Feature Article

Paleoenvironmental Reconstructions using Organic Source Indicators: A Summary of Examples from Sri Lanka

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Abstract

The qualitative and quantitative analysis of sedimentary organic matter (i.e., the residue of past biota) provides integrated histories of marine and continental past life and paleoenvironmental /paleoclimatic changes. Organic geochemical investigations are possible by combining (i) bulk properties such as elemental compositions, stable isotope ratios, and Rock-Eval pyrolysis data, and (ii) biomarker molecular compositions such as *n*-alkanes, sterol, and polycyclic aromatic hydrocarbons compositions. The analytical approaches described in this overview illustrate the published examples of lacustrine and marine organic geochemical studies in Sri Lanka. In summary, the Jurassic Andigama and Tabbowa Basins provide different sources of organic matter, followed by availability of nutrient for algal growth and the amount of land runoff to the basins. Rock-Eval analysis of the Cretaceous to Paleogene sedimentary rocks in the offshore Mannar Basin reveal the presence of gas-prone land-plant organic matter mainly and minor oil-prone algal organic matter. The amounts and types of organic matter variations in Bolgoda Lake sediments indicate changes in Holocene sea-level, coastal geomorphology, and continental climates during the last 7,000 years. In future directions, applications of novel organic geochemical proxies and understanding of original biologically synthesized materials in tropics would improve interpretations of paleoenvironmental changes. Besides, local and regional paleoclimatic proxy and model studies would refine future paleoenvironmental reconstructions in Sri Lanka.

Keywords: organic carbon, C/N ratios, biomarkers, n-alkanes, Sri Lanka

1. Introduction

Photosynthesis is the fundamental process for mass production of organic matter (OM) since the Precambrian. Terrestrial higher plants have contributed to primary production since the Devonian. Phytoplankton, zooplankton, and higher plants can be presently recognized as the main contributors to OM in sediments. The OM content can vary in sediments from less than 0.1% to 4%, and depends on aquatic primary productivity (autochthonous), secondary accumulation of terrestrial OM (allochthonous), and preservation against microbial decomposition in an oxygen-deficient environment (Berner, 1982; Meyers, 1997; Sampei et al., 1997a). The primary productivity in the aquatic environment is principally controlled by light, temperature, and nutrient such as phosphate and nitrate. Further, OM-rich sediments are present in lagoons, estuaries, and continental slopes followed by high primary productivity and/or deposition of land-derived terrestrial plant materials (Meyers and Ishiwatari, 1993; Sampei et al., 1997b). The fine-grained sediments enhance the preservation of coated OM in sediments by limiting the access of dissolved oxygen (Berner, 1984; Berner and Raiswell, 1984; Sampei et al., 1997a).

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Sedimentary OM is a mixture of lipids, carbohydrates, proteins, and other biochemical compounds such as lignin in higher plants (Sampei and Matsumoto 2001; Meyers, 2003). Microbial activities begin in recently deposited sediments, mainly near the water-sediment interface. This diagenesis process leads transforming biopolymers into geopolymers/geochemical fossils in sediments synthesised by plants/animals with minor changes (Sun and Wakeham, 1998; Marynowski and Wyszomirski, 2008; Chen et al., 2017). Sedimentary basins are diverse, and the sources and alterations of OM are geographically and temporally variable. Therefore, a variety of elemental, isotopic, and molecular indicators/proxiesin sedimentary OM could reconstruct past environmental changes in the watershed (e.g., Silliman et al., 1996; Versteegh et al., 2004; Suzuki et al., 2010).

2. Organic Geochemical Proxy

2.1 Bulk indicators

Total organic carbon (TOC) is a major proxy to determine OM richness in sediments (Meyers and Ishiwatari, 1993; Meyers, 1997; Hossain et al., 2009). TOC concentrations depend on several factors such as OM production, accumulation, and preservation, occasionally in a complicated manner. TOC (wt. %) values can decline due to clastic dilution under a high rate of sedimentation (e.g., Berner, 1982; Ibach, 1982; Sampei et al., 1997b). In addition, TOC concentrations commonly decrease with the increase of sediment grain size (e.g., Silliman et al., 1996; Meyers, 1997).

C/N ratios are the popular organic geochemical proxy to differentiate sedimentary OM of algae and terrestrial plants (Silliman et al., 1996; Meyers, 1997; Sampei and Matsumoto, 2001). Algae consist of protein-rich substance, whereas terrestrial plants consist of cellulose-rich substance. Algae, thus, record C/N ratios of 4 to 10, and C/N ratios of terrestrial plants are greater than 20 (Meyers and Ishiwatari, 1993; Meyers, 2003). C/N ratios can be partially enhanced during the early diagenesis due to selective decomposition of proteinaceous components. Alternatively, C/N ratios can be partially diminished in marine and fine-grained sediments due to ammonia absorption (Müller, 1977; Hossain et al., 2009). Grain-size distribution of sediments can also impact the C/N ratios. For example, coarse-grained sediments normally contain a larger proportion of terrestrial plant debris than fine-grained sediments (Silliman et al., 1996; Meyers, 2003).

Carbon isotope ratios can distinguish OM sources using paleoproductivity evidence. For example, $\delta^{13}C$ values of C_3 Calvin pathway and C_4 Hatch-Slack pathway plants are -27% and -14%, respectively. In addition, $\delta^{13}C$ values of marine OM range from -20 to -22% (Bender, 1971; Huang et al., 2001). The grain size distribution of sediments does not influence the $\delta^{13}C$ values, and thus, the $\delta^{13}C$ value is a useful OM type indicator with temporal changes indepositional conditions (Meyers, 1997).

Rock-Eval pyrolysis analysis is now frequently used in paleoenvironmental studies to identify kerogen types and their diagenetic alteration routes, based on modified van Krevelen-type diagrams (Tissot and Welte, 1978; Tissot et al., 1980, 1987). As discussed in 3.2, three main kerogen types could be identified from the pyrolysis results of Hydrogen Index (HI) and Oxygen Index (OI). Type I kerogen is primarily derived from algal OM in an anaerobic environment, especially lacustrine. Type II kerogen is typically derived from marine OM in reducing to suboxic environments, and Type III kerogen is predominantly derived from higher plants (Tissot et al. 1980; Jiang et al., 2015; Mashhadi et al., 2015).

2.2 Molecular (biomarkers) indicators

Molecular compounds such as *n*-alkanes and sterol are particularly valuable to paleoenvironmental reconstructions. These compounds, named as “biological markers” or “biomarkers”, are comparatively less sensitive to biodegradation than other OMs. Consequently, aliphatic hydrocarbon components such as *n*-alkanes area vital aspect of paleoenvironmental reconstruction due to its reasonably specific biological origins (Eglinton and Hamilton, 1967; Ficken et al., 2000; Hoffmann et al., 2013; Carr et al., 2014).

The sources of OMs are distinguishable mainly by the chain lengths of their molecular suites. The presence of short-chain *n*-alkanes (i.e., C_{17}, C_{19}, and C_{21}) and long-chain *n*-alkanes (i.e., C_{27}, C_{29}, and C_{31}) indicate algal contributions and land-plant epicuticular waxes, respectively (Eglinton and Hamilton, 1967; Zhou et al., 2005; Castańeda et al., 2009).

The *n*-alkane distributions of submerged and floating macrophytes commonly maximise at C_{23} and C_{25} (Ficken et al., 2000; Nott et al., 2000). The contributions of land-derived OM typically comprise higher amounts of *n*-alkanes compared to aquatic algae (Cranwell, 1990; Meyers and Ishiwatari, 1993). However, *n*-alkane distributions in sediments can help to reconstruct paleovegetational records in the watershed. For example, C_{31} and C_{27}/C_{29} are indicators for grasses and trees dominate watersheds, respectively (Nott et al., 2000; Zhou et al., 2005; Castańeda et al., 2009).

Sterol compositions is another approach to distinguish aquatic and terrestrial organic matters in sediments. In addition, the progressive changes of C_{27}, C_{28}, and C_{29} sterols in a ternary plot indicate the changes of aquatic and terrestrial organic sources (Huang and Meinschein, 1979; Meyers, 1997). This illustration is usually applied to reconstruct sources of OM in sediments deposited at temporal scale, as discussed under Section 3.1.

Polycyclic aromatic hydrocarbons (PAHs) are formed by diagenesis changes and low- and high-temperature combustion. Therefore, PAHs can indicate both paleoecological and paleoenvironmental characteristics (Jiang et al., 1998; Yunker and Macdonald, 2003; Han et al., 2014). PAHs distribution in recent sediments can act as an indication for anthropogenic activities in the watershed (e.g., Meyers, 2003; Yunker and Macdonald, 2003).

3. Organic Geochemical Evidence for Paleoenvironmental Changes in Sri Lanka

3.1 Comparison of the Jurassic OM contributions to the Andigama and Tabbowa Basins

Sri Lanka records two Jurassic sedimentary basins known as the *Andigama* and *Tabbowa Basins* along the northwest onshore margin (Figure 1).

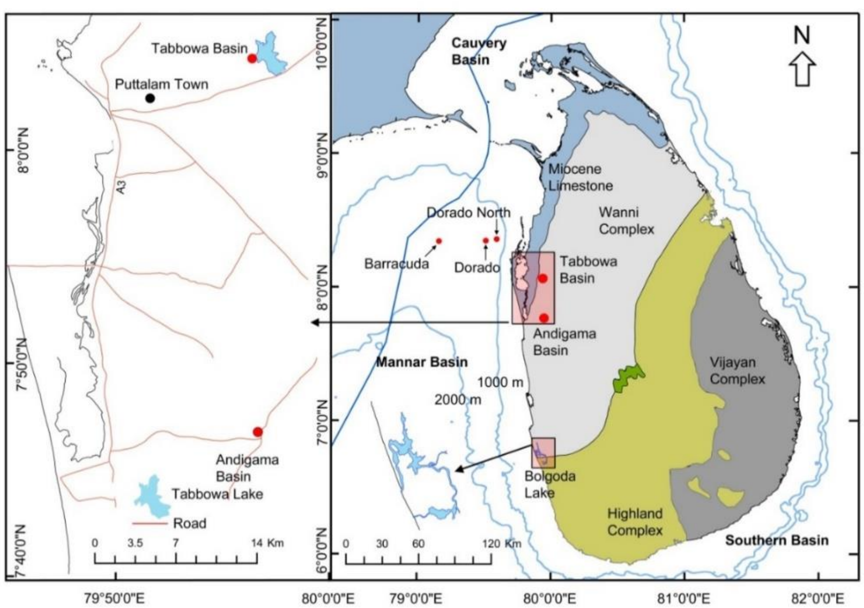

Figure 1: Simplified geological map of Sri Lanka shows the important study sites discussed in this overview (modified after Ratnayake et al., 2017a).

Organic geochemical studies have been concentrated on Andigama mudstone and Tabbowa fine-grained sedimentary rocks (Ratnayake and Sampei, 2005). The *n*-alkanes distribution of the Andigama Basin suggests the deposition of terrestrial higher plant dominated OM with some amount of plankton/algae Oms (Figure 2).

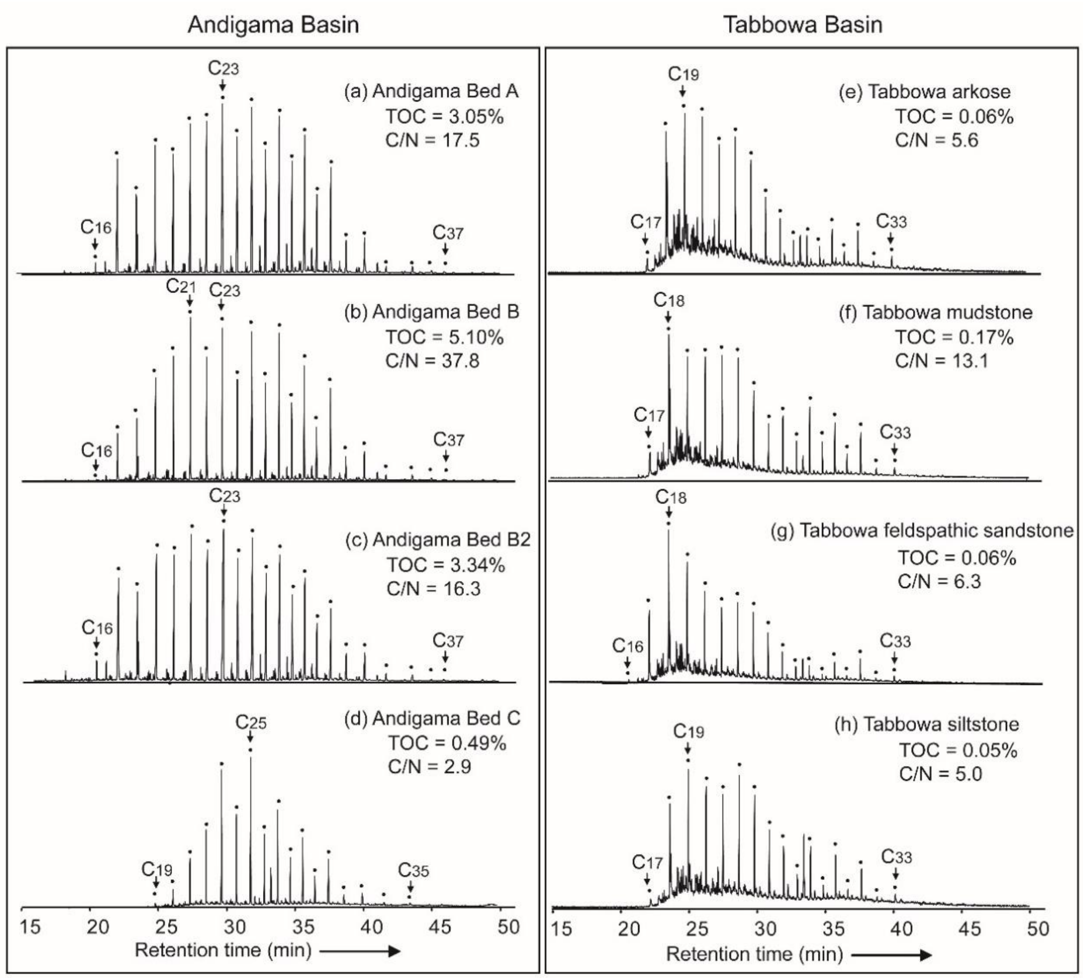

Figure 2: Comparison of representative *n*-alkane distribution in the Andigama and Tabbowa Basins (modified after Ratnayake and Sampei, 2015).

The middle chain length homologs of the Andigama mudstones can be interpreted as the development of lacustrine to swamp environments (e.g., Meyers and Ishiwatari, 1993; Ficken et al., 2000; Nott et al., 2000). However, the *n*-alkanes distribution suggests the deposition of plankton/algae dominated OMs in the Tabbowa Basin (Figure 2). Similarly, the ternary diagram of C_{27}-C_{28}-C_{29} steranes indicates the depositions of terrestrial OMs in Andigama Basin and algae OMs in Tabbowa Basin, respectively (Figure 3).

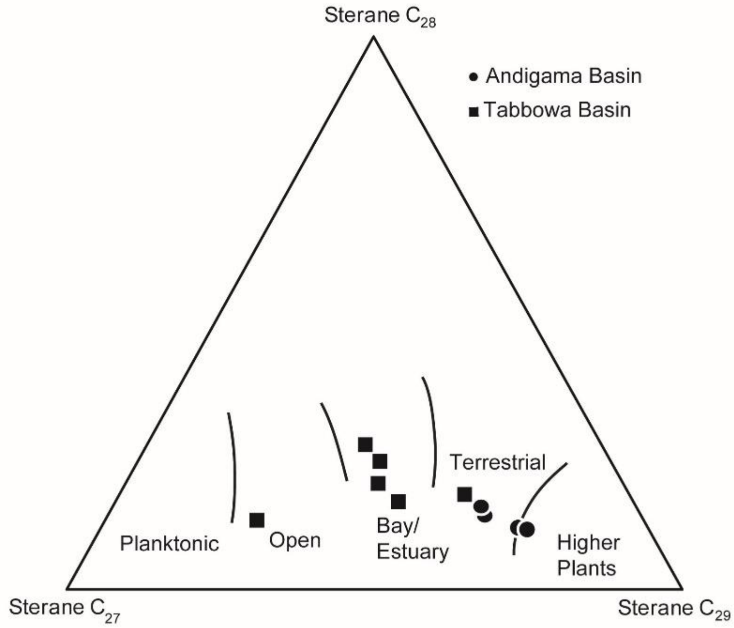

Figure 3: Comparison of OM sources and depositional environments of the Andigama and Tabbowa Basins using the ternary diagram of C_{27}-C_{28}-C_{29}steranes (modified after Huang and Meinschein, 1979; Ratnayake and Sampei, 2015).

Figure 4 illustrates the distribution of PAHs and alkylated phenanthrenes in the Andigama mudstones. Several organic sources indicating biomarkers are observed in PAHs fraction. For example, retene and simonellite can be derived from gymnosperm OM sources such as conifer resins (Jiang et al., 1998; Yunker and Macdonald, 2003; Yunker et al., 2011). Cadalene is also a terrestrial land plants indicator from both angiosperm and gymnosperm (Otto et al., 2002; Simoneit, 2005; Han et al., 2014). However, no angiosperm plants were recorded in the eastern Gondwanaland during the Jurassic (e.g., Aarssen et al., 2000; Friis et al., 2006). Therefore, in this study, cadalene can also be identified as an indicator for gymnosperm plant resins. Furthermore, perylene helps to reconstruct paleoenvironmental characteristics such as deposition of wood-degrading fungi OMs under the temperate humid climatic conditions (Aizenshtat, 1973; Suzuki et al., 2010; Marynowski et al., 2013). Moreover, the detected fluoranthene, pyrene, benzo[a]anthracene, chrysene/triphenylene, benzofluoranthene, benzo[e]pyrene, benzo[a]pyrene, indeno[cd]pyrene, and benzo[ghi]perylene PAHs (Figure 4) indicate medium-temperature ground fire to high-temperature crown fire in the watershed (Jiang et al., 1998; Denis et al., 2012).

Methylated aromatic isomers can be used as organic source indicators (Killops, 1991; Budzinski et al., 1995). For example, 1-methylphenanthreneand 9-methylphenanthrene (Figure 4) is mainly derived from terrestrial OM-rich sediments (Type-III kerogen) (Budzinski et al., 1995; Hossain et al., 2013). Therefore, 1-methylphenanthrene and 9-methylphenanthrenemayhave originated from coniferous gymnosperm in Andigama sediments. The 1, 7-dimethylphenanthrene (Figure 4) can also be originated from gymnosperm resin (Armstroff et al., 2006; Fabiańska et al., 2013).

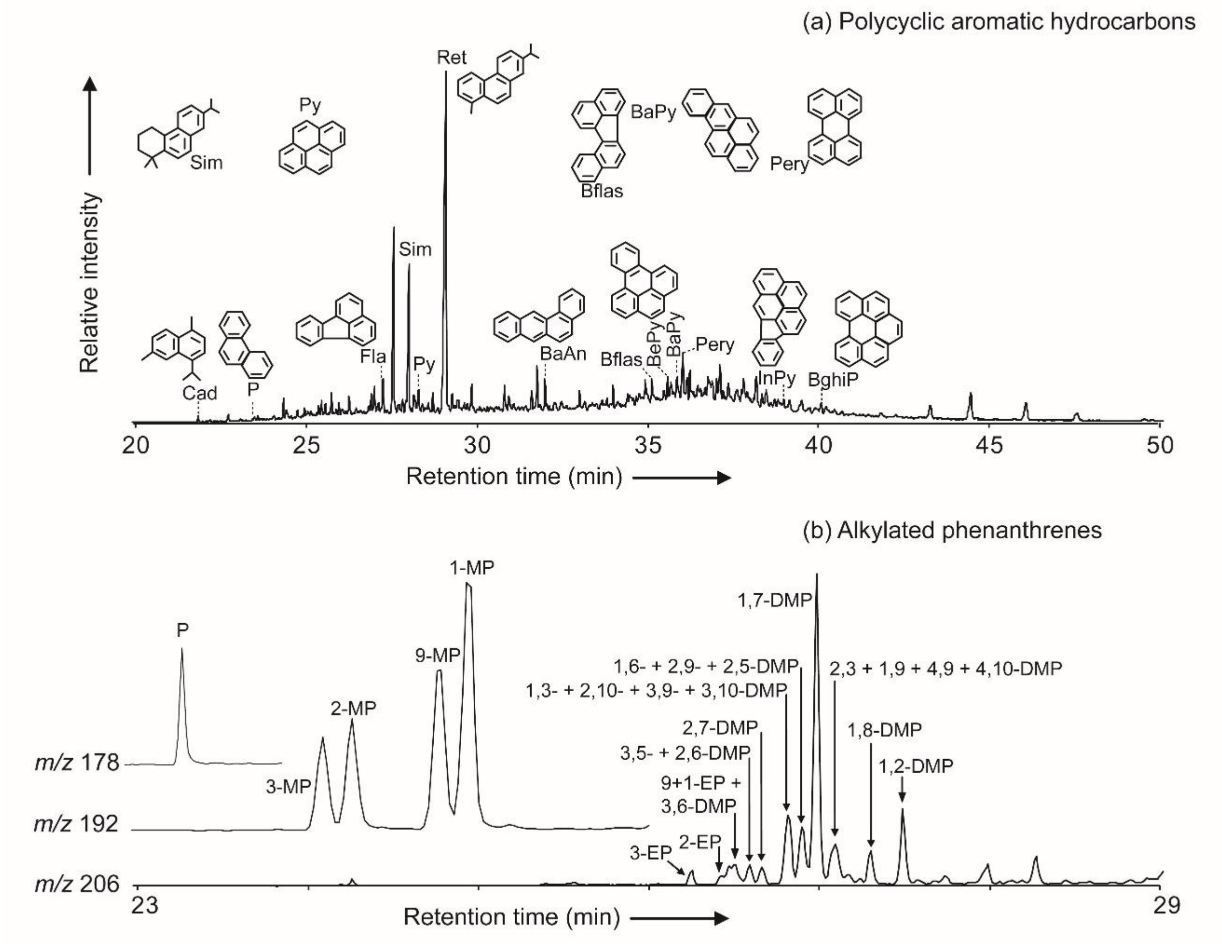

Figure 4: The representative (a) polycyclic aromatic hydrocarbons (PAHs) and (b) alkylated phenanthrenes distributions in the Andigama mudstones (modified after Ratnayake and Sampei, 2015).

P-phenanthrene, Fla-fluoranthene, Py-pyrene, BaAn-benzo[a]anthracene, Chry + Tpn- chrysene/ triphenylene, Bflas-benzofluoranthene, BePy-benzo[e]pyrene, BaPy-benzo[a]pyrene, Pery- perylene, InPy-indeno[cd]pyrene, BghiP-benzo[ghi]perylene, Cad-Cadalene,Sim:simonellite, Ret-retene, MP-methylphenanthrenes and DMP-dimethylphenanthrenes.

3.2 Cretaceous to Paleogene depositional environments in the offshore Mannar Basin

The Mannar Basin is an offshore sedimentary basin between southeast of India and Sri Lanka (Figure 1). The Mannar Basin covers around 45,000 km^2 in Sri Lankan jurisdiction and records organic carbon-rich sediments from the Jurassic to Recent in age. Rock-Eval pyrolysis analysis applies to the petroleum industry and the paleoenvironmental studies, as discussed in 2.1. In this method, types of kerogen and thermal maturity of sediments can be mainly interpreted using standard reference diagrams (e.g., Tissot and Welte, 1978; Tissot et al., 1980, 1987). The modified Van Krevelen diagram (i.e., hydrogen index (HI) versus oxygen index (OI) assists in identifying the dominant kerogen type of OM (Tissot and Welte 1978; Tissot et al. 1980, 1987; Jiang et al., 2015; Mashhadi et al., 2015). Figure 5 depicts that both Cretaceous to Paleogene sediments in the Mannar Basin consist of mixed Type III (mainly) and Type II kerogens. Therefore, geochemical characteristics of the Mannar Basin samples have a high potential for gas hydrocarbon generation.

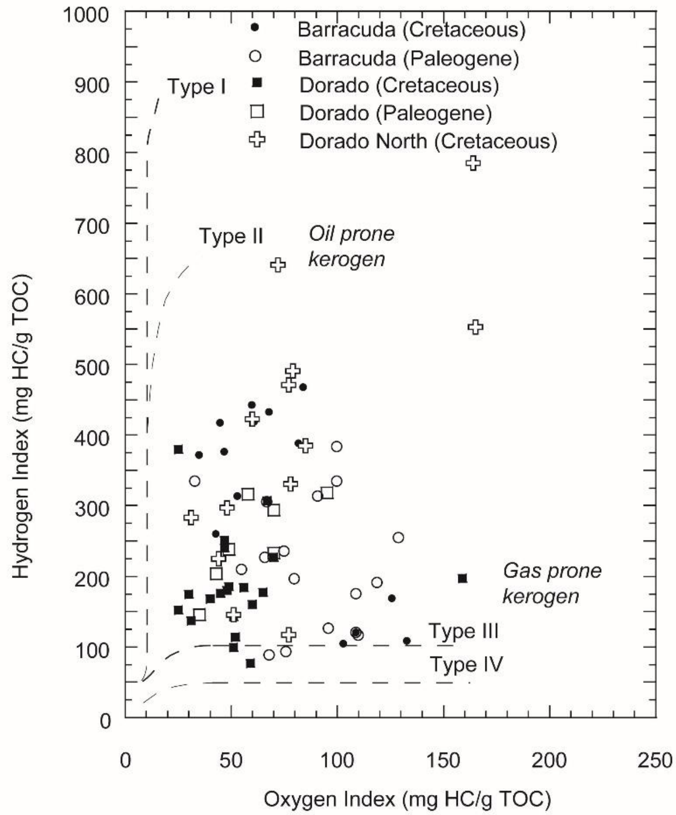

Figure 5: The modified Van Krevelen diagram shows kerogen types in the offshore Mannar Basin (modified after Ratnayake et al., 2018a).

3.3 Holocene paleoenvironmental changes in the coastal Bolgoda Lake

TOC content and C/N ratio of Bolgoda Lake surface sediments range from 1.31 to 6.89% and 10.4 to 15.8, respectively (Ratnayake et al., 2017b, and 2018b). Therefore, it suggests the deposition of algae OM dominant sapropelic sediments with some terrestrial contribution. Also, development of sapropelic condition depends on OM preservation under oxygen-poor to anoxic conditions. Sulfur and organic carbon relationship (C/S ratio) can identify oxic/anoxicdepositional conditions (Berner, 1984; Berner and Raiswell, 1984; Sampei et al., 1997a). The low C/S ratios (2.8±0.8) of Bolgoda Lake surface sediments suggest oxygen-poor stagnant bottom environment (Ratnayake et al., 2017b, 2018b). The core sediments of Bolgoda Lake can divide in to two sections as the lower sedimentary succession (from ca. 7.0 calky B.P. to ca. 2.5 calky B.P.) and the upper sedimentary succession (after ca. 2.5 calky B.P.) based on sedimentary facies, geomorphological observations and chronological and geochemical data (Ratnayake et al., 2017b). Figure 6 illustrates that TOC values have depleted in the lower sedimentary (1%-4%) compared to the upper sedimentary succession (3%-30%), respectively. This major environmental change, thus, indicates nutrient enrichment after ca. 2.5 calky B.P. (Figure 6).It was followed by variations in OM concentration/type and depositional conditions (Ratnayake et al., 2017b). Therefore, this major environmental change is mainly controlled by coastal geomorphological evolution from a bay of paleo-river to a semi-closed and oxygen-depleted brackish lagoon due to middle Holocene sea-level changes (Ratnayake, 2016; Ratnayake et al., 2017b). The reconstruction of paleo-mangrove vegetation using specific molecular indicators assists in identifying sea-level changes in tropics and sub-tropical regions (e.g., Behling et al., 2001; Versteegh et al., 2004). For example, taraxerone can be mainly derived from mangrove vegetation (Versteegh et al., 2004; Koch et al., 2005, 2011). Therefore, based on the distribution of taraxerone biomarker, Ratnayake et al. (2017b) concluded the initial seawater invasion occurred ca. 7.0 calky B.P. along the west coast of Sri Lanka.

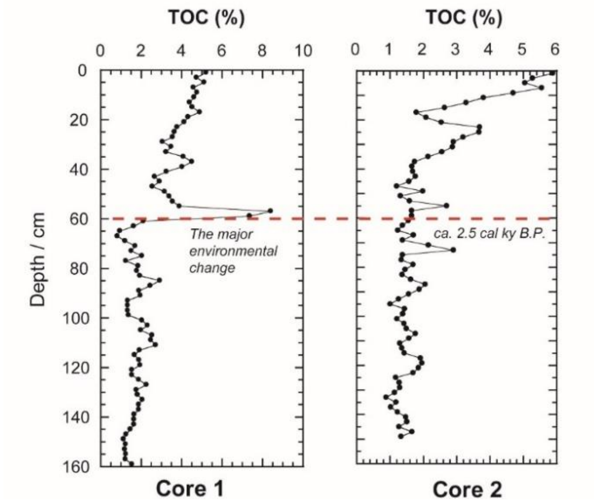

Figure 6: Total organic carbon (TOC) variations of Core 1 and Core 2 in Bolgoda Lake show the major environmental change (modified after Ratnayake et al., 2017b).

The *n*-alkanes distribution can reconstruct not only organic sources but also paleoclimate (Cranwell, 1990; Ficken et al., 2000; Hoffmann et al., 2013). According to Sachse et al. (2006) and Carr et al. (2014), broadleaf tree leaves contain longer chain *n*-alkanes in warm or dry regions compared to in cold or wet areas. Therefore, the distribution of n-C_{29}/n-C_{all} and n-C_{37}/n-C_{all} ratios in Bolgoda Lake sediments (Figure 7) indicates a gradual transition of paleoclimate from colder or wetter (middle Holocene) to warmer or drier climate (late-Holocene).

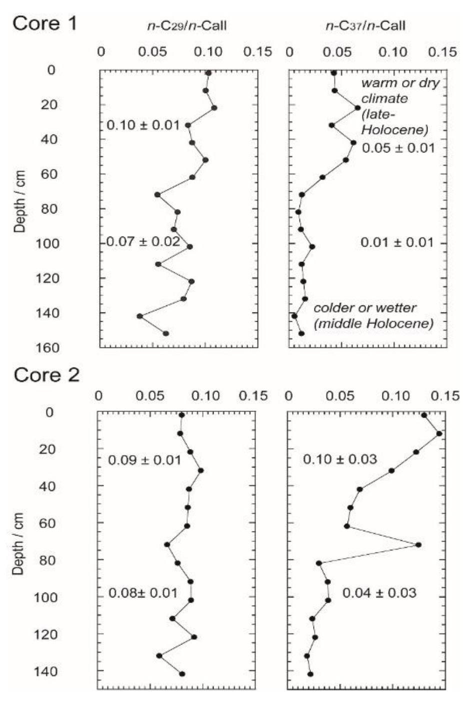

Figure 7: The distribution of n-C_{29}/n-C_{all} and n-C_{37}/n-C_{all} ratios in Bolgoda Lake sediments (modified after Ratnayake et al., 2017b).

4. Future Perspectives

In future research, several additional developments are necessary to quantify paleoproductivity of algae, terrestrial contribution/watershed vegetation, and to understand the transportation process. It is significant to identify novel biomarker information in tropics that can act as a proxy for reconstruction of paleoenvironment/paleoclimate. In addition, early diagenesis incorporation of microorganism in tropical sediments should be investigated in future research. At present, only limited studies have been conducted to reconstruct the paleoenvironmental changes in Sri Lanka. The future investigations should specifically focus on underexplored southeast and northeast coastal regions, and finally, it is crucial to link paleoenvironmentalproxy studies with multidisciplinary numerical/model interpretations.

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