



Re-evaluating modern and Palaeogene GDGT distributions: Implications for SST reconstructions



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ARTICLE INFO

Article history:

Received 3 November 2012

Accepted 20 June 2013

Available online 3 July 2013

Keywords:

TEX₈₆

Glycerol dialkyl glycerol tetraethers

Palaeogene

Sea surface temperatures

Organic geochemistry

Proxy development

Palaeoceanography

Palaeoclimate

ABSTRACT

In this paper, we review the TEX₈₆ palaeothermometer for sea surface temperature (SST) and evaluate its application to the Palaeogene, with a focus on the principal ecological, physical or chemical processes that can bias glycerol dialkyl glycerol tetraether (GDGT) distributions. Recent investigations of Palaeogene sediments have revealed temperature offsets between two different GDGT-based approaches, TEX₈₆^H and TEX₈₆^L, with the former agreeing with SST estimates derived from inorganic proxies (Hollis et al., 2012). These are surprising observations because the two GDGT approaches, although based on two distinct groups of compounds, apparently agree at SSTs > 15 °C in modern oceans. Here we reassess the relationship between raw GDGT distributions and the ratios used to construct TEX₈₆^H and TEX₈₆^L in both the modern core-top dataset and a new compilation of Palaeogene data. We show that the offset between TEX₈₆^H and TEX₈₆^L ($\Delta H-L$) is a function of the GDGT-2/GDGT-3 ratio ([2]/[3] ratio), and that this can be used to separate low- and high-latitude GDGT distributions in the modern core-top dataset: a range of [2]/[3] ratios and $\Delta H-L$ values occur in polar regions, whereas [2]/[3] ratios are high and $\Delta H-L$ values are small at temperatures > 15 °C. However, in the Palaeogene dataset, we observe a wide range of [2]/[3] ratios, even for SST estimates above 15 °C. Crucially, we find that water depth is a better discriminator of $\Delta H-L$ values and [2]/[3] ratios than SST in the combined modern and Palaeogene dataset: $\Delta H-L$ values are low (<3.0 °C) and [2]/[3] ratios are high (>5.0) where water depth is >1000 m.

Modern water column studies show that the [2]/[3] ratios in suspended particulate matter (SPM) increase with depth, suggesting that high [2]/[3] ratios reflect a contribution from Archaea living in the deeper water column. This suggests that export dynamics influence GDGT-derived SST estimates. We argue for new approaches to SST reconstruction: 1) continued use of core-top calibrations, in which export dynamics have been implicitly incorporated into the current core-top calibration datasets, but with the influence of water depth taken into account; and 2) use of SPM or mesocosm-based calibrations, with water depth and palaeo-export dynamics independently assessed.

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1. Introduction

Thaumarchaeota are among the most widely distributed Archaea on Earth, being apparently ubiquitous in soils and lacustrine settings as well as having a large depth range in both the photic and aphotic zones of the water column (Karner et al., 2001; Herndl et al., 2005; Agogué et al., 2008). Comparative genomics (Brochier-Armanet et al., 2008; Spang et al., 2010; Pester et al., 2011), biomolecular characterisation (Gupta and Shami, 2011) and cell cycle investigations (Pelve et al., 2011) have recently revealed that mesophilic marine ‘group I’ Archaea form a separate and deep-branching phylum within the Archaea, resulting in the reclassification of these organisms from Crenarchaeota (DeLong, 1992; Fuhrman et al., 1992) to Thaumarchaeota (Brochier-Armanet et al., 2008). Aside from ubiquity, the Thaumarchaeota appear to be important components in the nitrogen cycle, serving as ammonia oxidisers in both marine (Francis et al., 2005; Könneke et al., 2005; Wuchter et al., 2006b; Mincer et al., 2007; Agogué et al., 2008; Bemen et al., 2008; de la Torre et al., 2008) and terrestrial settings (Treich et al., 2005; Leininger et al., 2006; Zhang et al., 2010; Lehtovirta-Morley et al., 2011; Pratscher et al., 2011; J.G. Kim et al., 2012).

The lipids of Thaumarchaeota are well preserved in marine and lacustrine sediments and are increasingly used in palaeoclimate investigations. Archaeal membranes are comprised of ether lipids, including dialkyl glycerol diethers (DAGEs) and glycerol dialkyl glycerol tetraethers (GDGTs; Fig. 1A), the structures of which were first characterised over 30 years ago (De Rosa et al., 1977, 1980; Gliozzi et al., 1982). GDGTs, or their degradation products, have been found in a diverse range of settings, including virtually all marine environments (Hoefs et al., 1997; DeLong et al., 1998; King et al., 1998; Schouten et al., 1998), swamps and lakes (Pauly and van Fleet, 1986; Pancost et al., 2000), and hypersaline settings (Teixidor et al., 1993). In the former setting, it is now evident that pelagic Thaumarchaeota are the predominant source of GDGTs, most notably crenarchaeol, a unique GDGT bearing four cyclopentyl moieties and, unusually, a cyclohexyl moiety (Sinninghe Damsté et al., 2002).

The distributions of different thaumarchaeotal GDGTs – primarily with respect to the number of cyclopentyl moieties – are related to the organism’s growth temperature (Wuchter et al., 2004), providing the basis for their application in the reconstruction of ancient sea surface temperatures (SSTs). Such GDGT temperature proxies are widely used in time intervals or sites where other proxies are absent or compromised by diagenesis (e.g. Schouten et al., 2003b; Bijl et al., 2009; Hollis et al., 2009; Liu et al., 2009; Littler et al., 2011; Hollis et al., 2012). A recent review summarised the progress made in the last decade in the analysis, occurrence and recognition of sources of GDGTs, as well as their biosynthesis, application as lipid biomarkers and the development of proxies based on their distribution (Schouten et al., 2013). The work presented here complements that review. We briefly repeat the tenets of the calibration, as well as important caveats. We then critically evaluate the GDGT palaeothermometer by interrogating the most recent modern core-top calibration dataset in tandem with a compilation of all published and two unpublished (late Palaeocene, Bass River, NJ; and early Palaeocene, Waipara, NZ) Palaeogene datasets. Our analysis, coupled with recent ecological insights, leads us to reassess the controls on GDGT distribution and offer solutions to the problematic offsets between GDGT calibrations when applied to ancient datasets.

2. Development of the TEX₈₆ palaeotemperature proxies: indices and calibrations

Archaeal GDGTs can contain 0 to 8 cyclopentyl moieties incorporated into the two biphytanyl chains (Fig. 1), but GDGTs of Thaumarchaeota, as well as those occurring in mesophilic settings, contain only 0 to 4 of such groups (Schouten et al., 2013, and references therein). The degree of cyclisation for a given suite of GDGTs can be described by the weighted average number of cyclopentyl moieties (e.g. Uda et al., 2001; Schouten et al., 2002), and this is generally governed by the growth temperature of the archaeal communities producing the compounds (e.g. Shimada et al., 2002; Wuchter et al., 2004; Schouten et al., 2007b; A. Pearson et al., 2008). This phenomenon was initially identified for hyperthermophiles (De Rosa et al., 1980; Gliozzi et al., 1983; De Rosa and Gambacorta, 1988; Uda et al., 2001), but mesocosm studies have demonstrated that the mesophilic marine Thaumarchaeota exhibit similar behaviour (Wuchter et al., 2004; Schouten et al., 2007c). These observations are interpreted to reflect homeoviscous adaptation of the archaeal membrane (De Rosa et al., 1994; Gabriel and Chong, 2000): a higher degree of membrane lipid cyclisation creates a higher thermal transition point for the membrane, consistent with adaptation to warmer temperatures (Gliozzi et al., 1983).

Although there has been some mesocosm work (e.g. Wuchter et al., 2004; Schouten et al., 2007c) and analysis of suspended particulate matter (Wuchter et al., 2005; Lee et al., 2008; Schouten et al., 2012, 2013), current GDGT-based palaeothermometers are based on correlations between satellite-derived mean annual SST measurements and GDGT distributions in core-top sediments. The first correlation was determined by Schouten et al. (2002), who observed that the relative proportions of GDGT-2, GDGT-3 (where n in GDGT- n denotes the number of cyclopentyl moieties present) and crenarchaeol isomer (cren’) positively correlate with SST, whereas GDGT-1 and GDGT-0 negatively correlate with SST. Crenarchaeol exhibits no strong correlation. By extension, SST exhibits a strong correlation to the weighted average number of cyclopentyl moieties within GDGT biphytanyl chains (e.g. Shimada et al., 2002; Schouten et al., 2007b; A. Pearson et al., 2008; Pitcher et al., 2009), which we refer to as the GDGT ring average (Ring_{Av}):

$$\text{Ring}_{\text{Av}} = \left(\frac{(1 \times [\text{GDGT-1}]) + (2 \times [\text{GDGT-2}]) + (3 \times [\text{GDGT-3}]) + (4 \times [\text{cren'}])}{[\text{GDGT-1}] + [\text{GDGT-2}] + [\text{GDGT-3}] + [\text{cren'}]} \right). \quad (1)$$

GDGT-0 and crenarchaeol are typically excluded from the calculation of GDGT weighted averages. GDGT-0 in core-top sediments does exhibit an inverse relationship with SST (Schouten et al., 2002; Kim et al., 2008, 2010), but it is excluded from the calculation because it occurs in a range of other archaea, including methanogenic archaea (Koga et al., 1993; Schouten et al., 2000, 2002; Pancost et al., 2008; Blaga et al., 2009), that are widespread in marine sediments. Crenarchaeol is typically omitted as it is much more abundant than the other GDGTs and does not exhibit a strong relationship with SST (Schouten et al., 2002; Kim et al., 2010). Ultimately, Schouten et al. (2002) determined that the specific ratio of GDGTs that exhibited the highest correlation with SST in their dataset, termed TEX₈₆

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