



Predominance of archaea-derived hydrocarbons in an Early Triassic microbialite



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ABSTRACT

We investigated the distribution of lipids in Lower Triassic sedimentary rocks (252–247 myr) from South China, including a shallow water microbialite in the uppermost section of the outcrop. Archaeal derived hydrocarbons were the major constituents of the microbialite from the latest Early Triassic. Among these, we detected (i) abundant C₄₀ acyclic and monocyclic biphytanes (possibly derived from glycerol dialkyl glycerol tetraether lipids) and their degradation products, C_{30–39} pseudohomologues and (ii) a C₂₅ head-to-tail linked (regular) isoprenoid hydrocarbon [possibly derived from dialkyl glycerol diether lipids (DGDs)] and its degradation products, C_{21–24} pseudohomologues and abundant pristane and phytane. Through combination of compound-specific stable carbon isotope analysis of isoprenoid hydrocarbons, which had average δ¹³C values of –35‰ to –30‰, and their molecular distribution, it was not possible to unambiguously define the archaeal source for the biphytanes in the microbialite. The δ¹³C values for pristane and phytane were similar to those for head-to-tail linked C_{21–25} isoprenoids; potential source organisms for these compounds were halophilic archaea. Except for methane seep microbialites, no other ancient or recent phototrophic microbialites have been reported to contain predominantly archaeal isoprenoid hydrocarbons. Our findings suggest the presence of a new type of microbialite.

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1. Introduction

The Early Triassic, which followed the latest Permian mass extinction, is the interval in which microbialites and other unusual sedimentary structures, such as seafloor fans, occurred repeatedly in four successive events, the late Griesbachian to Dienerian, the early Olenekian and the latest Early Triassic (Baud et al., 2007).

Around the end of the Early Triassic, microbialites (with a thrombotic texture) occurred on an isolated pelagic plateau of the Western Tethys (Halstatt Limestone of Dobrogea, Romania; Atudorei et al., 1997), and microbialites (stromatolites and thrombolites) occurred in the Virgin Limestone, while seafloor fans (Union Wash Formation; Pruss and Bottjer, 2004, 2005; Pruss et al., 2005) occurred on the eastern Panthalassa margin (western USA). Pruss et al. (2005) and Pruss and Bottjer (2004) suggested that the formation of microbialites is associated with an alkalinity increase in seawater because they noticed that they occur during transgressive phases, suggesting that the flooding of shallower depositional environments by deeper alkaline ocean waters fosters

their formation (Pruss et al., 2005). Grotzinger and Knoll (1995) first proposed the upwelling of deep alkaline waters onto a shallower shelf as a mechanism for carbonate precipitation on the shelf, as the alkalinity increased. Because deep alkaline water may have contained abundant H₂S, mixing with surface water would have poisoned any metazoans present; the resulting depositional environments lacking metazoans facilitated the formation of microbialites (Fischer, 1965; Awramik, 1971, 1992; Riding, 2006). This mechanism explains the occurrence of carbonate fans in the late Early Triassic (Woods et al., 1999).

In recent studies of microbialites, investigations based on microbial 16S rRNA genes have indicated that modern microbialite-forming mats contain a plethora of microbial species, with species richness exceeding several thousand (e.g. Papineau et al., 2005). Often, the communities of so-called shallow water phototrophic mats contain 5–7 key groups of microbes (Dupraz et al., 2009), including photolithoautotrophs (cyanobacteria), aerobic heterotrophs, fermenters, anaerobic heterotrophs [predominantly sulfate reducing bacteria (SRB)], sulfide oxidizers, anoxygenic phototrophs (purple and green sulfur bacteria) and methanogens.

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The metabolism of cyanobacteria, anoxygenic phototrophs and SRB may promote the precipitation of CaCO_3 , whereas aerobic heterotrophs, sulfide oxidizers and fermenters promote the dissolution of CaCO_3 (Dupraz et al., 2009). The interplay of all the metabolic activities in a single mat ultimately determines whether CaCO_3 is precipitated or dissolved (Dupraz et al., 2009). Except for non-phototrophic methane-seep microbialites (e.g. Peckmann et al., 1999, 2009; Thiel et al., 1999, 2001; Goedert et al., 2003), archaea are usually minor constituents (ca. 10%) of modern microbial mat communities (e.g. Papineau et al., 2005).

Biomarker analysis has been applied to few other ancient non-methane seep microbialites (hereafter referred to as “microbialites” because this study focusses on non-methane seep microbialites) (e.g. Keupp et al., 1993; Malinski et al., 2009; Allen et al., 2010). Such studies have revealed contributions from various bacteria (including Cyanobacteria), algae, higher plants and archaea to microbialites. The contribution of archaeal lipids has been minor (Keupp et al., 1993; Allen et al., 2010) to moderate (Heindel et al., 2013) or undetectable (Malinski et al., 2009). Most studies of microbialites have shown a predominance of *n*-alkanes or *n*-fatty acids. However, it has been noted that the early Early Triassic microbial mats contain a relatively high proportion (ca. 20%) of archaeal-derived isoprenoid hydrocarbons, including biphytane and pseudohomologues, and C_{21-25} regular isoprenoids (Heindel et al., 2013). Benthic microbial mats occur in hypersaline low O_2 marine environments, although Heindel et al. (2013) imply that the farnesane in microbialites could have been produced by benthic and/or planktonic anoxygenic phototrophic bacteria.

Here, we report on the biomarkers in the hydrocarbon fractions from an upper Triassic microbialite in Chaohu, South China and constrain the environmental changes that arose during the formation of the microbialite.

2. Geological setting and methods

Samples were obtained from Lower Triassic sections in North Pingdingshan, West Pingdingshan and South Majiashan in Chaohu, Anhui Province, South China (Fig. 1). During the Early Triassic, South China was located in the low latitude eastern Tethys Sea.

The depositional environments of the Early Triassic successions now exposed in the study area included a distal offshore ramp, proximal ramp, lower shore face and marginal sea, with a water depth ranging from 20 to 200 m (Chen et al., 2011). The upper unit contains the microbialite, which has a carbonate content of >90%. The uppermost part of this section is overlain by evaporite-solution breccia sediments of Middle Triassic age (Tong and Zhao, 2005), which include large traces similar to the fish-swimming trail *Undichna* (Chen et al., 2011). These lines of evidence suggest that the upper unit of the upper sequence in the section was a marginal sea with restricted circulation (Chen et al., 2011).

The 118 samples include carbonates, shales and marls from the 250 m thick sedimentary sequence at Majiashan–Pingdingshan (Fig. 1). They were analyzed according Oba et al. (2006). Briefly, after any apparent surface contamination was cut away, the samples (10–100 g) were powdered. Solvent extracts were obtained with a Soxhlet apparatus using CH_2Cl_2 :MeOH (7.5:1 v/v) for 48 h. Each extract was dried over Na_2SO_4 and concentrated by evaporation under reduced pressure. The concentrated extract was separated into nine fractions (Fs) on a silica gel column (0.6 g silica, 63–200 μm mesh) as follows: 2 ml hexane (F1a); 4 ml hexane (F1b); 3 ml hexane/toluene, 3:1 v/v (F2); 3 ml hexane/EtOAc, 19:1 v/v (F3); 3 ml hexane/EtOAc, 9:1 v/v (F4); 3 ml hexane/EtOAc, 17:3 v/v (F5); 3 ml of hexane/EtOAc, 4:1 v/v (F6); 3 ml hexane/EtOAc, 3:1 v/v (F7); and 10 ml MeOH (F8). The aliphatic hydrocarbon (F1a) and aromatic hydrocarbon fractions (F1b + F2) were analyzed with gas chromatography–mass spectrometry (GC–MS). The branched and cyclic compounds were separated from the aliphatic hydrocarbon fraction using 5A molecular sieve, on the basis of the method by Murphy (1969).

Biomarkers were assigned and quantified using an Agilent 6893 GC instrument interfaced with an Agilent 5973 mass selective detector with an ionizing electron energy of 70 eV, and scanning from *m/z* 50 to 600 with a scan time of 0.34 s. A fused silica HP-5MS column (30 m, 0.25 mm i.d., 0.25 μm film thickness) was used with He as carrier gas. The samples were injected at 50 °C and maintained at that temperature for 1.0 min. The temperature was then raised to 120 °C at 30 °C/min and to 310 °C (held 20 min) at 5 °C/min.

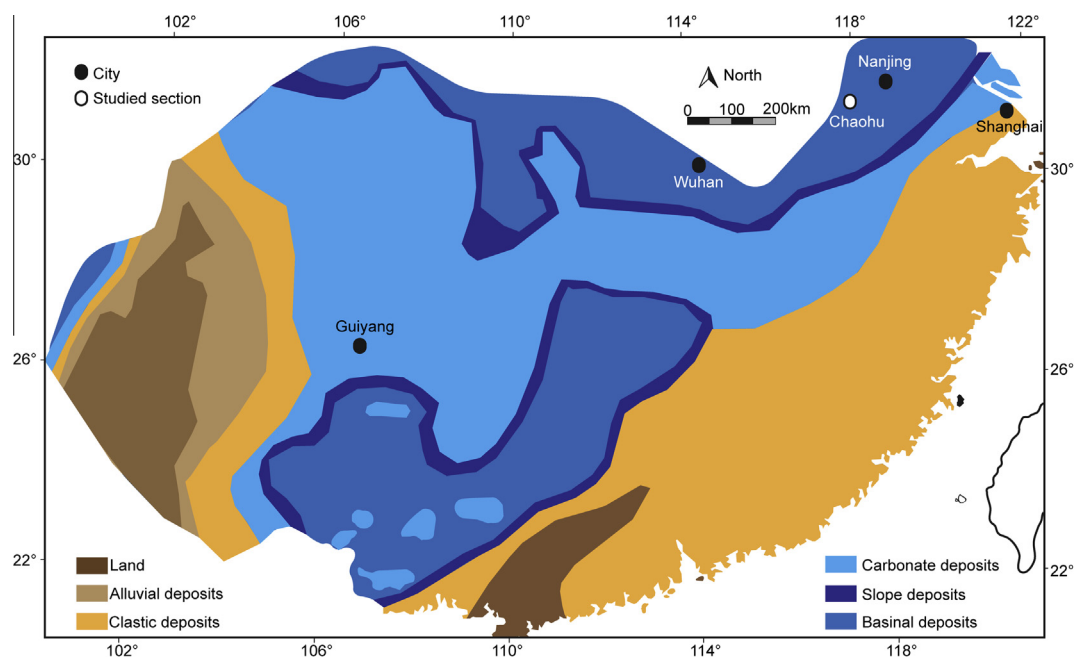


Fig. 1. Paleogeographic map of South China (modified from Song et al., 2013) and location of the Chaohu section.

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