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Constraints on the sources of branched tetraether membrane lipids in distal marine sediments

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ABSTRACT

Branched glycerol dialkyl glycerol tetraethers (brGDGTs) are membrane lipids produced by soil bacteria and occur in near coastal marine sediments as a result of soil organic matter input. Their abundance relative to marine-derived crenarchaeol, quantified in the BIT index, generally decreases offshore. However, in distal marine sediments, low relative amounts of brGDGTs can often still be observed. Sedimentary in situ production as well as dust input have been suggested as potential, though as yet not well constrained, sources. In this study brGDGT distributions in dust were examined and compared with those in distal marine sediments. Dust was sampled along the equatorial West African coast and brGDGTs were detected in most of the samples, albeit in low abundance. Their degree of methylation and cyclisation, expressed in the MBT′ (methylation index of branched tetraethers) and DC (degree of cyclisation) indices, respectively, were comparable with those for African soils, their presumed source. Comparison of DC index values for brGDGTS in global soils, Congo deep-sea river fan sediments and dust with those of distal marine sediments clearly showed, however, that distal marine sediments had significantly higher values. This distinctive distribution is suggestive of sedimentary in situ production as a source of brGDGTs in marine sediments, rather than dust input. The presence of in situ produced brGDGTs in marine sediments means that caution should be exercised when applying the MBT′–CBT palaeothermometer to sediments with low BIT index values, i.e. < 0.1, based on our dataset.

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1. Introduction

Marine sediments provide a unique archive for studying past environment and climate. Depending on the proximity of the study location to land, a varying proportion of the organic matter (OM) input to marine sediments may be land derived. This terrigenous OM can be delivered to marine sediments via several modes of transport, i.e. eolian dust input, suspended in river water, or by means of gravity transport over the sea bed such as through deep sea canyons or in turbidites. Several proxies have been developed that try to distinguish between marine and terrigenous derived OM. They can be based either on bulk properties like the C/N ratio and the stable carbon isotopic ($\delta^{13}C$) composition of OM (Hedges et al., 1997 and references therein), or on molecular composition, e.g. the (relative) abundance of lignin phenols (e.g. Goñi et al., 1997) or other land specific biomarkers like long chain, plant wax derived *n*-alkanes (Eglinton et al., 1962) or taraxerol (Killops and Frewin, 1994; Versteegh et al., 2004).

More recently, branched glycerol dialkyl glycerol tetraether (brGDGT) membrane lipids (Fig. 1) have been used as terrigenous biomarkers in marine sediments as they derive from soil bacteria, likely belonging to the Acidobacteria phylum (Sinninghe Damsté et al., 2000, 2011; Weijers et al., 2006b, 2009a) and their abundance in marine sediments quickly decreases with increasing distance from the coast (Hopmans et al., 2004; Herfort et al., 2006; Kim et al., 2006). Hopmans et al. (2004) proposed the branched vs. isoprenoid tetraether (BIT) index for quantifying the relative abundance of these brGDGTs in marine sediments by normalising them to crenarchaeol, an isoprenoid GDGT membrane lipid derived from ubiquitous pelagic Thaumarchaeota (Sinninghe Damsté et al., 2002). It was suggested that the index could be used as a proxy for tracing terrigenous OM input to marine sediments (Hopmans et al., 2004). However, subsequent studies demonstrated that the index actually traces soil rather than terrigenous OM input relative to marine OM input in near coastal marine sediments, as brGDGTs are present in soil and not in vegetation (Huguet et al., 2007;

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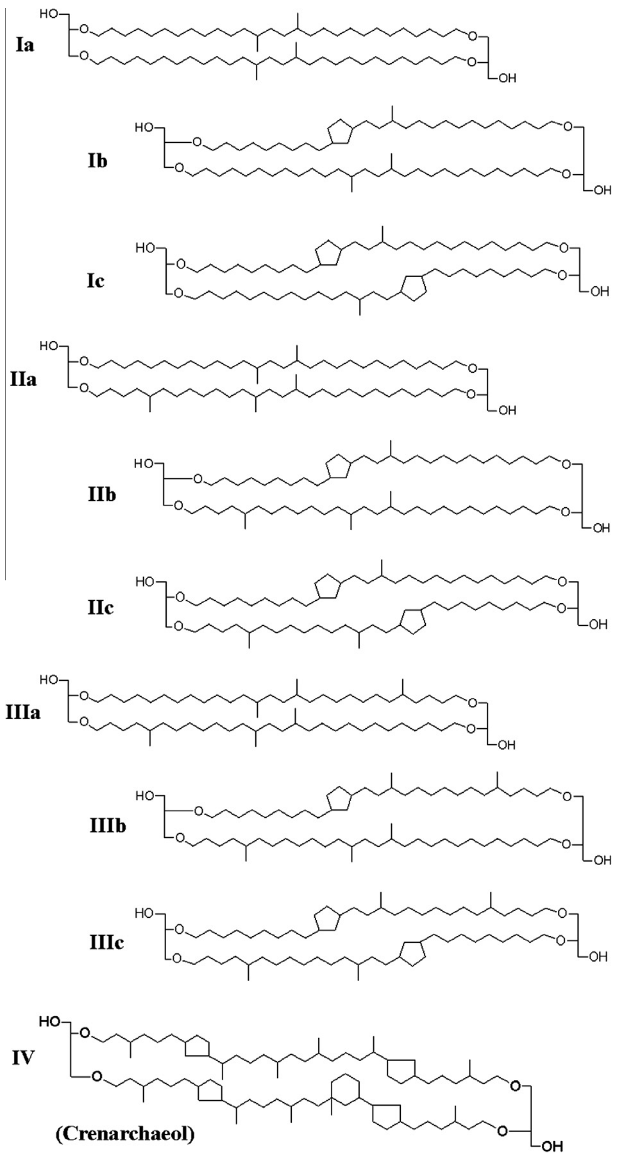

Fig. 1. Glycerol dialkyl glycerol tetraethers (GDGTs) referred to in the text.

Walsh et al., 2008; Kim et al., 2009; Weijers et al., 2009b; Smith et al., 2012).

Besides information on soil OM input, brGDGT distributions also provide information on climate conditions on land. BrGDGTs vary in the number of methyls at C-5 and C-5′ (recently it has been shown that methylation at C-6 and C-6′ also occurs; De Jonge et al., 2013) and contain one or two cyclopentane moieties (Weijers et al., 2006a). In a set of soils from across the globe, it was found that the degree of cyclisation, expressed in the cyclisation ratio of branched tetraethers (CBT), shows a strong relationship with soil pH, and the degree of branching, expressed in the methylation index of branched tetraethers (MBT), shows a strong relationship with both soil pH and annual mean air temperature (MAT; Weijers et al., 2007b). Upon transport of brGDGTs to the marine environment and deposition in the marine sedimentary archive, their distribution could hence be used to reconstruct past soil pH and past annual MAT using these parameters in the so-called MBT–CBT proxy (e.g. Weijers et al., 2007a). Recently the MBT index has been slightly modified (referred to as MBT′) by excluding two brGDGTs from analysis that usually occur in low abundance (i.e. < 1% of total brGDGTs) in soils (Peterse et al., 2012). The same study also provided a new calibration for the MBT–CBT annual MAT proxy using MBT′ and based on a larger number of soils. In addition to the CBT ratio, Sinninghe Damsté et al. (2009) introduced the degree of cyclisation (DC) index in order to define the degree of cyclisation of brGDGTs in a similar way as the degree of methylation in the MBT index, enabling a more direct comparison between the two.

Application of the BIT index and the MBT′–CBT proxy to marine sediments works under the assumption that crenarchaeol is produced in the marine realm and brGDGTs are derived from land. It has, however, been shown that Thaumarchaeota also thrive in soils, peat, lakes and river water and hence that crenarchaeol is also produced in these environments (e.g. Powers et al., 2004; Weijers et al., 2004, 2006b; Leininger et al., 2006; Bannert et al., 2011; Zell et al., 2013). The amount of crenarchaeol in soil and peat is, however, generally low relative to brGDGTs, causing soils and peat to be, on average, still characterised by a high BIT index value, though rarely reaching a value of 1 (0.90 on average; Schouten et al., 2013a and references therein). A similar situation is observed in marine settings: although the BIT index in distal marine settings is often low, it seldom reaches a value of 0 (avg. 0.04; Schouten et al., 2013a), as in most distal marine sediments small amounts of brGDGTs remain detectable. The source of these brGDGTs is uncertain. Long distance dust transport over the oceans might be a plausible mechanism for delivery of brGDGTs to remote ocean settings and brGDGTs have indeed been reported in atmospheric dust sampled off northwest Africa (Fietz et al., 2013). Alternatively, brGDGTs could be produced in situ in marine sediments, as suggested for nearshore marine sediments from a Svalbard fjord (Peterse et al., 2009) and East China Sea sediments (Zhu et al., 2011), based on differences in brGDGT distributions in marine sediments vs. soils on adjacent land. However, the extent to which these findings represent local, near coastal in situ production or general marine in situ production of brGDGTs is unclear.

To investigate whether or not dust or in situ production is responsible for the presence of brGDGTs in distal marine sediments, we analysed the distribution of brGDGTs in atmospheric dust and marine surface waters from the coast of western Africa and in globally distributed open ocean sediments. These data were compared with published brGDGT distributions from western African Congo deep sea river fan sediments and from global soils in order to constrain the potential source of brGDGTs in distal marine sediments and to identify potential implications for the use of GDGT-based proxies.

2. Material and methods

2.1. Samples

Atmospheric dust was sampled along the west coast of equatorial Africa onboard the R/V Meteor cruise M41/1 in 1998. Details of sampling and extraction procedures are provided by Schefuß et al. (2003). For the present study, the polar fraction of each extract from 13 dust filters (Table 1; Fig. 2) was dried, redissolved in *n*-hexane:*iso*-propanol 99:1 (v/v), and filtered through a 0.45 μm mesh PTFE filter prior to analysis of GDGT content.

Suspended particulate matter (SPM) in marine surface waters was sampled by filtration of ca. 100–400 l water, provided by the ship's seawater inlet (ca. 5 m water depth), through a 0.7 μm GFF filter onboard R/V Meteor during cruise M56 in December 2002 along the west coast of equatorial Africa (Spiess and Cruise Participants, 2008). The eight sampling locations comprised a transect along the equatorial African coast and included the lower salinity (down to 28.0‰) Congo River outflow plume (Table 2; Fig. 2). Filters were freeze dried, cut into small pieces and extracted

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