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RESEARCH ARTICLE

Molecular indicators of early stage diagenesis in the tropical coastal Bolgoda Lake, Sri Lanka

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Abstract: Bacterial reactions are one of the major processes that control nutrient cycles in sediments. The aim of this study was to trace the bacterial biomarkers (hopanes) to understand early diagenetic processes in tropical mangrove dominant brackish sediments over modern timescales. Hopane homologues were examined by gas chromatography-mass spectrometry (GC-MS). The results suggested that hopane concentrations were high in Bolgoda Lake surface sediments (average ± SD = 1336.5 ng/g TOC ± 748.7) compared to mangrove mud cores (676.4 ng/g TOC ± 268.5). Therefore, the variation of bacterial biomasses can be controlled by the available reactive organic matter type and depositional environments (sulfate concentration). Total hopane concentrations gradually declined with the depth in core samples. In addition, total hopane concentrations showed moderate and weak correlations with organic matter types in upper (0–60 cm) and lower (> 60 cm) sedimentary successions, respectively. These features in core samples suggest the formation of geohopanoids during diagenesis. The variations of C_{31} hopane isomerisation ratio suggest the minor influence of reworking bacterial biomarkers in the upper sedimentary succession. Therefore, C_{31} 22S (αβ) hopane distribution can indicate soil acidity (chemical origin) in the mangrove dominant swamp.

Keywords: Bacterial biomarkers, hopane, recent sediments, sediment-water interface.

INTRODUCTION

Abundance of archaea (methane-cycling dominate) and bacteria (sulphur-cycling dominate) inhabit a broad range of environments (Rothschild & Maninelli, 2001; Lipp *et al.*, 2008), and carbon and energy sources of these microorganisms are strongly controlled by geochemical settings of the living environment (Inagaki *et al.*, 2006; Chen *et al.*, 2017). Microorganisms are essential components in the biosphere that play an important role to control biogeochemical cycles in natural systems by early diagenesis (Berner, 1982; Sun and Wakeham, 1998; Marynowski & Wyszomirski, 2008). Bacteria can be widely recognised as a main diagenetic agent of recent sediments. In addition, bacterial biomarkers found in geological samples have been widely used to recognise the diagenetic status of sedimentary organic matter (OM), carbon preservation and changes of molecular compositions (Wakeham *et al.*, 1997; Farrimond *et al.*, 2003; Taylor and Harvey, 2011; Chen *et al.*, 2017).

Biological precursors of hopanoids are the product of cell membrane lipids of bacteria from diverse taxonomic groups (Rohmer *et al.*, 1984; Innes *et al.*, 1997; Inglis *et al.*, 2018). Diagenetic processes lead to preserve geohopanoids including hydrocarbons, acids, and hopanols in soils and sediments (Ishiwatari *et al.*, 2014; Ratnayake & Sampei, 2015a; Inglis *et al.*, 2018). Hopanes are well-known less biodegradable precursors of bacteria occurring since the Archean Eon (Brocks *et al.*, 1999; Brazelton *et al.*, 2006; Zheng & Huang, 2017). The distribution of hopane in recent and ancient sediments depends on a number of primary and

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secondary factors (Pan *et al.*, 2008; Mrkić *et al.*, 2011; Taylor & Harvey, 2011; Ratnayake & Sampei, 2015b). However, there are limited examples of early diagenetic incorporations of hopane in tropical mangrove-dominant sediments. This paper reports the analysis of hopane in the tropical brackish Bolgoda Lake sediments in Sri Lanka (Figure 1). In this study, the authors examine the intensity of bacterial activity to understand early diagenesis in the Holocene sediments of Bolgoda Lake, which is known to preserve variable amounts of terrestrial and marine sources (Ratnayake *et al.*, 2017). The authors also investigate bacterial reactivity with reference to different OM types during early diagenesis.

METHODOLOGY

Study area

Bolgoda Lake is the largest brackish lake in Sri Lanka (Ratnayake *et al.*, 2018), and is located in the southwest part of the island (Figure 1a). The lake is situated in a tropical zone (annual average temperature = 27 °C). The study area receives an annual average precipitation of 2500 mm. The lithology and ages of the sedimentary sequences can be divided into two major sections. The lower sedimentary succession (60–160 cm) is composed of the middle to late Holocene (from ~7.0 ky B.P. to ~2.5 cal ky B.P.) grey fine-grained sediments. The upper sedimentary succession (0–60 cm) is consisted of the late Holocene (from ~2.5 cal ky B.P. to the Recent) black fine-grained sediments with abundance of reworking terrestrial wood fragments (Ratnayake *et al.*, 2017). Total organic carbon (TOC) values suggest a significant enhancement of productivity in the upper sedimentary succession compared to the moderately productive lower sedimentary succession (Ratnayake *et al.*, 2017). In addition, the depositional environment of the study area was drastically changed from inner bay of larger paleoriver (in the lower sedimentary succession) to semi-closed estuary of local streams (in the upper sedimentary succession) at the end of middle Holocene highstand ca. 2.5 cal ky B.P. (Ratnayake *et al.*, 2017). Sedimentological observations and geochemical analysis in Bolgoda Lake illustrated the considerable teleconnection to the regional and global climatic changes such as middle Holocene sea-level rise and variations of northern hemisphere monsoon (Ratnayake *et al.*, 2017).

Materials and methods

Surface sediments (n = 28) and three sediment core samples (core 1 = 160 cm, core 2 = 150 cm and core 3 = 110 cm) were collected from the Bolgoda Lake (Figure 1b). In addition, four sub-aerial mangrove mud sediment core samples (VSC 1 = 15 cm, VSC 2 = 15 cm, VSC 3 = 15 cm and VSC 4 = 11 cm) were also collected along the Weras freshwater stream (Figure 1b).

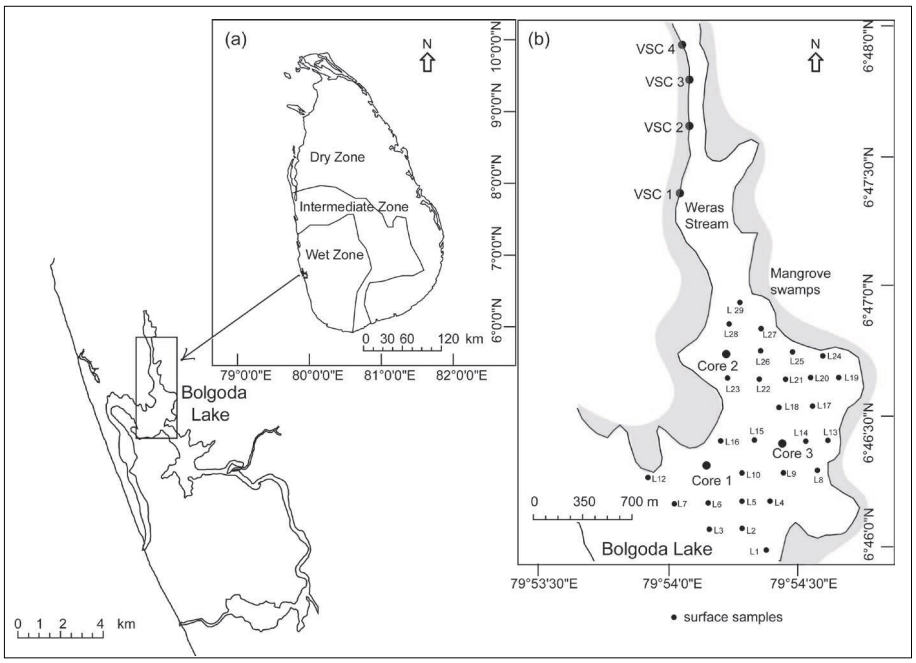

Figure 1: Location map, surface sampling and coring sites of the study area (modified after Ratnayake *et al.*, 2017)

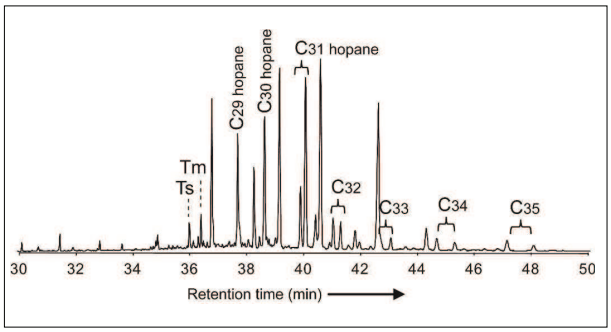

Figure 2: Representative *m/z* 191 mass chromatograms showing the distributions of hydrocarbons in the aliphatic hydrocarbon fraction. (Sample: surface sediments L 17)

Sedimentary OM were extracted from the powdered samples using the Soxhlet apparatus for 24 hours using dichloromethane and methanol (9:1 v/v) solvent system. Elemental sulphur was removed using activated copper granules. The extracts were dried at room temperature. The dried extractable OM were separated into aliphatic and aromatic hydrocarbons by a thin layer chromatography plate (silica gel 60 PF_{254} containing gypsum). The hydrocarbons were separated after washing with hexane. The extracted OM were directly used for gas chromatography (GC: Shimadzu 2010) coupled with a mass spectrometer (MS: Shimadzu GCMS-QP 2010). The GC was prepared with a fused silica capillary column (30 m × 0.25 mm DB 5MS). The temperature injection system was programmed from 50–300 °C at the rate of 8 °C min^{-1} and kept at 300 °C for 30 min with pure helium as a carrier gas. The MS was scanned every 0.5 s over *m/z* 50 to 850 at an electric energy of 70 eV. The spectral data were recorded in a computer system. The organic compounds were identified by comparison of GC retention times and mass spectra with published data. The compounds were quantified (ng/g TOC) by comparing the chromatogram peak area to the area of *n*-C_{24} tetracosane.

Table 1: Distribution of representative hydrocarbons in *m/z* 191 mass chromatograms of surface sediments in Bolgoda Lake

Sample	C_{31}-22S ng/g TOC	C_{31}-22R ng/g TOC	Total hopane ng/g TOC	C_{31} hopane (22S/22R)	22S/(22S + 22R) for C_{31}
L 1	377.8	799.6	2550.2	0.47	0.32
L 2	292.2	723.7	2008.1	0.40	0.29
L 3	95.8	201.7	561.5	0.48	0.32
L 4	220.0	632.1	1590.1	0.35	0.26
L 5	177.5	453.0	1238.5	0.39	0.28
L 6	255.9	509.9	1689.4	0.50	0.33
L 7	538.0	1060.0	3470.6	0.51	0.34
L 8	298.1	635.3	1916.9	0.47	0.32
L 9	202.0	553.2	1452.6	0.37	0.27
L 13	109.5	387.2	889.1	0.28	0.22
L 14	179.7	431.7	1214.4	0.42	0.29
L 15	285.7	776.8	2004.1	0.37	0.27
L 17	196.3	526.1	1388.0	0.37	0.27
L 19	100.3	275.3	682.0	0.36	0.27
L 21	150.0	476.5	1094.8	0.31	0.24
L 23	108.3	333.2	801.1	0.33	0.25
L 24	78.0	224.5	566.7	0.35	0.26
L 25	77.7	255.2	615.8	0.30	0.23
L 27	88.3	269.2	636.8	0.33	0.25
L 28	134.3	454.1	1028.0	0.30	0.23
L 29	82.5	306.2	666.5	0.27	0.21
Average	192.8	489.7	1336.5	0.38	0.27
Standard deviation	117.3	221.7	748.7	0.07	0.04

Table 2: Distribution of representative hydrocarbons in *m/z* 191 mass chromatograms of mangrove mud core samples

Sample	C_{31}-22S ng/g TOC	C_{31}-22R ng/g TOC	Total hopane ng/g TOC	C_{31} hopane (22S/22R)	22S/(22S + 22R) for C_{31}
VSC 1 (0–2 cm)	116.7	121.6	673.1	0.96	0.49
VSC 1 (6–8 cm)	150.2	124.0	805.9	1.21	0.55
VSC 1 (13–15 cm)	70.0	82.6	411.8	0.85	0.46
VSC 2 (0–2 cm)	192.4	191.6	1041.0	1.00	0.50
VSC 2 (6–8 cm)	183.1	207.1	1093.3	0.88	0.47
VSC 2 (13–15 cm)	55.2	66.6	284.0	0.83	0.45
VSC 3 (0–2 cm)	92.0	86.9	518.5	1.06	0.51
VSC 3 (6–8 cm)	152.5	154.3	882.9	0.99	0.50
VSC 3 (13–15 cm)	75.0	72.1	403.8	1.04	0.51
VSC 4 (0–2 cm)	139.0	119.9	787.4	1.16	0.54
VSC 4 (6–8 cm)	108.8	77.3	538.3	1.41	0.58
Average	121.4	118.5	676.4	1.04	0.51
Standard deviation	46.0	48.3	268.5	0.17	0.04

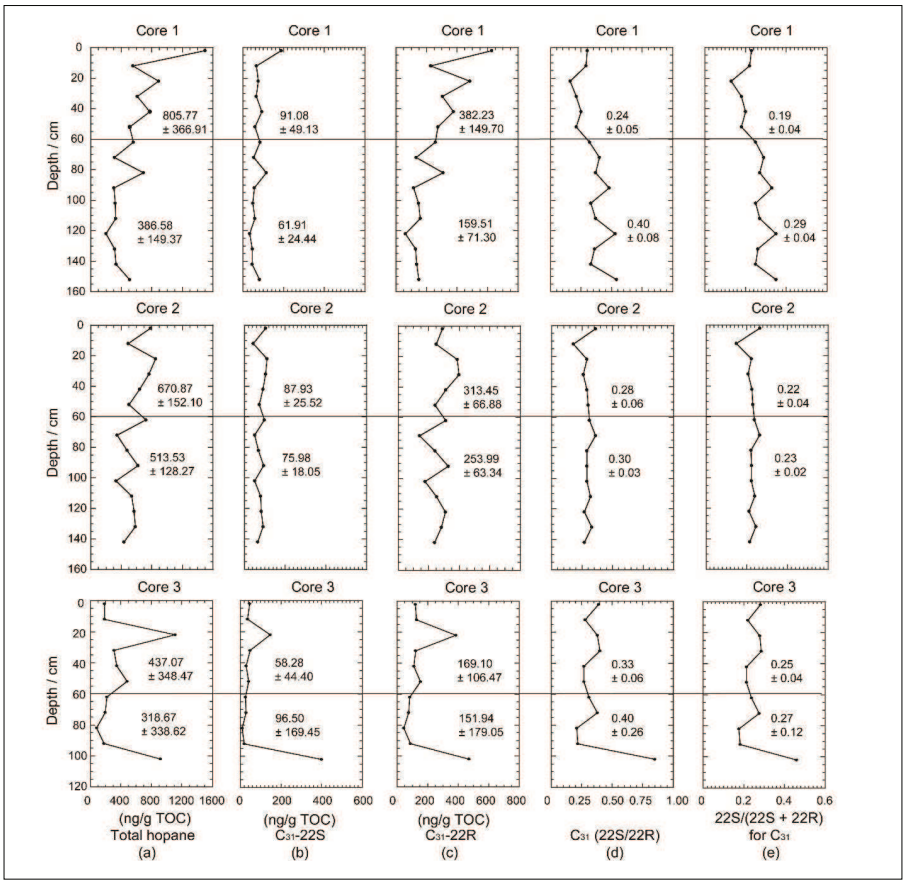

Figure 3: Depth profiles showing the distribution of (a) total hopane[a]; (b) C_{31} hopane – 22S isomer concentration; (c) C_{31} hopane – 22R isomer concentration; (d) C_{31} hopane (22S/22R) ratio and (e) C_{31} hopane 22S/(22S + 22R) ratio in core samples
Total hopane[a] = C_{29} hopane + C_{30} hopane + C_{31} hopane 22S isomer + C_{31} hopane 22R isomer

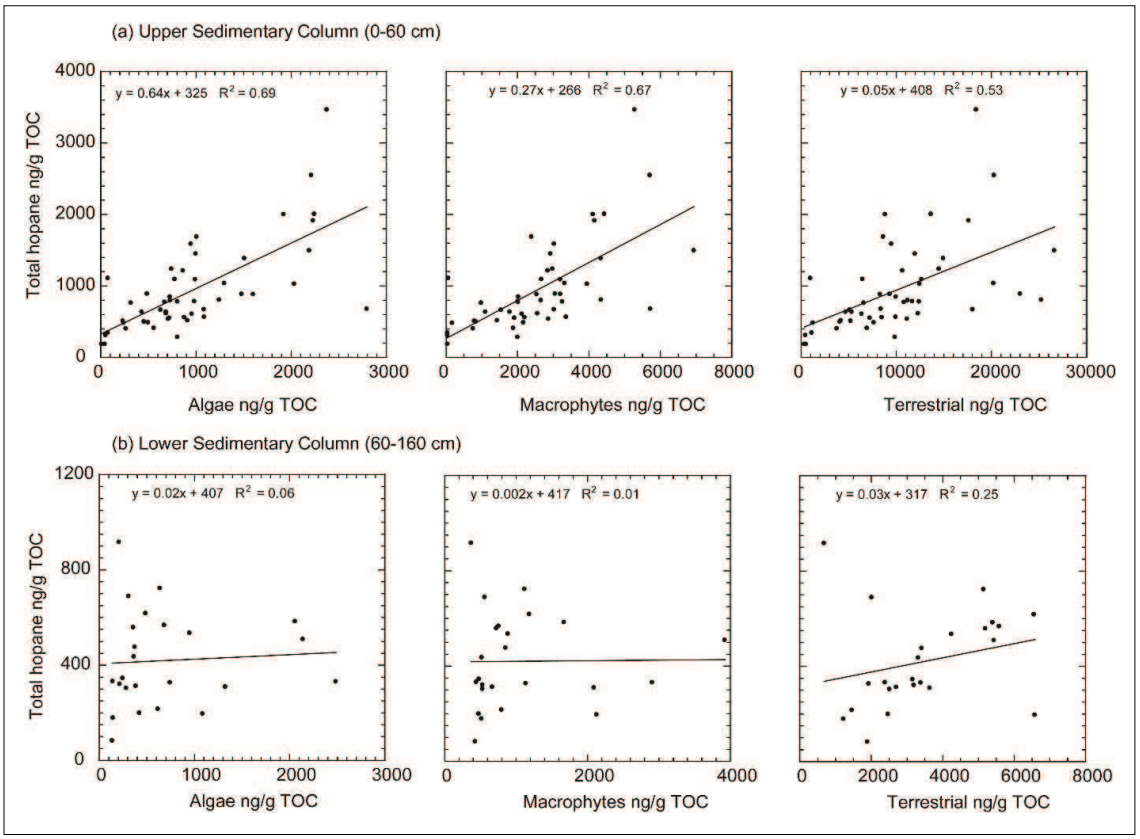

Figure 4: Relationship between total hopane *vs* algae[a] OM, macrophytes[a] OM and terrestrial[a] OM concentration in (a) the upper sedimentary succession (0 – 60 cm) and (b) the lower sedimentary succession (60 – 160 cm) of Bolgoda Lake ([a] algae, macrophytes and terrestrial data are from Ratnayake *et al.*, 2017).

RESULTS AND DISCUSSION

The characteristic partial *m/z* 191 mass chromatograms of triterpanes are shown in Figure 2. The C_{29}, C_{30} and C_{31} components are the most abundant hopane homologues in the *m/z* 191 mass fragmentograms. Homohopane concentrations are generally decreased with increasing carbon number from C_{31} to C_{35} homologues (Figure 2). However, the marked increase in C_{33}-22S hopane homologue can be interpreted as possible co-elutes with cedranes or its derivatives. The sesquiterpenoids cedranes or its derivatives are characteristic fossil resins of trees and shrubs (Grantham & Douglas, 1980; Pereira *et al.*, 2009), and is rare in algal rich microbial mats (Elias *et al.*, 1997).

The biomarker results of surface sediments and mangrove mud cores are summarised in Tables 1 and 2, respectively. The vertical distributions of biomarker proxies of core samples are shown in Figure 3. The cross plots of total hopane *vs* *n*-alkane based algae (sum of lower molecular weight nC_{17}, nC_{19} and nC_{21}), macrophytes (sum of middle molecular weight nC_{23} and nC_{25}) and terrestrial OM (sum of higher molecular weight nC_{27}, nC_{29}, nC_{31}, and nC_{33}) concentrations in the upper and lower sedimentary successions are summarised in Figure 4.

Total hopane distribution in surface and mangrove sediments

Total hopane concentrations in surface sediments ranged from 561.5 ng/g TOC to 3470.6 ng/g TOC (average ± SD = 1336.5 ng/g TOC ± 748.7) (Table 1). The total hopane concentrations in mangrove mud cores ranged from 284.0 ng/g TOC to 1093.3 ng/g TOC (average ± SD = 676.4 ng/g TOC ± 268.5) (Table 2). The variation of total hopane concentrations indicates the preservation of more bacterial diagenetic products (geohopanoids) in the Bolgoda Lake surface sediments

compared to the sub-aerial mangrove mud cores. Consequently, the total hopane distribution can be controlled by changes in OM type and depositional environments in these two depositional systems as follows. It is shown that Bolgoda Lake surface sediments consist of a significant amount of organic carbon [average TOC (%) = 4.4 ± 1.4] from algae and terrestrial organic matter [average C/N ratio = 11.8 ± 1.3; Ratnayake *et al.*, 2017]. The mangrove mud cores also indicate great accumulation of organic carbon [average TOC (%) = 11.8 ± 3.2] from mangrove root-biomass [average C/N ratio = 14.2 ± 1.6; Ratnayake *et al.*, 2017]. In addition, Bolgoda Lake surface sediments consist of anoxic to oxygen-poor brackish depositional environment (average C/S ratio = 2.4 ± 1.2). However, sub-aerial mangrove mud cores indicate a freshwater depositional condition (average C/S ratio = 129.2 ± 383.6) with cyclic seawater invasion (Ratnayake *et al.*, 2017, 2018). It has been widely discussed that bacterial reactions depend on a number of factors including reactive organic compounds mainly and dissolved sulfate concentration (e.g., Berner & Raiswell, 1984; Picard *et al.*, 2018). Algae OM are favourable mats for sulfate-reducing bacteria compared to lignin-rich terrestrial OM (Gong & Hollander, 1997; Sampei *et al.*, 1997; Zheng & Huang, 2017; Picard *et al.*, 2018). In this study, reactive algae OM concentration (regardless of carbon burial) and sulphate availability were relatively high for bacterial sulphate reduction in Bolgoda Lake surface sediments. Therefore, a significant amount of bacterial sulphate reduction can be expected in brackish surface sediments of Bolgoda Lake compared to sub-aerial mangrove mud cores. In nature, this fractionation leads to the breakdown of organic matter to methane and carbon dioxide in freshwater swamp ecosystems elsewhere (Berner, 1985; Bridgham and Richardson, 1992; Brix *et al.*, 2001).

Total hopane distribution in the lake core sediments

The total hopane distributions in core samples are shown in Figure 3a. Total hopane concentrations decreased with depth and flattened out to almost constant values in Core 1 and Core 2, and average hopane concentrations were high in the upper sedimentary succession of all core samples (Figure 3a). This pattern indicates the reduction of bacterial decomposition with increasing depth. However, the general trend has somewhat deviated in Core 3, which is composed of a significant amount of terrestrial OM (Ratnayake *et al.*, 2017).

The sediment/water interface is characterised by highly abundant microbial communities (Klump & Martens, 1989; Sun & Wakeham, 1998; Chen *et al.*, 2017). Aerobic microorganisms live within a few centimeters below the sediment/water interface. It depends on penetrated dissolved oxygen to bottom sediments. Sedimentation accumulation rate of Bolgoda Lake is 0.24 mm/year (Ratnayake *et al.*, 2017). Such a condition provides sufficient time for aerobic bacterial degradation in the sediment/water interface. After that, dysaerobic or anaerobic degradation arises due to prevention of oxygen migration to deeper sediment levels. Therefore, total hopane variations in the down core profiles can indicate the intensity of bacterial activity in sediments. However, bacterial activity depends on a number of other factors as discussed in the previous section.

The authors considered the influence of OM type for bacterial activity in the upper (0-60 cm) and lower (60-160 cm) sedimentary successions. Moderate correlations were identified between total hopane and OM concentrations in the upper sedimentary succession (Figure 4a). Further, it suggests an increasing trend of connection between bacterial activities and different OM types as follows: terrestrial plants ($R^2 = 0.53$) < macrophyte plants ($R^2 = 0.67$) < algae ($R^2 = 0.69$) (Figure 4a). However, hopanes did not show significant correlations with different OM types in the lower sedimentary succession (Figure 4b). This trend suggests the formation of more resistant geopolymers (macromolecular) after the progressive sulphate reduction in the studied samples. In summary, structural modifications, rearrangements and stereochemical changes of sedimentary OM can progressively occur in modern sedimentary successions during the early diagenetic reactions (e.g., Haven *et al.*, 1987; Innes *et al.*, 1997; Farrimond *et al.*, 2003; Inglis *et al.*, 2018).

Effects of reworking geohopanoids in the upper sedimentary succession

The distribution of hopane can also be associated with environmental factors. A significant amount of 22S C_{31} hopane isomer was detected in recent surface sediments (192.8 ng/g TOC ± 117.3; Table 1) and mangrove mud cores (121.4 ng/g TOC ± 46.0; Table 2). Similarly, 22S (αβ) configuration has been predominantly detected in recent marshy peat bogs, lignite and forest soils elsewhere (Dehmer, 1995; Crossman *et al.*, 2001; Pancost *et al.*, 2003). Previous studies clearly demonstrated that early diagenetic origin of 22S (αβ) hopane can occur *via* acid-catalysed reactions (chemical origin) in modern acidic peat bogs and forest soils (Innes *et al.*, 1997; Pancost *et al.*, 2003; Ishiwatari *et al.*, 2014). In this study, the αβ/ββ (22S/22R) ratio is significantly high in mangrove mud cores (1.04 ± 0.17; Table 2) compared to Bolgoda Lake

surface sediments (0.38 ± 0.07; Table 1). Consequently, C_{31} hopane 22S/22R ratio suggests a typical high acidity in the mangrove-dominant swamp sediments. However, hopane C_{31} 22R and 22S homologues have been traditionally identified as biologically- and thermally-derived isomers, respectively (Farrimond *et al.*, 1998; Pan *et al.*, 2008; Zheng & Huang, 2017).

According to Ratnayake *et al.* (2017), the upper sedimentary succession of core samples was characterised by deposition of reworking OM from the watershed. Therefore, thermally-derived 22S hopane can also be deposited in recent sediments as reworking OM, oil pollution or seepages. However, in this study, the increment in the concentration profile of C_{31} 22S hopane isomer was not significantly increased compared to C_{31} 22R hopane isomer in the upper sedimentary successions (i.e., 0–60 cm in depth, from ~2.5 cal ky B.P. to the Recent) (Figures 3b and 3c). Therefore, 22S C_{31} hopane distributions in core samples can be interpreted as chemical origin rather than thermal origin (16–60 cm in depth), and possible contamination by thermally derived reworking OM in the uppermost sedimentary succession (i.e., 0–16 cm in depth, after the 15th century) based on literature (Ratnayake *et al.*, 2017). The authors further examined the effects of reworking OM based on maturity indicating biomarker proxy under the normal thermal stress. The 22S/(22S + 22R) isomerisation ratio of C_{31} hopane is frequently used as a maturity indicator (Farrimond *et al.*, 1998; Pan *et al.*, 2008; Inglis *et al.*, 2018). Similarly, C_{31} hopane isomerisation ratio did not show a significant increment in the reworking OM abundant upper sedimentary succession (Figure 3e). Consequently, 22S C_{31} hopane isomer was mainly originated by chemical reactions.

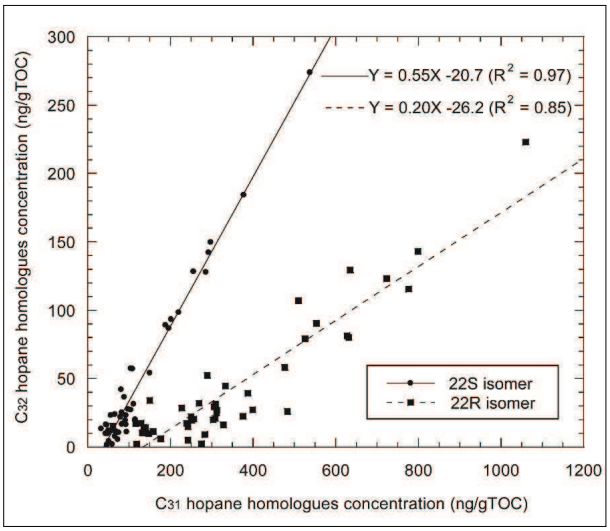

Figure 5: Relationship between 22S and 22R C_{31} hopane homologues *vs* 22S and 22R C_{32} hopane homologues

The plot of 22S and 22R C_{31} hopane homologues *vs* 22S and 22R C_{32} hopane homologues shows a strong linear relationship with each other (Figure 5). It suggests that C_{31} and C_{32} hopane homologues cannot be co-eluted with unknown compounds. Therefore, in summary, the gradual reduction of biologically-inherited C_{31} 22R isomer with depth suggests a general diagenetic trend with no significant accumulation of reworking bacterial biomarkers (geohopanoids) in the upper sedimentary succession.

CONCLUSION

The significant amounts of hopane in the brackish Bolgoda Lake surface sediments (1336.5 ng/g TOC ± 748.7) indicate prominent bacterial sulfate reduction compared to freshwater sub-areal mangrove sediments (676.4 ng/g TOC ± 268.5). It is followed by the reactive algae OM type and sulfate availability. Hopane distributions are moderately controlled by OM type in the upper sedimentary succession. However, OM type is not significantly influenced in the lower sedimentary succession may be due to the formation of geohopanoids (macromolecular) during early diagenesis. The variation of C_{31} hopane 22S/22R ratio can indicate acidity of soils in the tropical mangrove dominant swamp. The C_{31} hopane 22S isomer specifies almost a constant distribution throughout the whole sedimentary succession suggesting chemical origin. Hopane distribution in the Bologoda Lake is characterised by normal diagenetic evolution rather than the preservation of ancient bacterial biomarkers in the upper sedimentary succession.

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