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Geochimica et Cosmochimica Acta 189 (2016) 24–36

Geochimica et Cosmochimica Acta

www.elsevier.com/locate/gca

Evidence of moisture control on the methylation of branched glycerol dialkyl glycerol tetraethers in semi-arid and arid soils

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Received 20 December 2015; accepted in revised form 1 June 2016; available online 6 June 2016

Abstract

The distribution of bacterial branched glycerol dialkyl glycerol tetraethers (brGDGTs) is influenced by growth temperature and pH. This results in the widespread application of the brGDGT-based MBT(′)/CBT proxy (MBT – methylation of branched tetraethers, CBT – cyclization of branched tetraethers) in terrestrial paleo-environmental reconstructions. Recently, it was shown that the amount of precipitation could also have an impact on CBT, as well as the abundance of brGDGTs relative to that of archaeal isoprenoidal (iso)GDGTs ($R_{i/b}$) and the absolute abundance of brGDGTs, potentially complicating the use of MBT/CBT as paleothermometer. However, the full influence of hydrology, and in particular soil water content (SWC), on GDGT distributions remains unclear. Here we investigated variations in the GDGT distribution across a SWC gradient (0–61%) around Qinghai Lake in the Tibetan Plateau, an arid to semiarid region in China. Our results demonstrate that SWC affects the brGDGT distribution. In particular, we show that SWC has a clear impact on the degree of methylation of C6-methylated brGDGTs, whereas C5-methylated brGDGTs are more impacted by temperature. This results in a combined SWC and temperature control on MBT′. In this context we propose a diagnostic parameter, the IR_{6ME} (relative abundance of C6-methylated GDGTs) index, to evaluate the applicability of brGDGT-based paleotemperature reconstructions. Using the global dataset, expanded with our own data, MBT′ has a significant correlation with mean annual air temperature when $IR_{6ME} < 0.5$, allowing for the use of MBT′/CBT as temperature proxy. However, MBT′ has a significant correlation with mean annual precipitation (i.e., a substantial reflection of SWC impact) when $IR_{6ME} > 0.5$, implying that MBT′ may respond to hydrological change in these regions and can be used as a proxy for MAP.

Keywords: Soil water content; MBT; C6-methylated brGDGTs; Precipitation; Semi-arid and arid region

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1. INTRODUCTION

Lipid biomarkers, including bacterial branched glycerol dialkyl glycerol tetraethers (brGDGTs) and archaeal isoprenoidal (iso)GDGTs, are widely used to reconstruct past environmental conditions in a range of environments (Schouten et al., 2002, 2013; Weijers et al., 2007; Peterse

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http://dx.doi.org/10.1016/j.gca.2016.06.004
0016-7037/

et al., 2012; De Jonge et al., 2014a). Ultimately the reliability of paleo-reconstructions depends on how well the environmental factors controlling these proxies are known in modern settings. Due to the large heterogeneity of terrestrial environments, many proxies, such as the carbon isotopes of organic matter and leaf wax lipids (Zhang et al., 2006; Bendle et al., 2007; Ning et al., 2008; Rao et al., 2013), pollen distributions (Wu et al., 2007) and phytolith distributions (Lu et al., 2006, 2007), appear to be controlled by a range of environmental factors, complicating their interpretation (e.g. ambiguity of temperature vs precipitation control). Consequently, it is necessary to develop new proxies or refine existing ones.

BrGDGTs are presumed to be the membrane-spanning lipids of as-yet unknown bacteria that likely favor anaerobic settings (Weijers et al., 2004, 2009, 2010). The occurrence of brGDGTs-Ia (Fig. 1) in some culture isolates of Acidobacteria suggests that these bacteria are likely the producers of at least one of the brGDGTs observed in natural settings (Sinninghe Damsté et al., 2011). The brGDGTs comprise three series (I, II and III) according to their degree of methylation (Weijers et al., 2007), and each series includes components with 0–2 cyclopentyl rings (e.g. Ia, Ib and Ic) (Fig. 1). In global soil datasets, the MBT and modified MBT′ proxy, representing two different expressions of the degrees of brGDGT methylation, is empirically related to mean annual air temperature (MAT) and soil pH, whereas the degree of cyclization of brGDGTs, i.e. CBT, correlates only with pH (Weijers et al., 2007; Peterse et al., 2012). A combination of MBT (′) and CBT, i.e., the MBT(′)/CBT proxy, therefore, has been used to reconstruct continental temperature in loess-paleosols (Peterse et al., 2011) and sediments as far back as the Paleocene (Pancost et al., 2013; Kemp et al., 2014). Additional evidence, primarily from altitudinal transects (e.g. Sinninghe Damsté et al., 2008; Ernst et al., 2013), further supports the close relationship between MBT(′)/CBT and MAT.

However, the global soil database is characterized by large scatter in the relationship between MBT′ with either MAT or pH (Peterse et al., 2012), and when applied to modern alkaline soils from semi-arid and arid regions, the global MBT(′)/CBT calibration leads to estimates significantly lower than instrumental temperatures (Peterse et al., 2012; Yang et al., 2014). These observations imply that other environmental factors impact MBT(′)/CBT and consequently paleo-temperature reconstructions. For example, Dirghangi et al. (2013) suggested that mean annual precipitation (MAP) affects both brGDGT composition and the BIT-index, the ratio of brGDGTs to the dominant isoGDGTS (Hopmans et al., 2004; defined below), in soils from the USA. Similarly, a significant (negative) correlation between CBT and MAP (and SWC) was found in semi-arid and arid soils from China (Wang et al., 2014). Menges et al. (2014) found that MBT in soils from the Iberian Peninsula had a moderate correlation with the aridity index (AI = mean annual precipitation (MAP)/mean annual potential evapotranspiration), but the effect of MAP on MBT was blurred by its co-variation with pH. MAP has also been proposed to impact the distribution of brGDGTs in cold and wet and in warm and dry regions (Peterse et al., 2012), although excluding these soils did not improve the relationship with MAT in the global dataset (Weijers et al., 2007; Peterse et al., 2012), implying that other factors may exist. One such factor could be soil moisture, which at a given site can be decoupled from

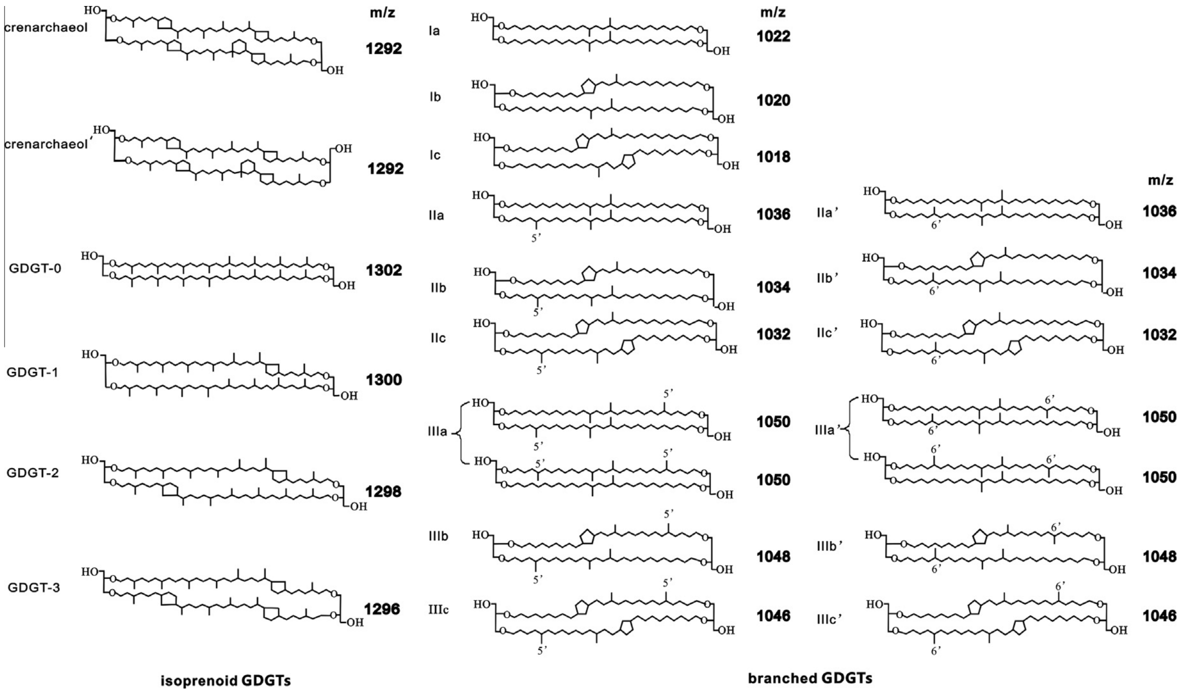

Fig. 1. The structures of archaeal isoprenoidal (iso-) and bacterial branched (br-) glycerol dialkyl glycerol tetraethers (GDGTs).