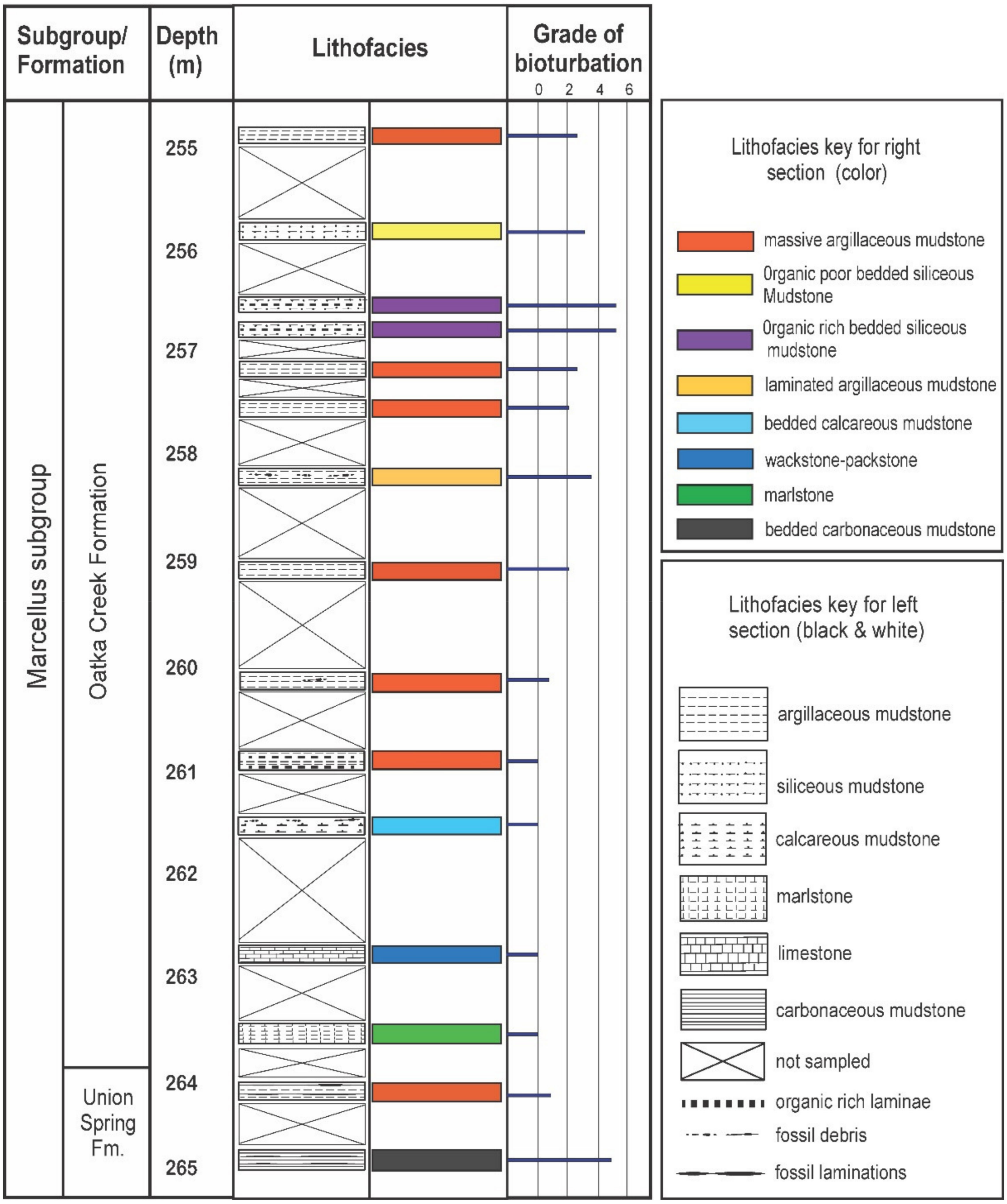


Figure 3. Log showing the lithofacies distribution and the grade of bioturbation (GB).

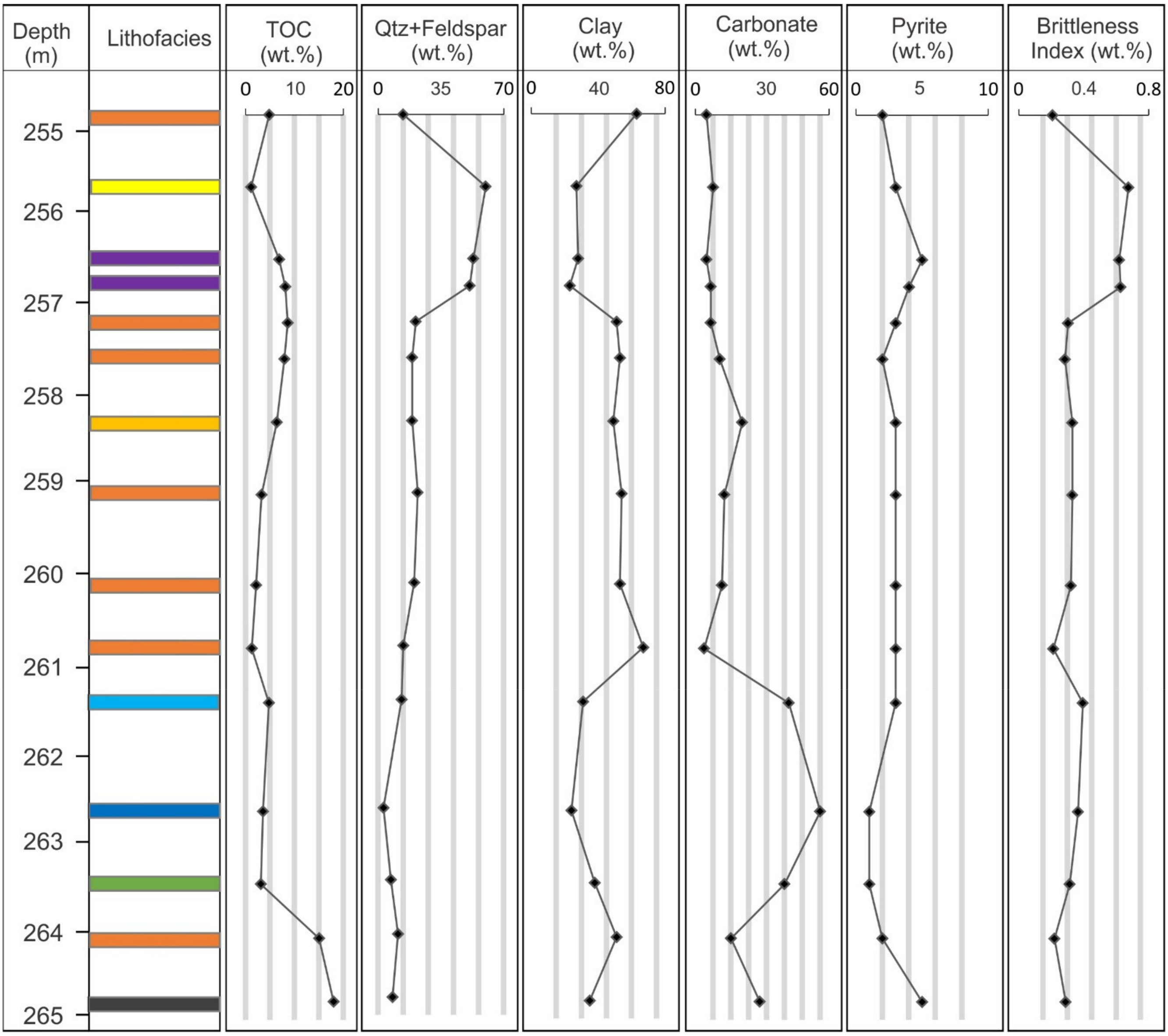


Figure 4. Total organic carbon (TOC), mineralogical (quartz, feldspar, clay, carbonate, and pyrite) and brittleness index (BI) profiles of the lithofacies in the studied interval.

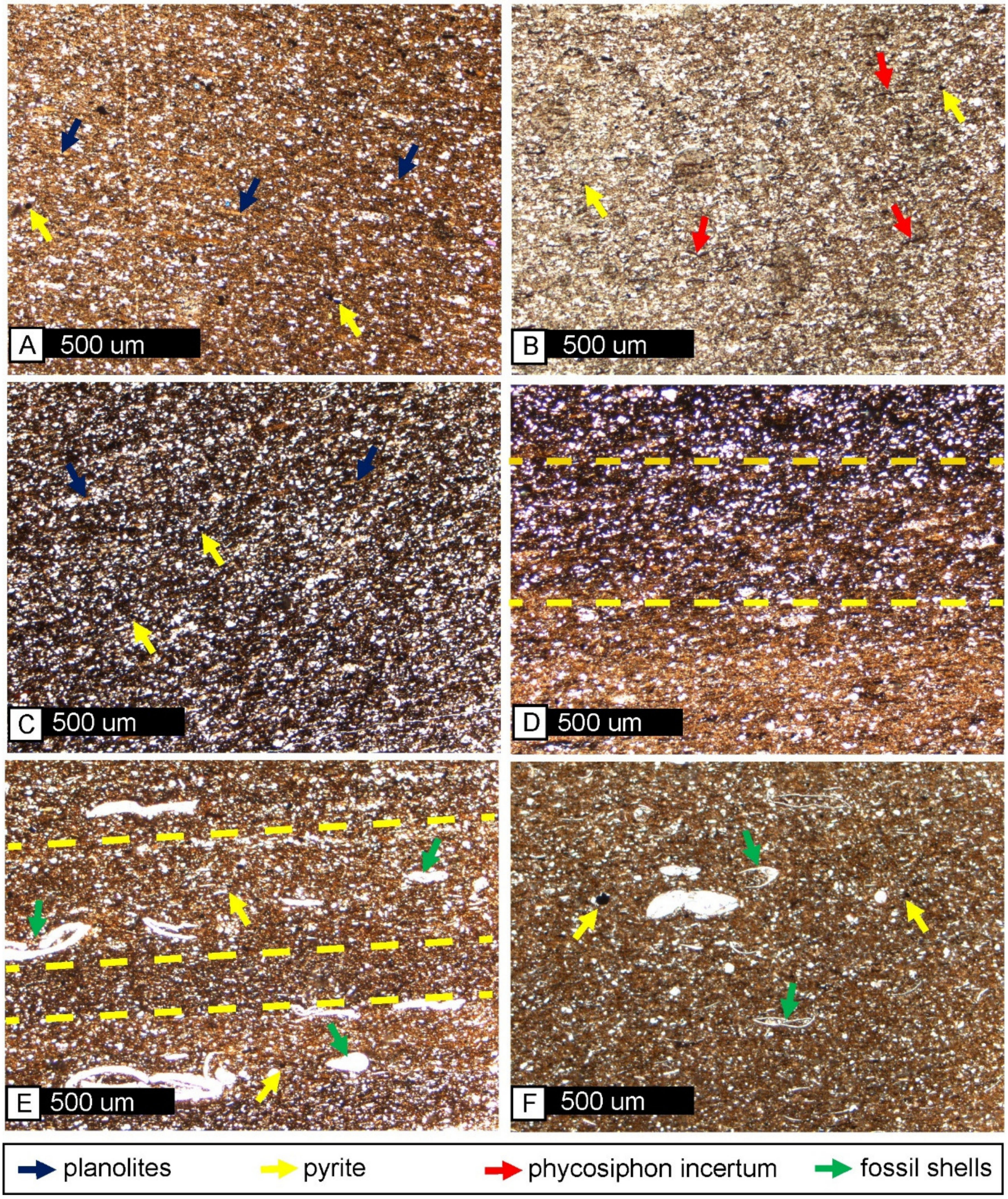


Figure 5. Plane light photomicrographs of mudstone lithofacies in the Oatka Creek Formation. (**A**) Bedded argillaceous mudstone (BAM). (**B**) Organic-poor bedded siliceous mudstone (Op-BSM). (**C**) Organic rich bedded siliceous mudstone (Or-BSM). (**D**) Gradational contact between argillaceous mudstone and organic-rich siliceous mudstone. (**E**) Laminated argillaceous mudstone (LAM). (**F**) Bedded argillaceous mudstone (BAM).

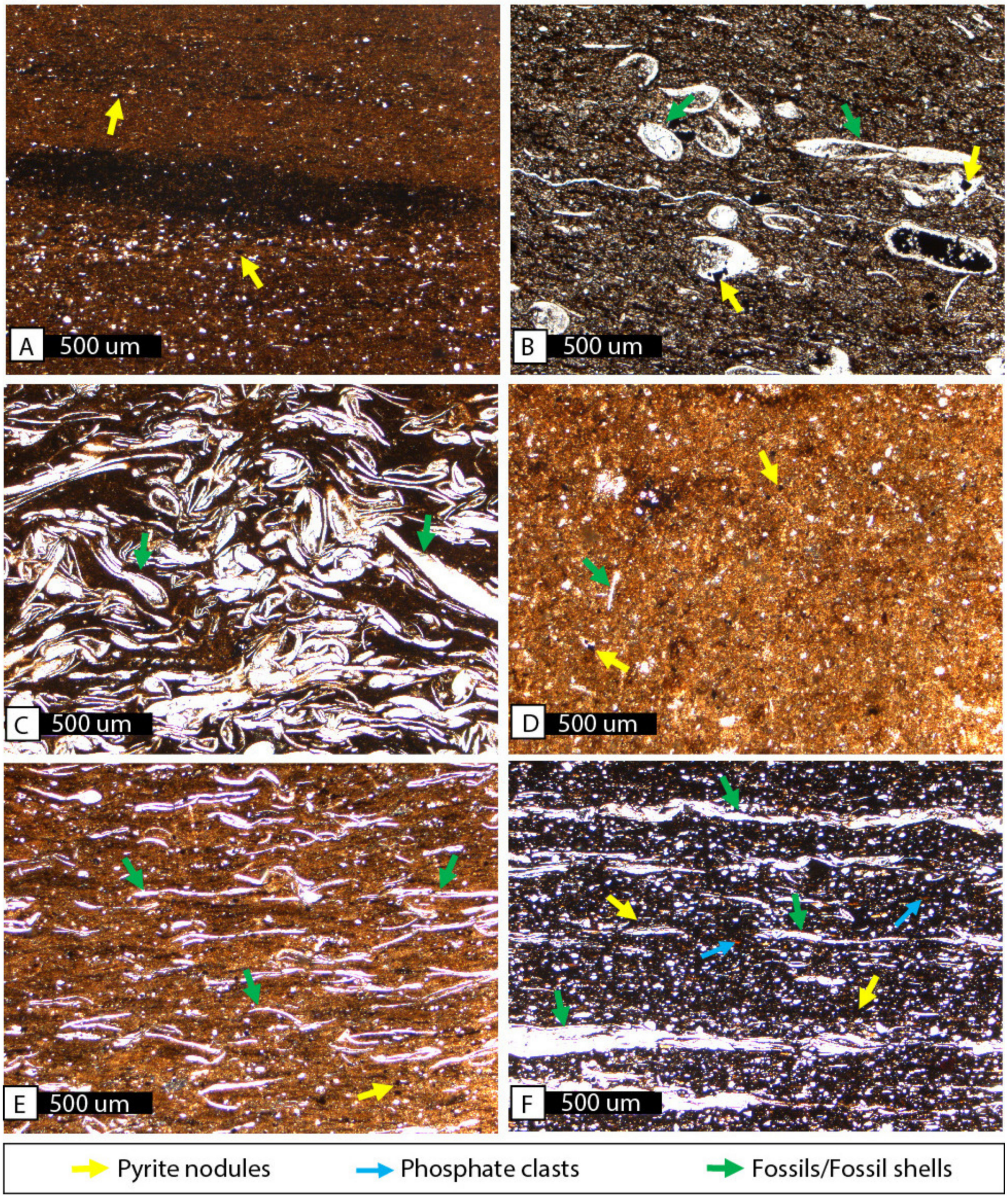


Figure 6. Plane light photomicrographs of the mudstone and carbonate lithofacies in lower part of the Oatka Creek Formation (**A**–**D**) and upper part of the Union Spring Formation (**E**,**F**). (**A**) Bedded argillaceous mudstone (BAM) facie not affected by bioturbation. (**B**) Bedded calcareous mudstone (BCaM). (**C**) Wackstone-packstone facies. (**D**) Bedded marlstone facies. (**E**) Bedded argillaceous mudstone (BAM) facies. (**F**) Bedded carbonaceous mudstone (BCM) facies.

4.3. Lithofacies

4.3.1. Lithofacies of the Oatka Creek Formation

Lithofacies in the Oatka Creek formation include: (1) bedded argillaceous mudstone (BAM), (2) bedded siliceous mudstone (BSM), (3) laminated argillaceous mudstone (LAM), (4) bedded calcareous mudstone (BCaM), and (5) carbonate facies i.e., marlstone and wackstone-packstone.

Bedded Argillaceous Mudstone (BAM)

The BAM lithofacies are found at various depths in the ten meter long interval (Figures 5A,D and 6A). The rock matrix in this case is rich in clay (avg. 57 wt.%), with minor silt size quartz grains distributed randomly throughout the matrix. Average quartz content is 18 wt.% while average carbonate content is 7 wt.%. The angular to sub-rounded silt grains are mostly of uniform size and are displaced in the entire matrix, where index of bioturbation is high (Figure 5A). At depths, in the Oatka Creek Formation, the matrix of this lithofacies is rich in organic material and pyrite but lacks fossils. The TOC content of this lithofacies is highly variable, from as low as 1.23 wt.% to a maximum of 8.63 wt.%. Planolites, together with mottled burrows (Figure 5A) are present at certain depths while there is no indication of trace fossil activity at other intervals. The bioturbation index of the BAM lithofacies varies between 0 and 3.

Bedded Siliceous Mudstone (BSM)

The matrix of the BSM consists of silt size angular to sub-angular sediments in the clayey matrix. The percentage of silt grains (avg. 54 wt.%), that are predominantly quartz, typically or always exceed the volume percent of clay in these lithofacies. These are further categorized as organic-poor (Op) BSM (Figure 5B) and organic-rich (Or) BSM (Figure 5C). The Or-BSM has an average TOC value of 7.49 wt.% and grades upward into Op-BSM with TOC value of 1.11 wt.%. The matrix of both Or-BSM and Op-BSM is extensively bioturbated, indicated by dark-colored mottled burrows. This reworking has destroyed all the structural (lamination) features. The filling material in the reworked areas is the same as that of the original rock matrix in both Op and Or-BSM. Pyrite crystals are dispersed throughout the matrix in both the cases and are readily identified from the typical shiny black luminescence under reflected light. Phycosiphon incertum are found, restricted to Op-BSM (Figure 5B), lying parallel to the bedding with a filling material of dark brown color. The grade of bioturbation varies from 3 in Op-BSM to 5 in Or-BSM.

Laminated Argillaceous Mudstone (LAM)

The matrix in the LAM lithofacies consists of silt-sized quartz and carbonate grains embedded in a clay (49 wt.%) supported matrix and up to several micrometer-long fossils and fossil shells. Quartz content is 23 wt.% while carbonate content is 15 wt.%. Lamination is clear and visible in the form of light brown clay-rich layer, a more organic rich dark brown clay-rich layer and a silt-rich layer (Figure 5E). These layers are intercalated with each other in a periodic fashion suggesting local fluctuations in the energy level and sediment input. Silt grains in clay-rich lamina are sub-rounded to angular in shape and are mostly of uniform size, whereas the size of the grains in the silt-rich lamina is variable. Internally, there is no apparent grading or sedimentary structures in these lithofacies. Devonian micro-fauna aligned parallel to the bedding is restricted to the silt-rich lamina in the matrix. The matrix also hosts abundant pyrite nodules; however, the concentration of the pyrite appears greater in the organic-rich clay laminae. The average TOC value of these lithofacies is 8.35 wt.% and the matrix is reworked to a certain extent by the mottled burrows resulting in displacement of some silt grains into the adjacent layers. The grade of bioturbation for the LAM lithofacies is 3 to 3.5.

Bedded Calcareous Mudstone (BCaM)

The matrix of the BCaM is rich in calcite (avg. 42%) with silt-size quartz grains dispersed randomly. Quartz content is 13 wt.% while clay content is 31 wt.%. Fossils and fossil shells are found, embedded in the clayey matrix, and aligned parallel to the bedding (Figure 6B). These fossils are mostly Cornulites (benthic, often encrusting, conical organisms) and some unidentified species of the Styliolina. The original composition of these fossils is not solely altered after deposition, the calcareous rims of the fossils are still intact and unaltered; however, the core has been altered to some extent as can be seen partially replaced by clay, microcrystalline quartz (authigenic) and pyrite minerals. Pyrite nodules, replacing calcite in these fossils, are of much greater size as compared to the rest of the matrix and present in the form of aggregates (Figure 6B). The TOC content of the samples analyzed from this lithofacies is 4.74 wt.% and no trace fossil activity is observed. The grade of bioturbation of BCaM lithofacies is 0.

Bedded Carbonate Facies

Wackstone-packstone facies are highly fossiliferous, and thematrix is filled with dark brown to black color organic-rich clay material. The fossils, mostly Styliolina, are non-aligned to bedding and appears like thick lags deposited during a stormy event (Figure 6C). The organic-rich clay material is acting as cement, holding fossils inside the matrix. The filling of these fossils is mostly unaltered and reflect the original calcic composition; however, sometimes replaced by organic-rich clay material after dissolution of the calcite. TOC of this facie in the Oatka Creek Formation is 3.54 wt.% and grade of bioturbation is 0.

4.3.2. Lithofacies in Union Spring Formation

Bedded Argillaceous Mudstone (BAM)

The matrix of the BAM is rich in clay minerals comprising to 51 wt.% with minor silt-size quartz and calcite grains distributed randomly. The quartz and carbonate content of the facies is 11 wt.% and 16 wt.% respectively. Pyrite, which accounts for 2 wt.% is scattered in the form of nodules throughout the matrix. The facies host various species of the Styliolina is aligned parallel to the bedding plane in the rock matrix (Figure 6E). The TOC content of this lithofacies, resting directly above the organic-rich bedded carbonaceous mudstone (BCM), is 15.05 wt.% and grade of bioturbation is 1.

Bedded Carbonaceous Mudstone (BCM)

BCM is rich in clay (35 wt.%) and organic material in the Union Spring Formation, with fossils aligned parallel to the bedding. Silt-size rounded to sub-rounded quartz (8 wt.%) and carbonate (29 wt.%) grains are dispersed in the matrix. Phosphate clasts, together with some dolomite rhombs and pyrite (5 wt.%) crystals, are also found. Fossils are thin Rhynchonelliformea brachiopods of the superfamily Camarotoechioidea, the Styliolina, and possibly some ostracods. The length of these fossils is from 1 to 2 mm and thickness varies from 0.1 mm to 0.4 mm. Due to the plane shape of these fossils they are running parallel to the bedding in the organic-rich matrix. The TOC content in these lithofacies reaches up to 18 wt.%. The effects of bioturbation are evident in the form of mottled burrows. The filling material (silt grains) of these mottled burrows, mostly sub-vertical, is the same as that of the host matrix. The grade of bioturbation for the BCM lithofacies ranges between 4 and 5.

4.4. Total Organic Carbon (TOC) Analysis

The profiles of TOC values of the lithofacies reflect systematic variations in the organic material enrichment. The minimum and maximum TOC values in the examined 10 m interval are 1.11 wt.% and 18.02 wt.% respectively. The organic-poor siliceous mudstone (OpBSM) in the Oatka Creek Formation has the lowest value of TOC, while TOC is highest in the Union Spring Formation, i.e., up to 18.02 wt.% in the bedded carbonaceous mudstone (BCM) facies. The values of TOC in the bedded carbonate facies, i.e., marlstone and wackstone-packstone that were identified at base of the Oatka Creek Formation are 3.11 wt.% and 3.54 wt.%, respectively. The organic carbon content of the clay-rich argillaceous mudstone facies is not uniform and ranges from 1.23 to 15.05 wt.%. The TOC of shale rocks has been related to the mineralogy and porosity of rocks/facies in many case studies, including the formations of the Marcellus subgroup [11,33,43–45]. However, the TOC content of the lithofacies analyzed in this study depends entirely on the enrichment of organic material in the matrix, and no correlation is found between increase and/or decrease of the TOC content with the abundance of clay, quartz, and carbonate minerals in the rock matrix (Figure 7).

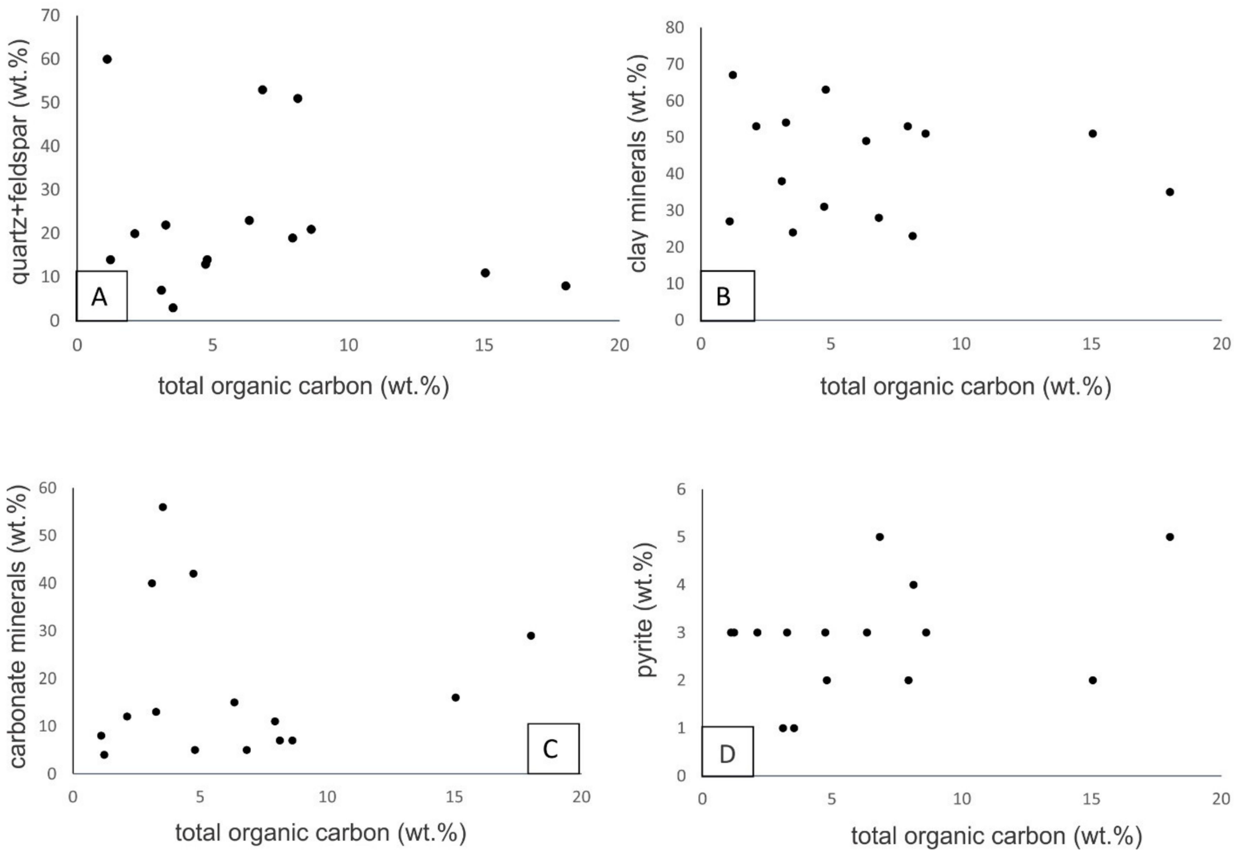


Figure 7. Correlation between total organic content (wt.%) and rock mineralogy (wt.%). (**A**) Correlation of total organic carbon with quartz and feldspar content. (**B**) Correlation of total organic carbon with clay minerals. (**C**) Correlation of total organic carbon with carbonate minerals. (**D**) Correlation of total organic carbon with pyrite content.

4.5. Brittleness Index of Lithofacies

The equation used in this study to calculate the brittleness index assumes that the brittleness of the rock is a response of abundance of brittle minerals compared to ductile minerals in a rock matrix. The brittle minerals here are defined as minerals having a hardness equal to or greater than 4 on the Mohs scale, while ductile are referred to as minerals having a hardness less than 3 on the Mohs scale. Calcite, which has a hardness of 3, is defined as a mineral intermediate between brittle and ductile.

Brittleness index delineated in this study shows that facies having a higher concentration of quartz, pyrite and calcite are brittle as compared to clay minerals and organic-rich facies. Bedded siliceous mudstone (BSM), both organic-poor and organic-rich, has the highest brittleness index followed by the bedded calcareous mudstone (BCM) and wackstone-packstone facies. Laminated argillaceous mudstone (LAM), bedded carbonaceous mudstone (BCM), bedded argillaceous mudstone (BAM), and marlstone shows ductile behavior among all the identified lithofacies.

5. Discussion

Sedimentological studies provide important clues to infer the depositional architecture and physiochemical conditions during the deposition of the shale successions. In order to distinguish proximal fine-grained sediments from the distal in shale/mudstone sequences, Egenhoff and Fishman [9] proposed a criterion based on the depositional trends of the facies. On the basis of this criteria, mudstone facies hosting phosphate clasts, large amounts of fossil shell fragments, erosional surfaces and coarser grains reflect deposition in a proximal environment, whereas facies dominated by fine-grained sediments (argillaceous/carbonaceous) are indicative of deposition in distal or low energy environments.

For the organic-rich shales of the Marcellus subgroup both shallow and deep-water hypothesis of deposition has been proposed. The deep-water model, as discussed by Lash and Engelder [46], is debated widely, but a wide range of sedimentological, paleontological data argue against this model [15,18].

5.1. Depositional Model from Petrographic Evidence

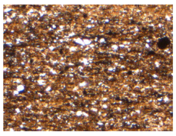
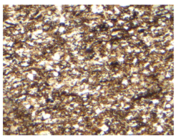
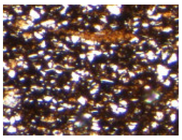
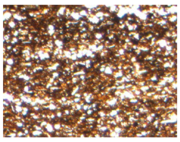
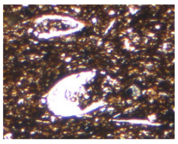
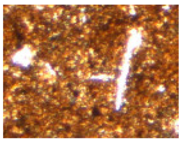
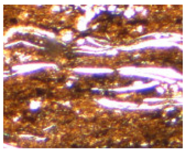
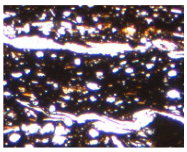
The occurrence of sedimentary structures in mudstone/shale lithofacies such as graded contacts, burrows, reworking of authigenic minerals, aligned fossils, basal fossil lag deposits, and the presence and enrichment of silt grains even in organic-rich facies are not typical of the deep-water depth. They are indicative of current activity [18].

Marlstone facies encountered at the base of the Oatka Creek Formation lie directly above the unconformity, overlying the bedded argillaceous mudstone (BAM) of the Union Spring Formation. This carbonate-rich facie is the result of the third-order relative sea level fall after deposition of the organic-rich mudstone facies of the Union Spring Formation [8]. The wackstone-packstone facie is the result of the fourth-order sea level fall [8]. Both these carbonate facies identified in the Oatka Creek Formation, i.e., marlstone and wackstone-packstone, constitute the Hurley and Cherry Valley members of the Oatka Creek Formation, respectively. These lithofacies represent deposition at the proximal ends in the depositional system. Variations in the mineralogy, organic carbon content, ichnofossils, and thickness of the mudstone lithofacies identified in the Oatka Creek Formation suggest fluctuations in the sea level, as well as proximity to the clastic source.

Argillaceous mudstone facies of the Oatka Creek Formation vary from laminated to bedded, hosting minor fossils/fossil lags, and scattered but aligned shell debris at some intervals. Bedded calcareous mudstone facies (BCaM) lie on the top of the Cherry Valley Limestone and are rich in calcite, having fossils aligned parallel to the bedding in the matrix (Figure 6B). Bioturbation has not affected the matrix, suggesting that sedimentation was continuous during that period with abundance of carbonate input from the source area. High sedimentation rate and a compact fabric of BCaM suggest that this facie was deposited during the time of early transgression, but after the deposition of carbonate facies in the Oatka Creek Formation. Bedded argillaceous mudstone facies (BAM) of the Oatka Creek Formation are characterized by silt-size quartz and carbonate grains, and has finer size than grains present in LAM. Occasional fossils in the clay-rich bioturbated matrix (Figure 5F) suggest deposition in the distal part of the facies belt, where currents envisioned to have slowed down carrying lighter carbonate fossils with minor silt and clay floccules. The inter-laminated nature of the laminated argillaceous mudstone (LAM) (Figure 5E) suggest deposition under low energy conditions. Most likely, the deposition of LAM occurred in the medial to distal part of the depositional system, with input from the mixed siliciclastic and carbonate sediments from the areas close to the paleoshoreline.

Organic-rich and organic-poor bedded siliceous mudstone facies (OrBSM, OpBSM), characterized by coarse silt-size quartz grains, reflect deposition under uniform and slightly high-energy conditions near to the proximal part of depositional belt (Table 2). The presence of burrows in these facies indicate that sedimentation was not continuous and low sedimentation rate allowed the burrowing organisms to disrupt the sedimentary structures giving the facies' a chaotic appearance. Variation in the organic material input of these two facies and gradational contact with massive argillaceous mudstone facies (Figure 5D) strongly reflects rapid fluctuations in the material input from the source area and is attributed to tectonics and change in climatic conditions during their deposition.

Table 2. Facies in the Oatka Creek Formation and Union Spring Formation.

No.	Facies	Description	Facies Appearance	Location	Interpretation
1	BAM facies (mudstone) (Oatka Creek Formation)	bedded argillaceous mudstone horizontal planolites burrowed		middle and upper part of the Oatka Creek Formation	distal part of the system dysoxic conditions clayey and ductile
2	Organic lean BSM facies (mudstone) (Oatka Creek Formation)	bedded siliceous mudstone organic-poor Phycociphon incertum burrowed		upper part of the Oatka Creek Formation	proximal part of the system oxic conditions silty and brittle
3	Organic rich BSM facies (mudstone) (Oatka Creek Formation)	bedded siliceous mudstone organic-rich horizontal planolites burrowed		upper part of the Oatka Creek Formation	medial part of the system oxic to dysoxic conditions silty and brittle
4	LAM facies (mudstone) (Oatka Creek Formation)	laminated argillaceous mudstone fossils aligned to bedding burrowed		middle part of the Oatka Creek Formation	medial part of the system oxic to dysoxic conditions clayey and ductile
5	BCaM (mudstone) (Oatka Creek Formation)	bedded calcareous mudstone fossils and fossils shells pyrite replacement		middle part of the Oatka Creek Formation	medial part of system oxic to dysoxic conditions fossiliferous and moderately brittle
6	Wackstone-packstone (limestone) (Oatka Creek Formation)	wackstone-packstone organic-rich fossiliferous		basal part of the Oatka Creek Formation	proximal part of the system oxic conditions fossiliferous and moderately brittle
7	Marlstone (limestone) (Oatka Creek Formation)	marlstone calcified bivalve components		basal part of the Oatka Creek Formation	proximal part of the system oxic conditions ductile
8	BAM facies (mudstone) (Union Spring Formation)	bedded argillaceous mudstone fossils aligned to bedding		upper part of the Union Spring Formation	distal part of the system dysoxic conditions clayey and ductile
9	BCM facies (mudstone) (Union Spring Formation)	bedded carbonaceous mudstone fossils aligned to bedding burrowed		upper part of the Union Spring Formation	medial part of the system dysoxic conditions organic-rich and ductile

Bedded carbonaceous mudstone (BCM) facies of the Union Spring Formation were deposited by bed-load processes. These processes supplied the sediments from shallow waters to the medial part of the basin during stormy events together with suspension sediment settling. This is indicated by thick pile of fossil lags aligned horizontally to the bedding in organic-rich matrix of BCM facies of the Union Spring Formation (Figure 6F). Although bioturbation has destroyed much of the original sedimentary features in BCM facies of the Union Spring Formation, phosphate clasts and mixed size of calcite and quartz grains suggest that this facie was deposited in the medial part of the system. On the other hand, presence of parallelly aligned fossils indicates that bed load transport was active periodically from the proximal end of the depositional system. In the upper part of the section, bedded argillaceous mudstone (BAM) facies of the Union Spring Formation host fine-silt particles in a clay-rich matrix. Fossils in this facie are also aligned parallel to the bedding; however, these fossils, i.e., various species of Styliolina, are different from those deposited in the BCM in shape and structure (Figure 6E). These features of BAM suggest deposition in a relatively deeper setting and a further transgression of the sea level after the deposition of BCM facies in the Union Spring Formation (Figure 8).

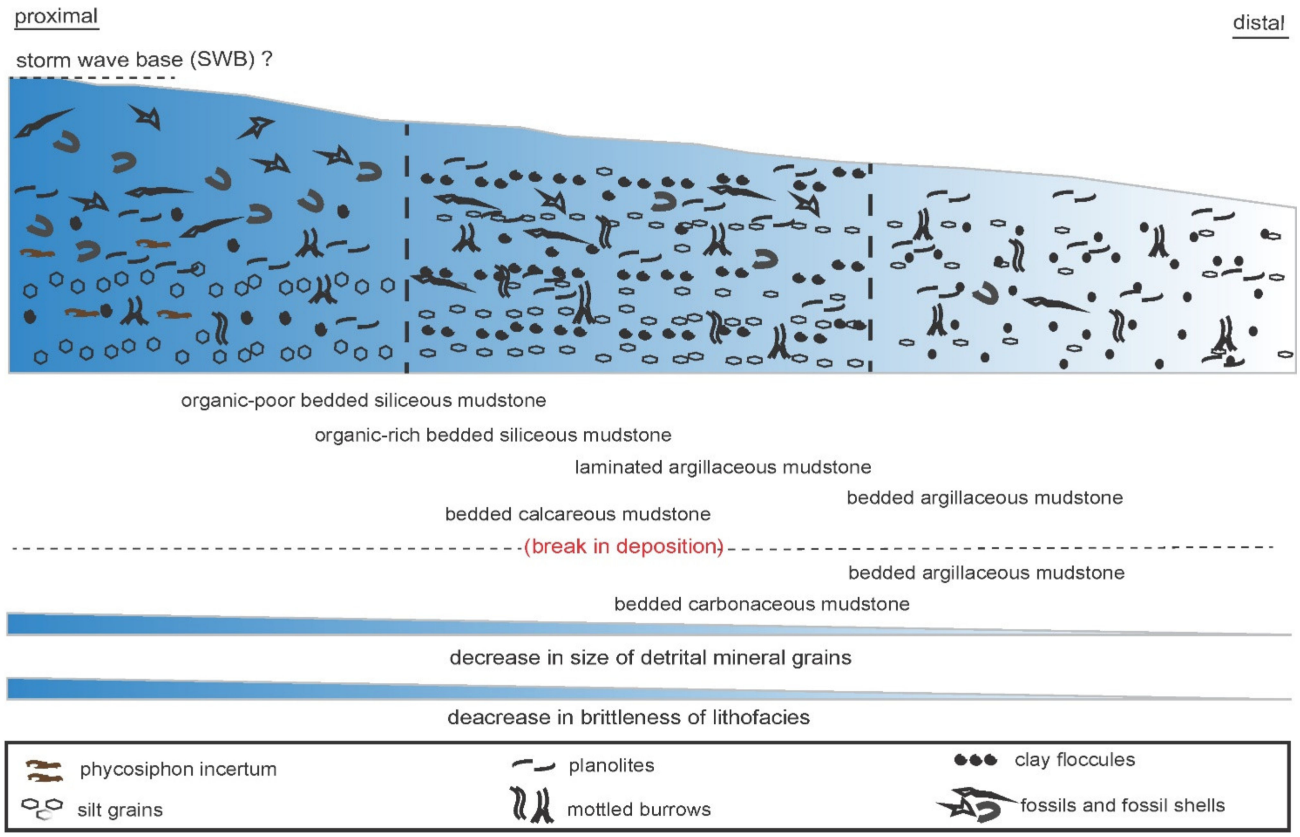


Figure 8. Conceptual model showing the distribution of mudstone lithofacies and burrow types along the facies belt. Note the decreasing trend in grain size of detrital mineral grains and brittleness of the lithofacies from the proximal to the distal zone [9].

5.2. Physiochemical Conditions during Deposition

Small-scale lamination, organic richness, fossil debris/lags, and the highly bioturbated nature of the studied lithofacies reflects the physio-chemical conditions during deposition. The grade of bioturbation of organic-poor as well as organic-rich bedded siliceous mudstone (BSM) facies ranges from 3 to 3.5, suggesting that oxygen supply to the bottom waters was sufficient during the deposition of these facies. Argillaceous mudstones, including laminated and bedded, are also affected by bioturbation. The organic-rich laminae in laminated argillaceous mudstone (LAM) have recorded high bioturbation activity as compared to the organic-poor laminae (Figure 5E). This suggests a favorable living environment for the burrowing organisms feeding on nutrients from the organic material. The presence of the mineral pyrite in the lithofacies indicates that a low oxygen environment prevailed in the system aiding pyrite formation by the pyritization of organic matter in the burrow fills and pyrite framboids replacing the calcite/quartz cement in the fossil cores. Carbonate facies in the Oatka Creek Formation deposited during the sea level fall, whereas bedded calcareous mudstone (BCaM), deposited during the early transgression period after the deposition of wackstone-packstone, lack signs of bioturbation.

The oxic and dysoxic bottom waters could persist between blooms of organic growth and deposition [14]. The diversity of fossils and high grade of bioturbation together with pyrite in the bedded carbonaceous mudstone (BCM) facies of the Union Spring Formation indicates that dysoxic conditions most probably were more persistent in the bottom waters or in the sediments just beneath the sediment-water interface. Infiltration of oxygenated bottom waters into the underlying sediments might have provided a mechanism of survival for the burrowing organisms responsible for mottled burrows in mudstone facies that are rich in organic material. The mottled burrows recorded in these lithofacies might also be alternatively interpreted to represent fluid-escape structures [47]; however, typical 'U' and 'Y' shapes of the bioturbation structures are indicative for faunal activity during sediment deposition. The interpretation of documented burrowing is in accordance with the recent findings of burrows in organic-rich shales [7–9,14].

5.3. Input to the Fracking Design

Effective and successful fracking of shale reservoirs is achieved by stimulating the reservoir to a safe maximum extent. This is reflected in micro-seismic maps and production history by a very slight decline in hydrocarbon production with time. Therefore, the identification of brittle and ductile zones in shale reservoirs is therefore important to the design of a hydraulic fracturing operation. Although hydraulic fracture growth depends on several factors, including the minimum principal stress direction, difference in the horizontal stress magnitudes and perforation design, reservoir zones that are brittle enough aid in the successful vertical and lateral growth of hydraulic fractures resulting in the maximum stimulation of the reservoir. It has been reported in the literature that the brittleness index of the rocks calculated using mineralogical composition of the rocks relates closely with the brittleness index calculated using elastic parameters of the rocks [3,48].

The brittleness index of the facies was calculated using Equation (1) and is shown in Figure 4. These facies were then assigned to different zones of brittleness. The bedded siliceous mudstone facies, both organic-rich and organic-poor, fall in the brittle zone, while all the other lithofacies, including carbonate lithofacies, lie in the ductile zone (Figure 9C). Bedded argillaceous mudstone (BAM) and bedded carbonaceous mudstone (BCM) are the least brittle among all the lithofacies, because they are predominantly composed of ductile material, i.e., clay minerals and organic material. The design of the reservoir stimulation via hydraulic fracturing will vary accordingly keeping in view the ductile and brittle behavior of lithofacies, as identified in this study, together with strength and elastic behavior from geomechanical characterization.

The limitation of this study includes the following: (1) the study is based on well data only available at discrete stratigraphic intervals; and (2) the non-availability of standard core samples for geomechanical property evaluation. For future work, it is recommended to carry out detailed petrographic and geochemical analysis on the Devonian succession of the Appalachian basin southward in the Pennsylvania and West Virginia states by analyzing various species of fossils and ichnofossils for correlation and further information on their distribution. Evaluation of geomechanical properties of individual lithofacies is also recommended to understand the controls of depositional heterogeneities on the strength of these rocks.

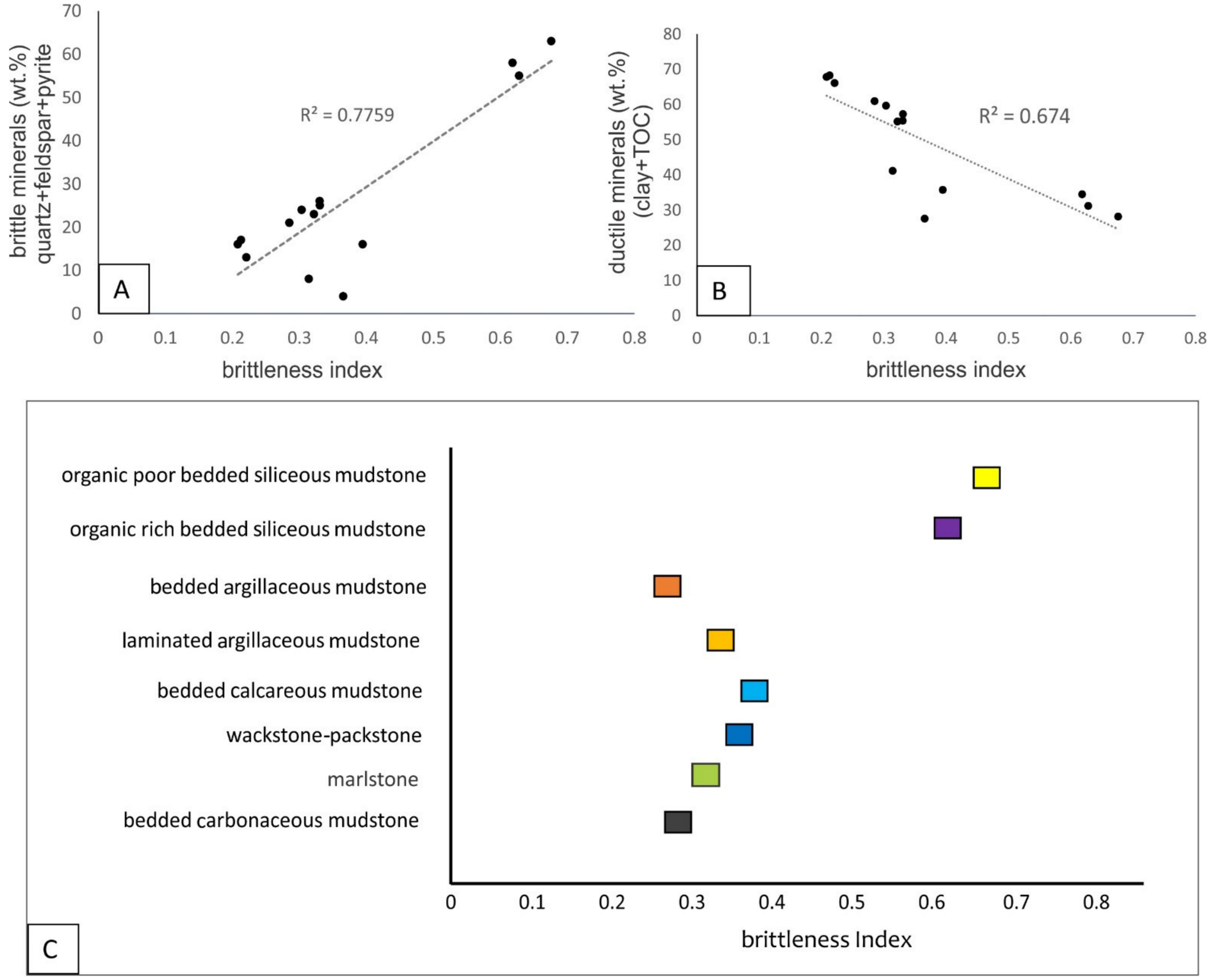


Figure 9. (**A**) Relationship between brittleness index and brittle minerals in the rock matrix. (**B**) Relationship between brittleness index and ductile minerals in the rock matrix. (**C**) Summary diagram showing the distribution of mudstone lithofacies into ductile, less brittle, and brittle zones on the basis of respective brittleness indexes.

6. Conclusions

The studied interval of the Marcellus subgroup, one of the most explored shale gas reservoirs in the Appalachian Basin, consists of several lithofacies. The lithofacies range in composition from argillaceous to siliceous, calcareous, and carbonaceous mudstone. Heterogeneities in the succession are identified in the form of varying mineralogical composition, slightly bioturbated to highly bioturbated chaotic matrix, organic-rich and organic-lean lamina planes, scattered fossil shells in the matrix, and fossils acting as lamina planes. The total organic content of lithofacies varies from top to bottom in both the formations and is recorded as high as 18 wt.%.

A proximal to distal transect indicates that bedded siliceous mudstone facies were deposited in the proximal facies belt of the depositional system showing a high silt content, pyrite, horizontal burrows, and fecal strings, whereas the distal facies belt sediments include bedded argillaceous mudstone facies characterized by a high clay mineral content, pyrite, and horizontal burrows. Sediments in the lithofacies of the medial facies belt were likely laid down under varying levels of energy and shows an equal proportion of quartz, calcite, and clay minerals. Fossils in this zone are aligned parallel to the bedding direction and act as lamina planes in the lithofacies of Union spring Formation. The grain size of the detrital mineral grains decreases from proximal towards the distal zone of the depositional system and the delivery of the sediments was likely dominated by unidirectional currents.

Abundant bioturbation recorded in all of the lithofacies argues against a complete anoxic environment during deposition. The oxygen level of the bottom waters was sufficient to facilitate the faunal burrowing in the lithofacies, suggesting that deposition took place in an environment that changed episodically and repetitively under conditions that were favorable for the activity of the burrowing fauna. Brittleness is found to be directly linked with the percentage of brittle and ductile minerals in the lithofacies and decreases from the proximal towards the distal zone of the depositional system with decrease in size of the sediments.

Author Contributions: Conceptualization, I.U.H. and E.P.; methodology, I.U.H.; software, I.U.H.; validation, E.P. and O.I.; formal analysis, I.U.H.; investigation, I.U.H.; resources, E.P.; data curation, I.U.H.; writing—original draft preparation, I.U.H.; writing—review and editing, O.I.; visualization, I.U.H. and O.I.; supervision, E.P.; project administration, E.P.; funding acquisition, E.P. All authors have read and agreed to the published version of the manuscript.

Funding: This work is supported by Petroleum Research Fund (PRF, 0153AB-A33) research grant awarded to Eswaran Padmanabhan, head of the Shale Gas Research Group (SGRG) at UTP.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The study did not report any data.

Acknowledgments: The authors are highly grateful to Jeffery Over (SUNY College, Geneseo, NY, USA) for providing the well-core data for this study. We would also extend our thanks to Nadeem Ahmad Sheikh for support in editing and formatting.

Conflicts of Interest: The authors declare no conflict of interest.

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