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論文内容の要旨

Sri Lanka records on of the longest and complete tectonic evolution from the mid-latitude in the southern hemisphere (during the Jurassic) to the equatorial northern hemisphere. Therefore, onshore and offshore sedimentary basins in Sri Lanka provide the natural laboratory to reconstruct paleoenvironmental and paleoclimatic during its northward voyage from Gondwana to Asia. The Jurassic Gondwana sediments were collected from the onshore Andigama and the Tabbowa Basins. The drillcore cutting samples from the Late Cretaceous to Miocene were obtained from the two exploration wells (the Barracuda and Dorado North) in the offshore Mannar Basin. The Late Quaternary sediment samples were collected from the coastal Bolgoda Lake in the southwest of Sri Lanka. CHNS elemental analysis (n = 1279) and gas chromatography and mass spectrometer (GC-MS) analysis (n = 177) were performed for sediment samples. The standard burial history, thermal maturity, and kinetic models were prepared for the Mannar Basin using etroleum system modeling software (BasinMod 1-D). The 14C radiometric dating was carried out using accelerated mass spectrometry for the Late Quaternary Bolgoda Lake samples.

The Jurassic Andigama and Tabbowa Basins: Total organic carbon (TOC) contents are high (3.05-5.10%) in the Jurassic Andigama Basin, while the sandy sediments in the Jurassic Tabbowa Basin have very low TOC (0.04-0.17%). The Andigama mudstones are thermally immature, and the Tabbowa sediments are moderately matured in the oil generation stage based on sterane and hopane isomers proxies. The Jurassic Andigama Basin indicates a large proportion of middle-chain *n*-alkanes (*n*C21-C25), enriched C29 steranes, high C/N ratios (16.3-37.8), very low total sulfur (TS <0.001%) and higher pristane/phytane ratios (Pr/Ph = 2.1-3.0). These results suggest that terrestrial organic matter (OM) were deposited in the

freshwater swamp under oxic condition. Abundant retene, simonellite, perylene, 1-methylphenanthrenes, 1, 7- dimethylphenanthrene and 1, 2, 7- + 1, 2, 9-trimethylphenanthrenes in the Andigama mudstones indicate that the OMs were mainly originated from gymnosperm with fungi. In contrast, sandy sediments in the Tabbowa Basin have predominant *n*C16-C21 alkanes with a minor peak of waxy *n*-alkanes (*n*-C29, *n*-C31, and *n*-C33), very low TS (<0.001 %), abundant C27 steranes and higher Ts/(Ts + Tm) [Ts: 17 α (H)-22, 29, 30-trisnorhopane, Tm: 18 α (H)-22, 29, 30-trisnorhopane] triterpane ratios, which suggesting the OMs from algal origin with a significant terrestrial higher plants.

The pericratonic Mannar Basin (lithology): The Late Cretaceous to Late Paleocene lithology of the Barracuda well recorded mud dominant sediments and interbedded sandstone with volcanogenic materials, and the Dorado North well recorded sand dominant sediments and interbedded mudstone. At the end of the Late Paleocene, sedimentary facies were drastically changed from calcareous mudstone to argillaceous marl/ marlstone in the both wells. These facies variations have an apparent relation with the sedimentation rates in the basin. This shift is interpreted as the continuous subsidence of the basin and changes of an arid climate into warm and humid tropical conditions. The lowest sedimentation rate was recorded during the Eocene suggesting that the timing of collision between Indian and Asian plates. Burial history by 1D modeling indicates rapid subsidence from the Late Cretaceous to the Paleocene during the rift transition stage. Subsidence rate was decreased during the Eocene.

(Carbonate burial): The Late Cretaceous CaCO3 rich (average of Dorado North = 24.7 ± 9.4 and Barracuda = 32.5 ± 9.4) sediments could indicate high-productive stage under warm climate. The Deccan-Reunion basalt could be acted as a significant contributor to the mass extinction of coccolithophores/ foraminifera followed by reduction of CaCO3 and organic carbon burial of the basin. The deposition of CaCO3 rich sediments (average of Dorado North = 29.3 ± 11.4 and Barracuda = 40.2 ± 12.1) could indicate movement of Indian plate into northward warmer tropical latitudes since the Late Paleocene. It is correlated with the Cenozoic global cooling towards the present glaciated Earth.

(Organic carbon burial/ source rock beds): TOC contents are relatively low (< 1%) in the lower most Early Campanian sediments of the Dorado North (average = 0.45 ± 0.22 %) and Barracuda (0.97 ± 0.23 %) wells. However, the Early Campanian to Late Maastrichtian sediments (3320-3060m) of the Dorado North (1.64 ± 0.57%), the Late Campanian to Late Maastrichtian sediments (4540-4270 m) of the Barracuda (1.34 ± 0.36%) and Middle Oligocene to Early Miocene sediments (2520-2139 m) of the Barracuda (2.51 ± 1.2 %) can be recognized as OM rich source rock beds in this basin. This Middle Oligocene to Early Miocene OM rich bed contains black carbon and laminations suggesting that seasonal events in this area possibly due to development of monsoon activity.

(OM type): The higher C/N ratios of the OM rich the Late Cretaceous sediments of the Dorado North (27.13 ± 12.4) and the Barracuda (20.36 ± 7.6 and 23.5 ± 15.0) wells can probably indicate accumulation of terrestrial OMs from higher plants. The *n*-alkane compositions of these samples indicate abundant middle-chain (*n*C21-*n*C25) and long-chain (*n*C27-*n*C31) wax

from vascular plants with a significant amount of short-chain (*n*C18-*n*C19) wax from plankton. Therefore, the OM rich bed can probably contain gas prone Type II-III kerogen. The tectonic activities (separation of Laxmi
Ridge-Seychelles and Seychelles) from the Indian plate could be accompanied enhancement of TOC and terrestrial OMs during the Late Cretaceous.

(Thermal maturity and kinetic models): These OM rich beds are thermally mature based on maturity parameters of C29 sterane 20S/(20S + 20R) ratio (0.27-0.56 in the Barracuda and 0.20-0.33 in the Dorado North) and C31 hopane 22S/(22S + 22R) ratio (0.49-0.56 in the Barracuda and 0.41-0.53 in the Dorado North). These results suggest that sediments in the Barracuda are more thermally matured than the Dorado North. In addition, OM rich Oligocene-Miocene sediments could not deeply subsidence to natural gas stage based on relatively lower maturity (C29 sterane 20S/(20S +20R) ratio = 0.28-0.37 and C31 hopane 22S/(22S + 22 R) ratio = 0.45-0.48). Biomarker proxies are consistence with the maturity models. In detail, the standard rifting heat flows can be extrapolated with observed thermal maturity and gas deposit in the Mannar Basin. The kinetic model of the representative Cretaceous sediments (4260-4470 m) in the Barracuda well can indicate in-situ gas generation (mainly) since the Early Eocene. The in-situ gas generation was gradually increased and reached peak conditions during the Miocene (ca. 20 Ma). However, the Cretaceous sediments of the Dorado North and the Tertiary sediments of the both wells indicate poor cumulative hydrocarbon generation.

(Depositional environments): The OM rich sediments from Campanian–Maastrichtian and Oligocene–Miocene could be formed by the abundant input of terrestrial OM with nutrients for enhanced primary production in the marine area. The relatively higher C/S values in these beds could indicate weaker microbial activities (sulfate reduction) in terrestrial OMs rich sediments. In contrast, C/S values of the Early-Late Paleocene sediments (average of Dorado North = 32.07 ± 20.21 and Barracuda = 16.21 ± 20.14) suggest oxic depositional environment due to regression of relative sea-level.

The coastal Bolgoda Lake: The history of the Bolgoda Lake can be divided into two major chronostratigraphic divisions that are quasi-steady state (from ~7.5 ky B.P. to ~2.5 ky B.P.) and non-steady state (from ~2.5 ky B.P. to the Recent). The moderate productive lower sediment sequence was mainly deposited in marine-terrestrial influence, oxygen-poor and anoxic conditions during mid-Holocene highstands. The major environmental change was characterized by enhancement of TOC (%) and accumulation of reworking terrestrial OM in the semi-closed aquatic system after the sea-level regression (~2.5 ky B.P.). The mid-Holocene regression has changed geomorphology of the study area from part of the bay of larger paleoriver system to semi-closed and separate fluvial dominant estuary of local streams. The *n*-alkanes ratios of *n*-C29/*n*-Call and *n*-C37/*n*-Call suggested a gradual climatic transition from wetter to dryer since middle Holocene. Accumulations of petroleum residues and pyrogenic polycyclic aromatic hydrocarbons (PAHs) in modern sediments identified anthropogenic activity after the European settlement (15th century).

論文審査結果の要旨

スリランカはインドと共に過去約 2 億年間に南半球中緯度地域から北半球低緯度地域まで大規模な移動を経験しており，Amila Sandaruwan Ratnayake 氏は，その間の気候帯の変化と堆積物有機物の濃集システムの変化，有機物組成の応答および石油・天然ガス根源岩の形成などの関係を明らかにした。CNS 元素分析で 1279 個の膨大なデータを得，GC-MS 分析 177 個の結果を加え，ビトリナイト反射率，一次元堆積盆地解析ソフト BasinMod 1-D による解析を併せて網羅的に行った研究成果は，質・量ともに博士研究に相応しいと判断される。

本論文は 3 つのケーススタディから構成されている。同氏は，ジュラ紀以降の広範な時代の堆積盆地の有機物濃度・炭化水素・バイオマーカーを用いて主に陸源有機物の影響の観点からその特徴を明らかにした。 ①[第一のケーススタディ] ジュラ紀の Andigama 盆地と Tabbowa 盆地を対象とした。両盆地は当時南半球中緯度地域のゴンドワナ大陸中部のテチス海の湾奥に位置していた。前者では TOC 濃度が高く(3.05-5.10%)，後者では対照的に低かった(0.04-0.17%)。前者は主に淡水の酸化的堆積環境ではあり(TS <0.001%，Pr/Ph = 2.1-3.0)，*n*-alkanes (nC_{21}-C_{25})，retene, simonellite, perylene, 1, 7- dimethylphenanthrene の供給が多く，陸源裸子植物および抽水植物の供給が多い湿潤な堆積環境であったことが明らかになった。 ②[第二のケーススタディ] 白亜紀～中新世の Mannar 海底盆地の 2 本のボーリングで採取されたカッティング試料を用いた。ここでは，乾燥気候帯に属した後期白亜紀には堆積速度が小さく(約 24 m/Ma)，低い TOC と低い炭酸塩炭素濃度(約 1.1%および 約 2.5%)の石灰質泥岩で特徴づけられ，古第三紀暁新世になると熱帯性の湿潤気候となり，堆積速度がやや大きく(約 22～29 m/Ma)，高い TOC と高い炭酸塩炭素濃度(約 1.5%および 約 4%)の泥質石灰岩に変化していった。また天然ガスを産出させた根源岩は後期白亜紀のカンパニアンからマーストリヒチアンの層準であることを明らかにした。 ③[第三のケーススタディ] 現世堆積盆地の Bolgoda 汽水湖から採取した 3 本のコア試料，28 地点の表層試料，および接続河川の 4 地点のマングローブ泥を用いた。この熱帯湿潤地域で 2500 年前までは TOC 濃度が 1～5%（C/N 比 15～30）で堆積速度は約 0.12mm/y であったのに対し，2500 年以降には TOC 濃度が 2～30%（C/N 比 15～50）で堆積速度は約 0.23mm/y となり，陸源有機物の供給増加が堆積物有機物濃度を極めて高い濃度まで増加させたことを明らかにした。

これらの 3 つのケーススタディーから，以下のことが共通して見出されたことに高い学術的意義が認められる。(1) スリランカのジュラ紀から現在までの主要堆積盆地において有機物濃度を増加させた要因は陸源有機物の供給に由来する。(2) スリランカ沖の石灰質プランクトン生産性の増加は陸源有機物・細砕物の供給に伴って行われた。(3) 陸源有機物の供給増加は泥質堆積物の堆積速度増加を伴った。

Amila Sandaruwan Ratnayake 氏が用いた地質試料は，自らの調査によって採取されたもののほか，海洋ボーリングコア試料については Petroleum Resources Development Secretariat (PRDS), Sri Lanka の許可により提供を受けたものである。分析は，Amila Sandaruwan Ratnayake 氏が自ら全て技術を習得して質の高いデータを得た。一部，^{14}C 年代値・$\delta^{13}C$ 値については外部委託により行ったがデータの解釈は自ら行った。Amila Sandaruwan Ratnayake 氏は有機地球化学的手法を広く修得し幅広い知識・技術を身に付けたため，今後，研究者として活躍する基礎が確立したと判断される。博士論文・発表からは，知識・技術・研究遂行能力が十分であることを読みとることができ，博士の学位を授与されるのに相応しいと判断される。