Geophysical Research Abstracts
Vol. 17, EGU2015-1493, 2015
EGU General Assembly 2015

Methanogenesis produces strong 13C-enrichment in stromatolites of Lagoa Salgada, Brazil: A modern analogue for Palaeo-/Neoproterozoic stromatolites?

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Holocene stromatolites characterized by unusually positive inorganic carbon isotope values (δ13C up to +16 ‰ relative to the Vienna Peedee Belemnite Standard; VPDB) are present in Lagoa Salgada, a seasonally brackish to hypersaline lagoon near Rio de Janeiro (Brazil). Such positive values cannot be explained by phototrophic fixation of CO_2 alone. Instead, strong carbon isotope fractionation is commonly observed during methanogenesis, where isotopically light C is preferentially incorporated into methane and the residual inorganic carbon is 13C-enriched. We suggest that methanogenesis was the dominating process of organic carbon mineralization during the growth of the stromatolites. Indeed, the presence of dissolved methane in porewater (up to 5 mM) and the archaeal membrane lipid archaeol showing relatively high δ13C values (–15 to 0 ‰ VPDB) indicates that methanogenic archaea are present and active in the modern lagoon sediment. Moreover, 13C-depleted hopanoids diplopterol and 3-methylated bishomohopanoic acid (both –40‰ VPDB) are preserved in lagoon sediments and are most likely derived from aerobic methanotrophic bacteria thriving in the methane-enriched water column. Loss of isotopically light methane through the water column to the atmosphere would explain the residual 13C-enriched pool of dissolved inorganic carbon from where the carbonate constituting the stromatolites precipitated. The predominance of methanogenic archaea in the lagoon is most likely a result of sulphate limitation suppressing the activity of sulphate-reducing bacteria under brackish conditions in a seasonally humid tropical and arid environment. Also in the modern sediments, sulphate reduction activity is very low. Under dominating methanogenic conditions and in absence of an efficient carbonate-inducing metabolic process, we propose that stromatolite formation in Lagoa Salgada was abiotically induced while the 13C-enriched organic and inorganic carbon pools are due to methanogenesis. Radiogenic Sr-isotope compositions support a mixed water scenario with continental ground or surface water as additional source of alkalinity. Evaporative concentration during dry periods increased the carbonate saturation state in the brine inducing spontaneous or perhaps microbially catalysed precipitation of carbonate. Unusually 13C-enriched stomatolitic deposits also appear in the geological record during prolonged periods in the Palaeo- and Neoproterozoic. Thus, Lagoa Salgada represents a possible modern analogue to conditions that may have been widespread in the Proterozoic, at times when low sulphate concentrations in seawater allowed methanogens to prevail over sulphate-reducing bacteria.