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Sources and distributions of branched tetraether lipids and crenarchaeol along the Portuguese continental margin: Implications for the BIT index

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ABSTRACT

The branched vs. isoprenoid tetraethers (BIT) index, which is based on the relative abundance of non-isoprenoidal, so-called branched glycerol dialkyl glycerol tetraethers (brGDGTs) versus a structurally related isoprenoid GDGT “crenarchaeol”, has been used to trace soil organic carbon (OC) from the continent to the ocean. However, it has been found in some locations that the BIT index can be primarily influenced by crenarchaeol concentrations and brGDGT production in fresh water rather than by soil-derived brGDGT concentrations. This may hamper the application of this proxy as an indicator for the input of soil OC. In order to constrain the applicability of the BIT index along the southern Portuguese continental margin, we examined the source of brGDGTs and crenarchaeol, by investigating their concentration and distribution as well as variations in the BIT index in marine surface sediments from five transects (Douro, Mondego, Estremadura, Tagus, and Sado) and in marine suspended particulate matter (SPM) from the Douro and Tagus transects. Higher BIT values and brGDGT concentrations (normalized to OC content) were found close to the river mouths and coast than in deep offshore sites. This clearly indicated the continental input of brGDGTs and revealed that, at least in this setting, the BIT index was primarily influenced by the delivery of brGDGTs from the rivers. BrGDGT concentrations and distributions in sediments and SPM close to the rivers were similar to those of SPM in the Tagus River. This indicates that degradation processes in the estuaries had no significant effect on the riverine brGDGTs. Therefore, brGDGTs should be a good indicator for the recalcitrant OC fraction transported from the continent to the ocean. Our results also indicated that there are multiple sources of brGDGTs in the marine environment, i.e. the water column and the sediment, which complicates the use of the brGDGT distribution as an indicator for terrestrial vs. marine produced brGDGTs.

1. Introduction

The branched vs. isoprenoid tetraether (BIT) index has been used to trace the transport of soil OC to the marine environment. It is based on a ratio of membrane lipids of heterotrophic bacteria that are common in soil and membrane lipids of Thaumarchaota, which are common in the marine environment. The lipids are branched glycerol dialkyl glycerol tetraethers (brGDGTs) and crenarchaeol (Fig. 1), respectively (Hopmans et al., 2004). Initially, it was thought that the BIT index equaled 0.0 in the marine environment and 1.0 in the terrestrial environment (Hopmans et al., 2004).

However, this is often not the case, it was found that crenarchaeol is not only produced in the marine environment but also occurs in soil and fresh water (e.g. Weijers et al., 2006). Especially in alkaline soils crenarchaeol concentrations are high (e.g. Kim et al., 2010; Weijers et al., 2007; Xie et al., 2012; Yang et al., 2011). In addition it is influenced by variations of the crenarchaeol concentration in the marine environment (e.g. Castañeda et al., 2010; Fietz et al., 2011; Schmidt et al., 2010; Smith et al., 2012). Similarly, brGDGTs are also not exclusively produced in soil but also in lakes (Bechtel et al., 2010; Buckles et al., 2014; Loomis et al., 2011; Sinninghe Damsté et al., 2009; Tierney and Russell, 2009; Tierney et al., 2010, 2012; Zink et al., 2010) and rivers (De Jonge et al., 2014; Kim et al., 2012; Yang et al., 2013; Zell et al., 2013a,2013b; 2014; Zhang et al., 2012; Zhu et al., 2011). Furthermore, brGDGTs are also produced in the marine environment, but in much lower concentrations than in continental

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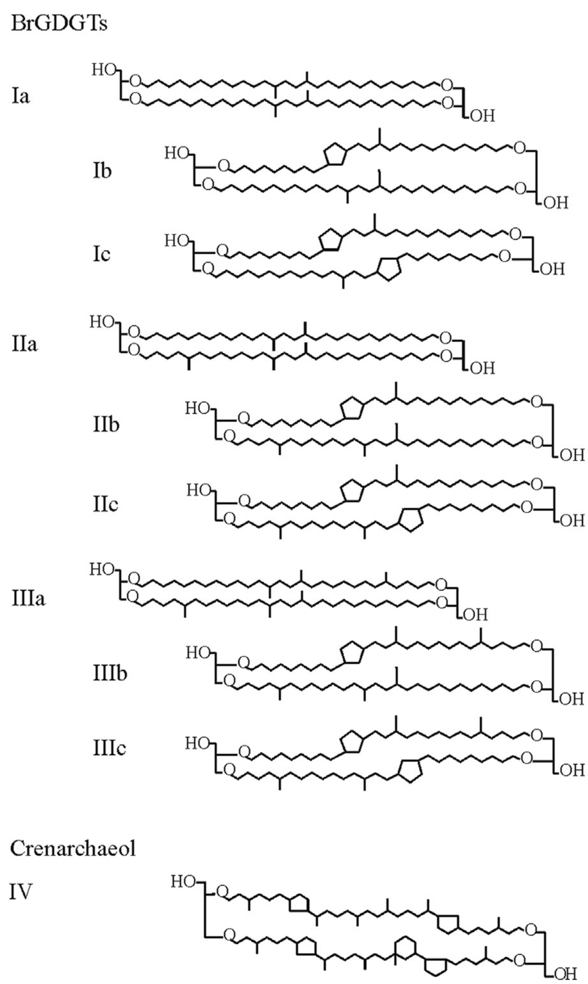

Fig. 1. Chemical structure of brGDGTs (Ia-IIIa) and crenarchaeol (IV).

settings, therefore the marine production does not have a strong influence on the BIT index (e.g. Hu et al., 2012; Peterse et al., 2009; Weijers et al., 2014; Zhu et al., 2011). Overall, this means that the BIT index is influenced by various factors and does not only indicate organic matter coming from soils, but rather a more general continental signal.

A better understanding of the source of crenarchaeol and brGDGTs could help to indicate how the BIT index can be used to trace the input of the continental OC. Therefore, we analyzed their concentration and distribution in five transects along the southern Portuguese continental margin (Fig. 2). The transects I and IV start close to the river mouth of the Douro River and the Tagus River, respectively (Data of transect IV is already published in Zell et al., 2014a). Transects II and V start close to the mouth of substantially smaller rivers, the Mondego River and the Sado River, respectively, while transect III does not receive any direct input from a river. Transect III is, therefore, thought to be less influenced by the terrestrial environment. Organic matter transported by rivers is often strongly degraded in the estuary (e.g. Zhu et al., 2013). This degradation processes might also influence the amounts of brGDGTs that are delivered by the rivers. The Douro and Tagus river systems have different types of estuaries. The Tagus River has a large estuary (the largest in Western Europe), which retains the majority of the terrestrial organic matter of the river (Jouanneau et al., 1998). By contrast, the Douro estuary is smaller and has a higher discharge rate, thus the riverine organic matter has a relatively short residence time in this estuary (a few days) (Abril et al., 2002).

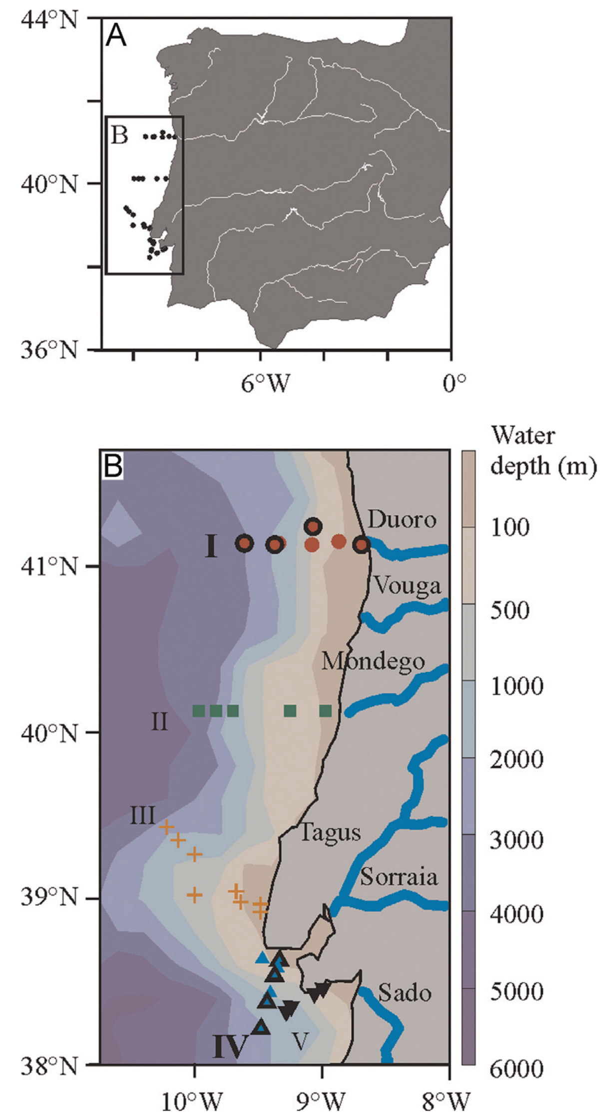

Fig. 2. (A) Overview of the study area with the sampling sites of surface sediments and (B) detailed sampling locations of surface sediments along the five (I: Douro, II: Mondego, III: Estremadura, IV: Tagus, and V: Sado) transects. Symbols with a black outline indicate the sites at which SPM samples were also taken.

2. Study area

The Portuguese continental shelf ranges from 20 to 34 km in width. The shelf break varies between 130 and 150 m water depth. Several canyons (e.g. Nazaré, Cascais, and Setúbal–Lisbon canyons) intersect the shelf (e.g. Vanney and Mougenot, 1981). In summer (May–September), the Azores high pressure system is driven closer to the coast, which together with the associated northerly winds makes the colder, less salty and nutrient enriched subsurface water (from 60 to 120 m depth) rise to the surface along the Iberian margin (Fiúza, 1983). This upwelling leads to increased productivity in summer along a ca. 50 km wide zone. In winter, the Azores high moves south, which results in southerly winds and downwelling conditions. The downwelling conditions in winter lead to deposition of sediment on the shelf (Frouin et al., 1990). Winter storms can remobilize sediment and transport it northward by bottom currents (Dias et al., 2002; Vitorino et al., 2002) and eventually deposit it in the mid-shelf mudbelt (between 50 and 130 m water depths) (Vitorino et al., 2002). Hence, the upper 10–15 cm of sediment on the inner shelf (above 100 m water depth) represent a mixing layer which decreases in mixing