Fluid Evolution of Permian Carbonates in Muak Lek District, Saraburi province, Central Thailand: A Tectonically Driven Fluid System

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Abstract

The nature of deposition and diagenesis in carbonates of Central Thailand is not yet well documented and understood. This study focuses on an outcrop exposed in an active rock slab quarry in Central Thailand, extracting the Khao Kwang formation of the Saraburi Group. To quantify the depositional setting and diagenetic evolution of rocks in this area, especially related to levels of porosity and permeability modification, work was done via the integration of outcrop observations, petrography, X-ray powder diffraction (XRD) analysis, and carbon and oxygen stable isotope determinations.A unique giant bivalve (Alatoconchidae) and a characteristic fusulinid assemblage show the studied strata were deposited in the Middle Permian. Muddy lime sediments were deposited in a biotically-restricted, outer back-reef lagoon. The bottom sediments were occasionally reworked by storm events as evidenced by the frequent association of mud-dominated intraclastic limestones with un-layered fusulinid aggregates. Floatstone and rudstone are the dominant rock types defined at the field scale, while at the microscale the same rocks classify as grain-dominant wackestone and packstone. The $\delta^{13}C_{PDB}$ and $\delta^{18}O_{PDB}$ values plot into two groups in a C-O covariance plot; 1) A trend of positive carbon value from (0 to +6.1‰) versus more negative oxygen values (-4.7 to -16.7‰), and 2) A trend of increasingly negative carbon (-10.5 to -11.3‰) and oxygen values (-6.6 to -12.8‰). The more negative carbon and oxygen values show that precipitated calcites were bathed by subsurface waters driving refractionation and re-equili-bration under the influence of increasingly warmer burial fluids. By the moderate mesogenetic stage (with its pervasive pressure solution) carbonate sediments and bioclast pores were largely cemented. In addition, any remnant porosity was continually reduced though cementation along open fractures, which were filled with equant-shaped syntaxial vein calcites. Then, after more than 200 million years in the mesogenetic realm (since the Triassic), the distal effects of Himalayan Orogeny uplifted the Saraburi Group sediments. These limestones once again experienced active phreatic circulation as they entered the zone of telogenesis and karstification, with the associated development of enhanced levels of secondary porosity. However, a terra rossa soil was transported and deposited inside many of the vugs and solution channels, via infiltration of meteoric water; this resulted in a reduction of this newly formed karstic porosity.To summarise, this Permian limestone has a low chance of becoming a reservoir unless the overprinting effects of telogenetic groundwaters can be preserved. These overprints tend to erode the limestone, creating vugs and solution enhanced fissures and can also drive dedolomitisation.

Keywords: Permian carbonate, depositional setting, diagenetic evolution, carbon and oxygen stable isotope

1. Introduction

Permian carbonate rocks have been exposed many region in Thailand and its fractured equivalents are produced in the Nang Nuan and Phu Horm fields. One of the best regions of exposure of a variety of Permian carbonate in Thailand is in the Central Plain where tectonic thrusting has uplifted a variety of lithologies. However, these outcrops are not widely researched in terms of either depositional ordiagenetic fabrics. So, an outcrop exposed in an active rock slab quarry, extracting the Khao Kwang Formation in the Saraburi Group, was chosen for a detailed field and laboratory study.

Objectives of this study are 1) To describe the outcrop in terms of its geology, especially lithology and its relationship to the regional tectonic evolution. 2) To describe the rock's fabrics and petrophysical properties by interpreting samples down to the microscopic scale. 3) To document the outcrop in a framework of diagenesis, burial history, and also paleo-depositional environment by interpreting a combination of field observations and laboratory analysis.

2. Study area

The outcrop is located in the Lam Praya Klang area, near road 4005 (latitude 14°53’54.9”N, longitude 101°21’50.8”E), Muak Lek district of Saraburi province, in the Central plain of Thailand (Figure 1). The location is an active facing-stone quarry showing fresh outcrop surfaces in 2D and 3D, as seen and sampled in large facing stone blocks over an area some 100 meters long and 8 meters high (Figure 2).

3. Methodology

This research consists of the combination of detailed outcrop and laboratory analyses.

3.1 Field study

A total eight meters of outcrop in vertical succession parallel to bedding is described in a high detail: observations were made of lithology of rocks (texture, color, and grain size), biota and sedimentary structures. All rocks were classified in the field using Embry & Klovan’s classification (1972). Sixteen samples of representative rocks were collected and used in subsequent laboratory analysis. The outcrop was photographed and measures of bedding/fracture/vein orientation were documented.

3.2 Laboratory analyses

The main laboratory techniquese used comprised petrographic study (microscopic analysis), X-ray diffraction (XRD), and carbon and oxygen stable isotope determinations.

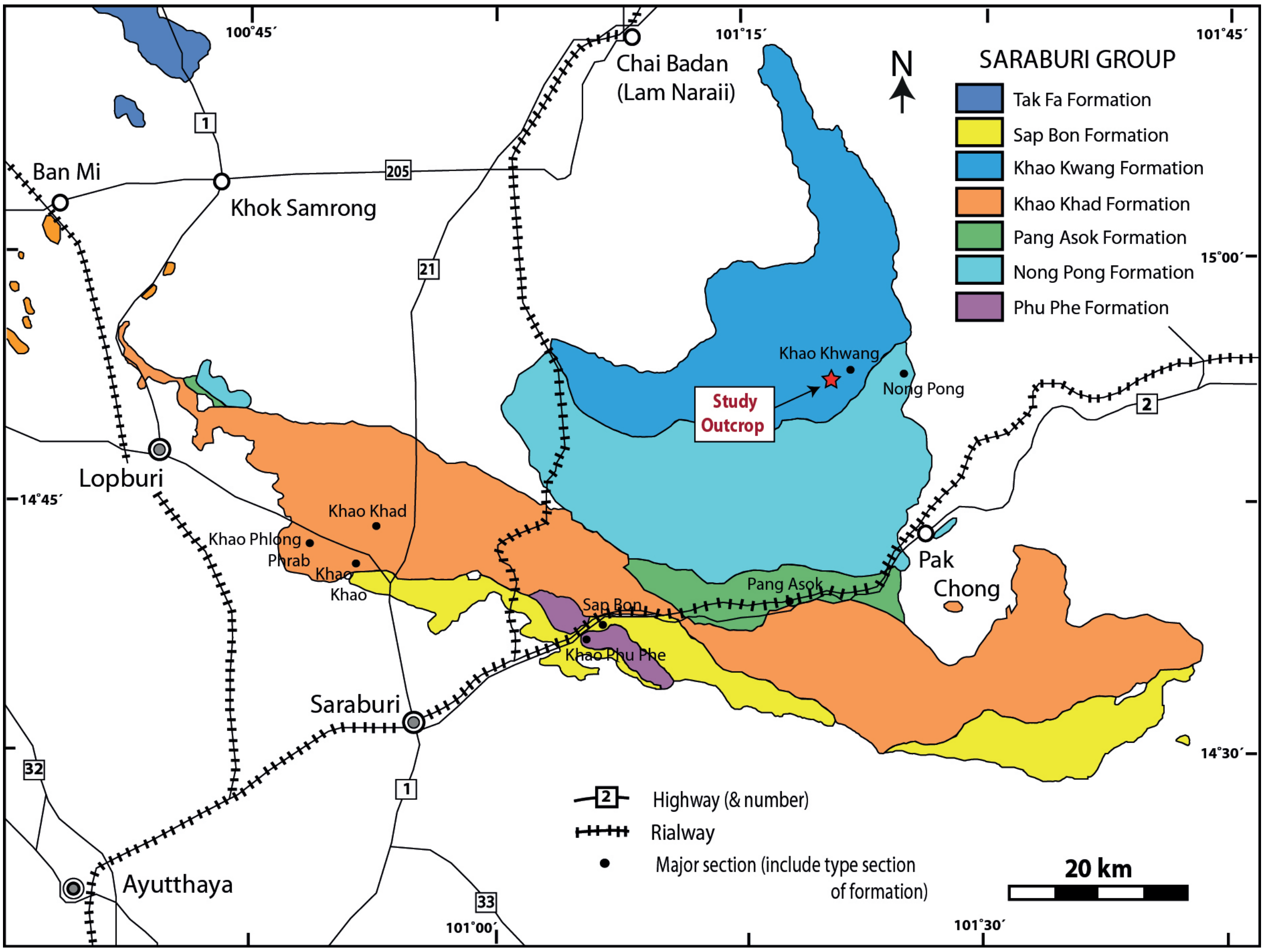

Figure 1 Geological map of Saraburi province with location of the studied outcrop (red star) and the positions of the type sections as defined by Hintong (1981) (modified from Warren et al., 2014).

1) *Petrology study*

Twelve thin sections were made from eleven rock samples collecting from the outcrop. They were described in terms of variations in textures which is important for interpreting a fluid driven cement character and timing. As well as identifying grains and cements, percentages of grain, cement, and matrix were made in order to classify the variety of rock types present.

2) *XRD*

Unknown materials found as mineral fills in vein, cements in coral, and sediment filling in vugs were drilled in powder form (by using dental technician's drill) to collect approximatly a gram for XRD analysis.

3) *Carbon and oxygen stable isotope*

Samples for the stable isotope analysis collected in a powder form, as for the XRD samples. 74 powders derived from drilling the faces of sixteen rock samples (most with corresponding thin sections) were taken across a selection of calcite veins, (likely) early marine cement from bioclast rims, rock matrix, and recent soil fills in some vugs and fractures. The purpose of the latter is to better understand fluid types present during uplift and karstification.

4. Results

4.1 Field study

1) *Lithology and stratigraphic log*

Dark grey limestone is the main lithology in the studied outcrop. It is composed mostly of floatstone with a packstone to wackestone matrix and lesser amounts of rudstone. Floatstone is a matrix supported rock, which contains more than 10% grains that are coarser then sand-size. Whereas, rudstone used to describe a grain-supported carbonate rock where the coarser than sand-sized clasts are in clast to clast contact, as in some layers with abundant with coarse-grained bioclasts deposited as possible storm layers. In addition, some larger grains or bioclastsconsist of the remains of *Alatoconchidae* sp. (giant bivalve) 10-20 cm long (Figure 2A), Fusulinids (many larger than 2 mm), Corals (whole and fragmented) 5-30 cm, and Algal plates 1-3 cm long (Figure 2B). Their pore spaces were partially filled by marine calcite cement (indicating eogenetic processes). The deposition of limestone in the lower outcrop section (lower 4 m of the 8m outcrop thickness) shows a high abundance of coarse bioclasts spread laterally over approximately 10-30 meters (rudstone) interbedded with fusulinid-floatstone layers (see logs in Figure 3). In the upper section (upper 4m of the slabbed outcrop thickness), floatstone dominates and grain to grain coarse bioclasts are less common. Karst features (vugs, soils and caverns) cross cut the outcrop, and formed as a consequence of uplift and karstification. Moreover, some intervals show limestone breccias in cavern intervals, resulting from dissolution and filling of the pore space by combinations of calcite cement (Figure 2D) and soil (terra rossa) (Figure 2F), which infiltrate from near the modern landsurface, due to the gravitational-driven descent of meteoric waters.

2) *Depositional environment prediction from field study*

In general, the giant bivalves prefer to live in a low-energy still-water lagoonal environment which can allow growth to "giant" sizes. However, the giant bivalves found in the study are were mostly found as small but related fragments, or with distorted shapes in a bedding-parallel plane. Neither association is indicative of storm events or tectonic forces. The rugose corals seem to be transported from nearby areas, because they are not preserved in growth position and edges of some are also disrupted (Figure 2C). Moreover, fusulinids are sometimes found isolated or as aggregates of un-layered fusulinids (Figure 2E). Therefore, the depositional area was likely a restricted marine area perhaps a lagoon influenced by occasional storm events.

3) *Bedding orientation and fracture-stylolite development*

The bedding trend is NNW-SSE strike with an average 20° dip, as measured from a major

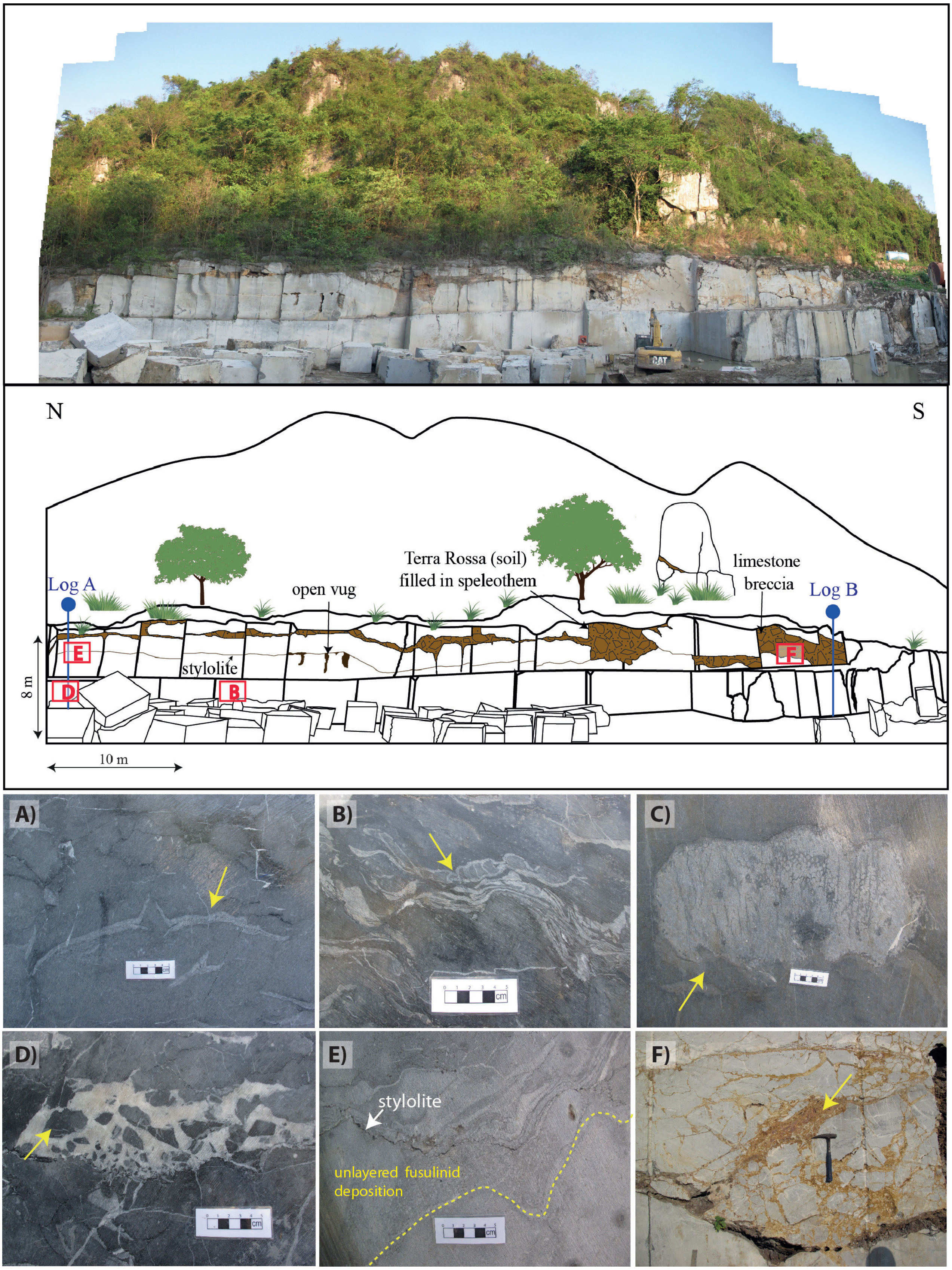

Figure 2 outcrop panorama and outcrop sketch showing relative position of logged sections A and B (Figure 3) with outcrop photograph positions. A) giant bivalve, B) algae, C) coral, D) limestone breccia with calcite cement filled, E) unlayered fusulinid deposition, and F) terra rossa in brecciated limestone.

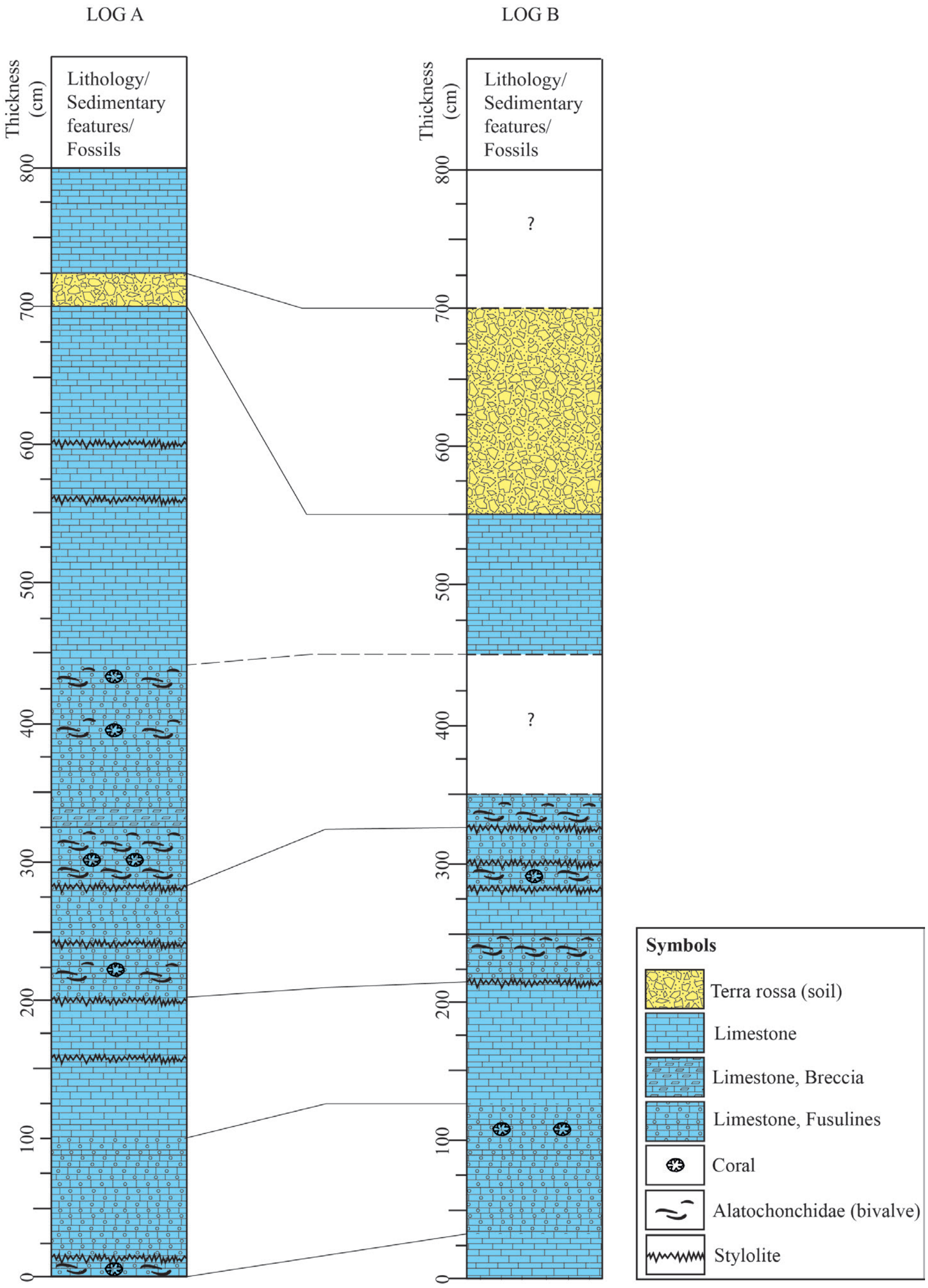

Figure 3 log A and B correlation. There is no obvious lateral facies change between Logs A and B situated some 60 meters apart (see Figure 2 for log's location).

sub-horizontal stylolite which formed parallel to a biota-enriched layer, containing Alatoconchidae.This layer indicates a paleo-seafloor and the bedding of carbonate rock in the quarry. There are least two generations of fractures exposed in the quarry. All fractures are filled by calcite and are typically offset by stylolites.

4) Karst features

Karst features observed in the outcrop faces are mostly vugs and karst collapse breccias of various sizes, ranging from 1 cm to meters wide. They were developed as rock uplift and telogenesis enhanced the second porosity levels and pore connectivity. Brownish-orange sediments fill in vugs are terra rossa soils. They were carried into the expanding pore space by meteoric water inflows. Terra rossa is likely to develop during dissolution of the associated carbonate rock so it is typically found within karst collapse breccias, which are angular-shaped rock pieces that collapsed into karstic accommodation space from the surrounding country rock (Figure 2F). Vugs without the soil fills are called open vugs. It can indicate that meteoric water has eroded all soil that used to be deposited inside the vug, or it may be a vug that never contained any fill.

4.2 Petrography study (Microscopic analysis)

Eleven rock samples were collected from the studied outcrop in order to make stained thin sections. Half of each thin section was stained by a combination of the organic dye Alizarin red S (ARS) and with potassium ferricyanide (Hitzman, 1999). Mineralogically, all studied thin sections are composed of calcite (pink to red stain) and no ferroan carbonates were present. At the scale of a standard thin section the rocks are wackestones and packstones (Figure 4).

1) Matrix

The thin sections of floatstone and rudstone (at the outcrop scale) show a fine-grained carbonate matrix dominated by lime mud (Figure 6A).

2) Grain (Bioclast)

Bioclasts are either fragments or complete skeletons of organisms such as fusulinids, foraminifera shells, crinoid fragments, and bryozoans (Figure 6C, 6D, 6E). Fusulinids identified were *Pseudodoliolina* sp., *Verbeekina* sp., *Neoschwagerina* sp., and *Yangchienia* sp. (Figure 5). They indicate the age of carbonate deposition is Middle Permian. Moreover, micrite envelope were found surrounding some grains (Figure 6F), forming a dark-coloured, fine-grained, and carbonate exterior to the skeletal fragments. A micrite envelope forms by endolithic microbial infestation on the surface of grain fragments that then fill with Mg-calcite or aragonite mud after the microbes die. Repeated generations of boring and microcrystalline cement creates the micrite envelope and if the process goes to completion the grains are completely micritized and evolve into peloids. Sediments with pervasive development of micrite envelopes are and evolve into peloids. Sediments with pervasive development of micrite envelopes are usually interpreted as accumulating in oxygenated seafloor conditions (Ahr, 2008).

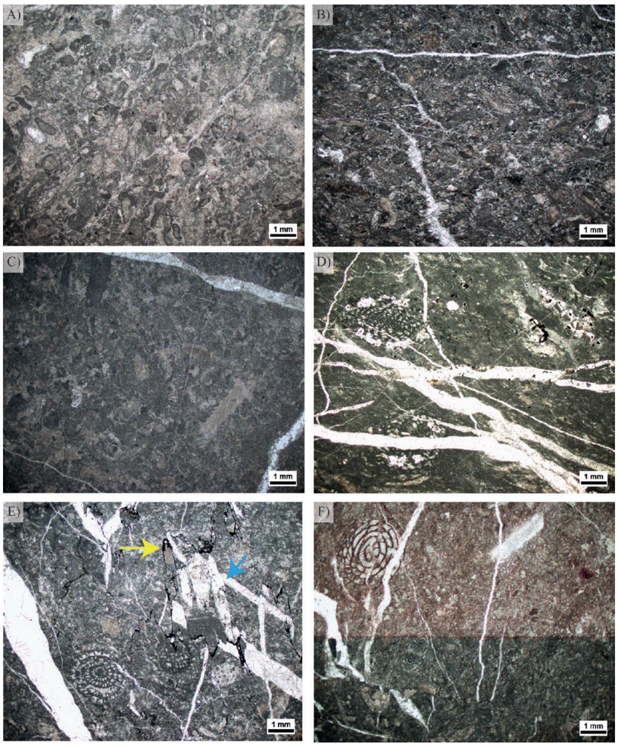

Figure 4 Photomicrographs of: A) and B) packstone, containing more than 10% bioclasts, grain supported, C) and D) wackestone dominated by lime mud, matrix support, E) stylolite (yellow arrow) formed after the calcite-filled fractures (blue arrow) were created, and F) one side of a thin section dyed red by ARS, indicating calcite.

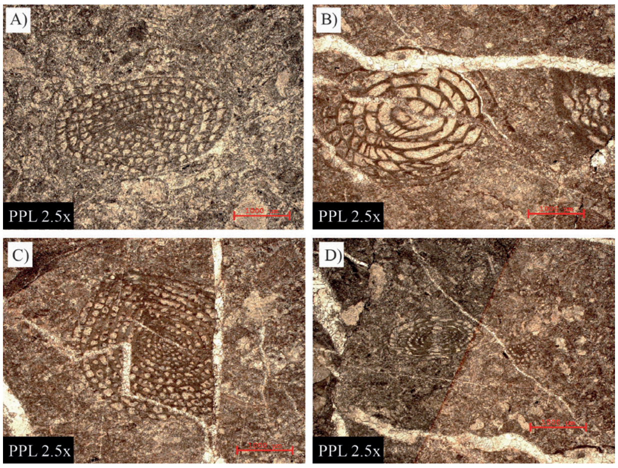

Figure 5 Photomicrographs of various index fusulinids in thin section A) *Pseudodoliolina* sp., B) *Verbeekina* sp., C) *Neoschwagerina* sp., D) *Yangchienia* sp.

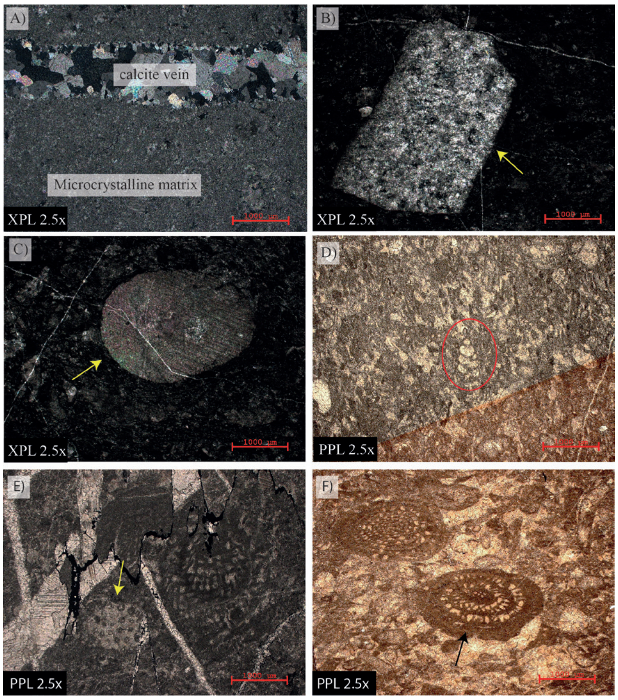

Figure 6 Photomicrographs of A) microcrystalline matrix cut by a calcite vein, B) limestone breccia fragment, C) recrystallized crinoid columnal, D) smaller uniserate foraminifera, E) bryozoan adjacent to stylolite, and F) micrite envelope developed on a fusulinid test.

3) Calcite fill in fractures

Calcite filled fractures with various orientations and responses to stylolitisation typify the quarry faces. Some fracture fills were first precipitated karst collapse as marine calcite cement linings that then evolve into larger crystals. Small aligned calcite crystals first formed at both edges of fracture. Then, syntaxial growth formed later crystals found in the middle of the fracture fill made up of larger equant-shaped crystals that completely fill in the fracture pore space (Figure 6A). In some larger fractures there are recrystallized limestone clast breccias surrounded by a sparry calcite fill (Figure 2D).

4) Microfractures and stylolite

When the carbonate sediment was deeper inthe burial environment (mesogenesis), overburden-driven compaction of sediment created microfractures cutting cemented bioclasts and stylolites that are mostly parallel to bedding. Likewise, non-parallel bedding stylolitisation took place in later mesogenesis during tectonic compression or folding. Bed-parallel and later stylolites form as carbonate grains and matrix are dissolved by pressure solution and display serrated contacts made up of insoluble residues. At the microscale, stylolite sutures display as dark colored fills in both plane polarized light (PPL) and cross polarized light (XPL). In the samples studied it is thought that the stylolite material is mostly iron-oxide, aluminosilicate clay, or organic matter residues.

4.3 XRD

Three samples were chosen for mineral confirmation; E1: mineral fill in a bioclast (coral), E2: mineral precipitated in a fracture (sparry crystals), and E3: sediment fill in vug.The mineral fills in the bioclast and in the fracture are pure calcite ($CaCo_3$), the exception is in the terra rossa, which is composed of low Mg-calcite as a dominant mineral but muscovite KAl_2 Si_3AlO_{10} $(OH)_2$) is also present a trace component in the soil fill.

4.4 Stable isotopes

Stable isotope values of $\delta^{13}C_{PDB}$ and $\delta^{18}O_{PDB}$ were determined using a total of 74 samples from the study outcrop. Samples include: 6 samples from sediment fill in speleothems, 40 samples from various calcite veins, 14 samples from matrix, 11 samples from bioclasts, 2 samples from stylolites, and 1 sample from a limestone breccia

Carbon isotope values ($\delta^{13}C_{PDB}$) show a range from -12.8 to +6.1‰ and oxygen isotopes ($\delta^{18}O_{PDB}$) from -4.7 to -16.7‰ (Figure 7). There are two separate grouped trends seen in the resulting C-O covariant plot: 1) A trend of positive carbon value (above 0 ‰ $\delta^{13}C$) versus more negative oxygen values, and 2) A trend of increasingly negative carbon and oxygen values. The increasing trend of negative carbon and oxygen values indicate moderate burial passing into deeper burial or deformation fluid (elevating temperature trend). For the majority of carbonates, deposition and precipitation began in marine (eogenetic) setting and passed via burial into the deep burial realm by ongoing deposition

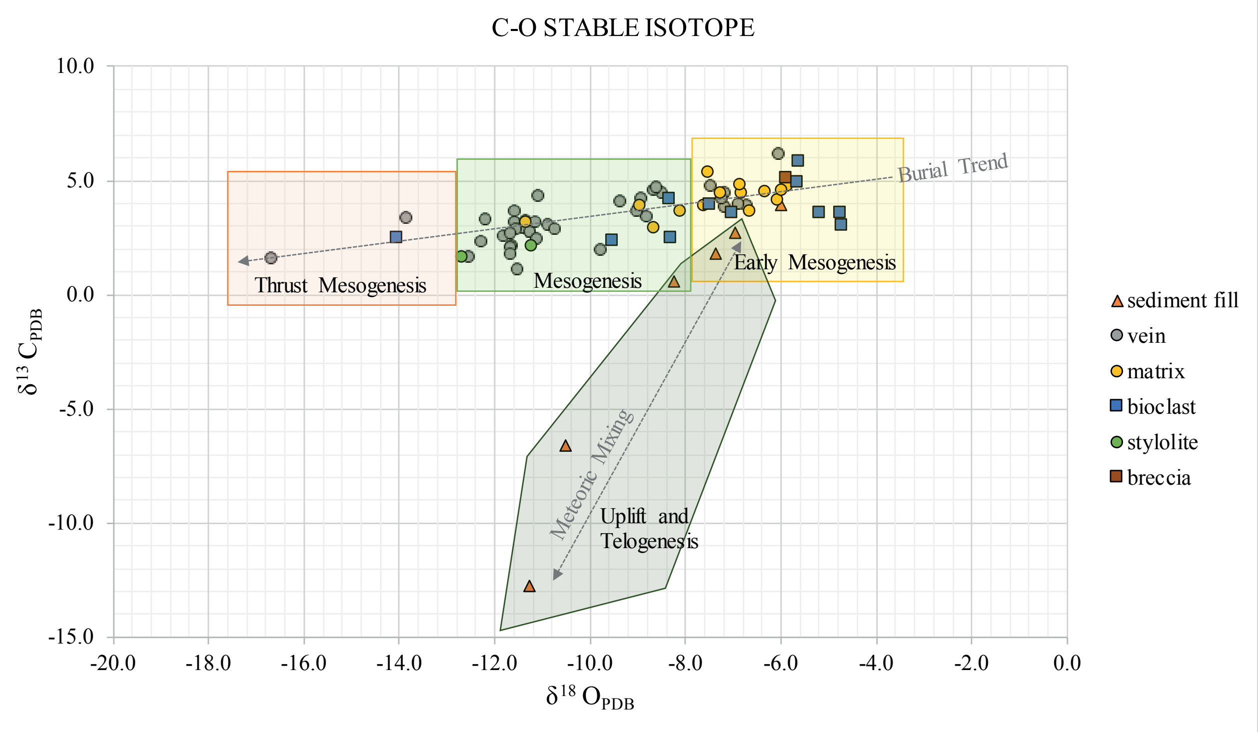

Figure 7 plot of $\delta^{13}C_{PDB}$ and $\delta^{18}O_{PDB}$ of the samples from study area, superimposed on the shaded plot areas defined in Warren et al., (2014).

of overburden, and associated tem-perature induced refractionation and re-equilibration of the calcite. The documented burial trend overlays the curves of Warren et al., (2014), and indicates that marine carbonate sediments were initially cemented in the eogenetic realm by marine calcite (including microcrystalline cement and cement rinds) and the same calcite cement also precipitated in bioclast pores (confirmed by XRD). With burial during early mesogenesis to middle mesogenesis the calcite in the sediments then experienced ongoing re-equilibration while flushed by increasingly warmer burial waters. Outside this early to moderate burial field there is one exception, a bioclast sample that plots in in the thrust mesogenesis field. When field checked, the sample was collected from a 20 cm elevation in the log and it was then seen in the slab/field photograph that a late stage calcite vein cut through part of the bioclast, implying that it was probably recrystallized by a later calcite cement during regional compression.

Samples of the calcite veins show an increasingly negative trend in both oxygen and carbon values, so following the burial trend first documented in Warren et al., (2014). Some of the later calcite veins developed in the thrust mesogenesis period (indicated by the orange shading in Figure 7. Sediment fill samples collected from vugs show a separate group of values that occur in the plot field of uplift (telogenesis) related to telogenetic inflows of meteoric water and speleothem fill in the evolving pore space.

5. Interpretation

5.1 Paleo-depositional environment

Paleo-depositional environment of the study area is interpreted as a somewhat restricted outer back-reef lagoon (Figure 8). This setting, with its mostly lower-energy levels, allowed fine-grained carbonate sediment to deposit in a restricted area evidenced by microcrystalline calcite matrix and micrite envelopes surrounding bioclasts. Dark-grey limestone classified as mud-dominant floatstone with lesser rudstones are the main lithologies. Bioclasts are abundant in some layers as a result of winnowing from storm events, as evidenced by muddy matrix, un-layered fusulinid aggregates and solitary rugose corals not showing in-situ growth positions. Restriction is indicated by a relatively low-diversity assemblage of organisms compared to a normal marine unrestricted open platform biotal assemblage. Moreover, Alatochonchidae sp. mostly prefer to live in low energy environments.

5.2 Diagenetic history

Integration of petrography and stable isotope analysis is used to identify the evolution of fluids preserved in the calcite cements in the studied Permian carbonate. Since these carbonate sediments were deposited in a lagoonal environment, pervasive framestone cements were not present. In the first stage of carbonate diagenesis (eogenesis), the sediments were not completely cemented until they were buried and overprinted by cements from chemical compaction in the mesogenetic stage. At such burial levels, marine and then rock-derived bicarbonate characterized fluids flowing through the pore space between carbonate grains, solutions of Ca-Mg ions were crystallized and then experienced thermal re-equilibration as pore fluid temperatures and pressures increased. The increasingly negative $\delta^{13}C$ and $\delta^{18}O$ trend seen in the stable isotope values and associated textures are evidence of this. The first development of fractures are tied to stylolites that are parallel to sub-parallel bedding and are the result of increasing overburden thickness. Later, another set of fractures and stylolites were developed at steeply inclined angles to bedding that were driven by tectonic compression during thrusting of Saraburi group in the Triassic period (Indosinian orogeny ~250 million years ago). Two hundred million years later, the distal effects of Himalayan Orogeny uplifted the Saraburi group sediments so that they entered the zone of telogenesis and karstification. This created vugs, caverns and speleothems in rising limestone masses as it formed then enhanced levels of secondary porosity. Terra rossa (red soils) that covered the top of the limestone exposure in the study area infiltrated into some of the vugs, carried by meteoric water. As dissolution continued, vertical to sub-vertical shafts filled or partially filled with limestone breccia and were surrounded by crackle breccia that was produce during karst collapse. This allowed new broader pathways for carbonate soils and so reduced the porosity and permeability in the opening vugs.

6. Discussion

6.1 Hydrocarbon potential

Khao Kwang Permian limestones as documented in this study are another key to hydrocarbon reservoir understanding as they show how second porosity and pore connectivity can be enhanced during karstification in the later stage of diagenesis (telogenesis). Although terra rossa soils and other spleothems can fill in the vugs, they remain porous (between soil grains and open vugs) which if buried once more would allow hydrocarbon accumulation even though the unaltered surrounding rock matrix lacks any porosity and permeability. Unless telogenetic groundwaters erode the limestone to create vugs

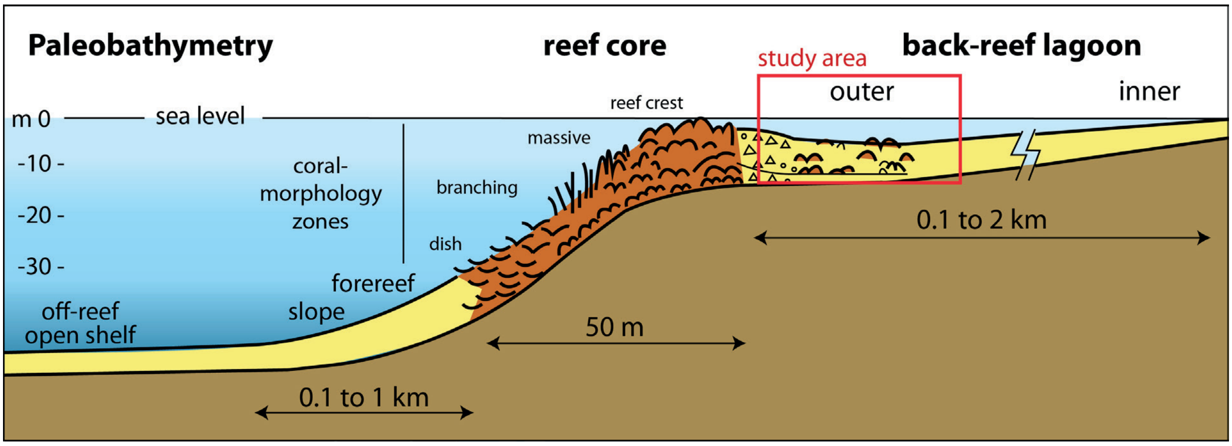

Figure 8 depositional setting of carbonates in the study area (modified from Pomar, 2004).

or drive de-dolomitisation, the rock will lack effective porosity and permeability. But, based on the isotope plot fields identified in this study, one could sample cuttings in a future well intersecting subsurface Permian carbonates and identify if any calcites in the cuttings showing evidence of meteoric signatures (lie in the meteoric mixing trend) that have overprinted an otherwise deeply buried rock (values along the burial trend).

7. Conclusions

1. Lithologies are mostly muddy limestone that classify the field as mud-dominated floatstone and rudstone. Samples of the same rock under the petrographic microscope classify as wackestone and packstone.

2. These carbonate sediments were deposited in a restricted-marine lagoonal environment. Bioclasts show various sizes from below a millimeter to many centimeters. Fusulinids are dominant and indicate a Middle Permian age.

3. The carbonate sediment has experienced chemical compaction (stylolitisation) during burial (mesogenesis) and interparticle porosity no longer exists. Matrix first formed as grains composed of microcrystalline calcite with interparticle porosity. Micrite envelopes developed on bioclasts as can be seen at the microscale. The rock is now heavily recrystallized, and isotopes indicate ongoing burial re-equilibration of the calcite.

4. Many fractures cut though the matrix and early calcite cement and then were filled with equant-shaped syntaxial vein calcites. Some of these earlier calcite fractures are displaced by bedding-parallel stylolites that formed in response to increasing overburden. Later stylolites formed at angles that were steeply inclined to bedding and the bedding-parallel stylolites. These later pressure solution features were driven by tectonic compression perhaps tied to the Indosinian Orogeny. Higher temperature fluids were driven along these later fractures as calcite vein precipitation continued, and are indicated by a depletion of $\delta^{13}C$ and $\delta^{18}O$ values in the stable isotope plot.

5. Subsequent uplift and karstification of the deeply buried carbonates created secondary porosity and enhanced the permeability as a result fracture widening, as well as vug and cavern creation (telogenesis).Where meteoric water flowed from the surface into the growing vugs and caverns, a terra rossa soil was transported and deposited inside the vugs, so reducing porosity (Figure 9).

Time	Ma	Stage	Process	Porosity		Permeability	
				decrease	increase	decrease	increase
Middle Permian	~270	Eogenesis	1. Deposition of carbonate sediment in lagoon				
			2. Early marine cement (syndeposition)	←		←	
↓		Mesogenesis	3. Grain compaction and marine calcite cement precipitates between grains	←		←	
			4. Development of sub-horizontal fractures and by vertical compression of overburden		→		→
			5. High temperature fluid drive cement precipitation in fractures, formed calcite crystals in equant shapes	←		←	
			6. Development of sub-vertical fractures by compressional stress		→		→
Triassic	~250		7. High temperature fluid continuely precipitate calcite along new fractures	←		←	
			8. Horizontal stylolites created by pressure solution in deep burial	←		⇠	
↓			9. Thrusting and compression create vertical stylolites	←		⇠	
		Telogenesis	10. Dissolution of limestone along fractures		→		→
Quaternary	~0.01		11. Karst collapse		⇢		⇢
			12. Terra rossa forming in vugs by meteoric water driven	←		←	

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Figure 9 Summary of carbonate diagenetic evolution in the study area.

6. Today, this Permian limestone has low chance of becoming a reservoir unless it is rapidly buried below a muddy seal, so allowing it to retain its telogenetic porosity.

7. Isotope values measured in this study define, 1) a regional burial trend and 2) a meteoric mixing trend, which likely have application in future subsurface studies.

8. Acknowledgement

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