

Global scale production of BrGDGTs by benthic marine bacteria: Implication for developing ocean bottom environmental proxies

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Abstract :

The in-situ production of branched glycerol dialkyl glycerol tetraethers (brGDGTs) in marine environments has been generally accepted. However, whether it takes place in the water column or the sediments remains elusive. Here, we compiled a dataset composed of globally distributed marine sediments from continental shelves to hadal trenches to address this important knowledge gap. Firstly, a cutoff of $\Sigma\text{IIIa}/\Sigma\text{IIa} \geq 0.92$ was applied to distinguish the marine vs. terrestrial signals of brGDGT production. For marine derived brGDGTs, the cyclization of branched tetraethers (CBT) index, related to ambient pH, displays a large variability, suggesting that marine derived brGDGTs in sediments are mainly produced within sediments where the pH of sediment porewater is highly variable, rather than in the water column where pH is relatively constant. This is also supported by good correlations between the bottom water temperature and the methylation of branched tetraethers (MBT') index, between the water depth and the $\Sigma\text{IIIa}/\Sigma\text{IIa}$ index, as well as between the brGDGT-based proxies and sediment redox conditions during glacial/interglacial cycles. We thus propose that the benthic production of brGDGTs in marine sediments is ubiquitous on a global scale, and is largely affected by the ambient temperature and pH. Our findings support that brGDGT-based proxies have potential of recording environmental conditions of the ocean bottom.

Highlights

► A dataset of brGDGTs in global marine sediments was compiled. ► Benthic production of brGDGTs in marine sediments is ubiquitous on a global scale. ► Marine derived brGDGTs in sediments reflect ocean bottom environmental conditions.

Keywords : Branched GDGTs, in-situ production, marine sediment, global scale, paleoclimate reconstruction

1 Introduction

Branched glycerol dialkyl glycerol tetraethers (brGDGTs), consisting of two branched alkyl chains with a differing number of methyl groups (4–6) and cyclopentane moieties (0–2), are widely distributed in marine and terrestrial settings (Sinninghe Damsté et al., 2000; Weijers et al., 2006). They were thought to be produced in soil, peat and hot spring by bacteria including, but not necessarily limited to, Acidobacteria (Sinninghe Damsté et al., 2011; Zhang et al., 2013). In the past decade, several indicators on the basis of brGDGT compositions, such as branched/isoprenoid tetraethers (BIT), methylation and cyclization of branched tetraethers (MBT and CBT), have been developed and widely used for reconstruction of organic carbon source, paleo-temperature and soil pH (Schouten et al., 2013). Recently, improvements in chromatography methodologies enabled the separation of 5-, 6- and 7-methyl brGDGTs (De Jonge et al., 2013; Ding et al., 2016), which led to the redefinition and recalibration of brGDGT-based proxies (e.g., De Jonge et al., 2014; Xiao et al., 2015).

The application of brGDGT-based proxies in marine settings is based on a presumption that brGDGTs are only derived from land. However, mounting evidence suggests that brGDGTs can also be produced by marine organisms in estuary and shelf (De Jonge et al., 2015; Sinninghe Damsté, 2016; Xiao et al., 2016; Zell et al., 2014), distal marine (Weijers et al., 2014) and hadal trench sediments (Xiao et al., 2020; Xu et al., 2020). Taken Svalbard as an example, the brGDGT distributions in fjord sediments exhibited a strikingly different pattern from the nearby soils (Peterse et al., 2009). Sinninghe Damsté (2016) proposed that the degree of cyclization of tetramethylated brGDGTs ($\#rings_{tetra}$) can be an indicator for assessment of brGDGT sources with significantly higher values in marine derived brGDGTs than soil derived brGDGTs. By

compiling globally distributed marine sediments and soils, Xiao et al. (2016) developed a new index, as expressed by the abundance ratio of hexamethylated to pentamethylated brGDGT ($\Sigma\text{IIIa}/\Sigma\text{IIa}$), to evaluate the sources of brGDGTs with significantly higher $\Sigma\text{IIIa}/\Sigma\text{IIa}$ values for marine derived brGDGTs.

Although in-situ production of brGDGTs in marine environments has been generally accepted, it remains vague whether they are produced in the water column or the sediments. This source appropriation is important for the application of brGDGT-based proxies in modern and paleo-environments since they may record the signal where they were biosynthesized. Weijers et al. (2014) investigated brGDGTs in distal marine sediments and suggested that marine derived brGDGTs are mainly produced in marine sediments where the pH of porewater is variable. Sinninghe Damsté (2016) suggested that in-situ production of brGDGTs was a widespread phenomenon in shelf sediments that was especially pronounced at water depth of ca. 50–300 m. Marine in-situ production may also take place in the water column but few studies have focused on brGDGT distributions in marine sinking particles (De Jonge et al., 2015; Liu et al., 2014; Yamamoto et al., 2016; Zell et al., 2014). Liu et al. (2014) suggested that brGDGTs are preferentially produced in-situ by planktonic microorganisms favoring anoxic waters. If so, the prevailing of oxygenated waters in the ocean will inhibit the production of brGDGTs, and the benthic production of brGDGTs will become important in marine sediments where oxygen is depleted below sediment-water interface. Nevertheless, our knowledge of relative importance of planktonic vs. benthic bacteria (water column vs. sediment) to the sedimentary brGDGT pool in global ocean is limited, which hampers the application of brGDGT-based proxies in marine environments. Here, we compiled a dataset composed of globally distributed marine sediments from continental shelves, slopes, abyssal plains and hadal trenches, aiming to determine the

source of marine derived brGDGTs preserved in global marine sediments (allochthonous water column and in-situ sediment), and assessing their environmental implications.

2 Data compilation and methods

The information of dataset used in this study is listed in Tab S1. It consists of 749 samples compiled from the northern Atlantic Ocean (n = 29, Peterse et al., 2009), southern Italian shelf (n = 48, Leider et al., 2010), Arctic Ocean (n = 18, Smith et al., 2002), western Pacific warm pool (n = 5, O'Brien et al., 2014), distal marine sediments (n = 38, Veijers et al., 2014), eastern Indian Ocean (n = 36, Chen et al., 2014), Chilean margin (n = 15, Kaiser et al., 2015), East Siberian Arctic Shelf (n = 92, Sparkes et al., 2015), Gulf of Lions (n = 8, Kim et al., 2015), Portuguese margin (n = 15, Warden et al., 2016), Beaulieu River delta (n = 43, Sinninghe Damsté, 2016), western North Pacific (n = 5, Yamamoto et al., 2016), western Bering Sea and northwest Pacific (n = 10, Meyer et al., 2016), Chinese marginal seas (n = 323, Cao et al., 2020; Chen et al., 2018; Dong et al., 2015b; Ge et al., 2013; Liu et al., 2021; Lü et al., 2014; Xiao et al., 2016; Xing et al., 2016; Xing et al., 2014), Brazilian continental margin (n = 46, Ceccopieri et al., 2019), Japan Sea (n = 5, Wu et al., 2020), Mariana Trench (n = 5, Xiao et al., 2020) and Chukchi Sea shelf (n = 10, Gao et al., 2021) (Fig. 1a). For the sediment cores in the dataset, the top five samples were selected. The temperature data used in this study are extracted from WOA18 0.25 degree dataset (Locarnini et al., 2018). The time periods are annual, and time spans denote all data used in WOA18 regardless of year.

The roman numbers in following equations denote the fractional abundance of GDGTs whose structures are depicted in Fig. S1. It should be noted that most of previous studies did not separate 5- and 6-methyl brGDGTs. Therefore, we used the CBT and MBT' to represent the cyclization and methylation of brGDGTs, respectively. The BIT, $\Sigma\text{IIIa}/\Sigma\text{IIa}$, CBT and MBT'

indexes were calculated according to the definitions of Hopmans et al. (2004), Xiao et al. (2016) and De Jonge et al. (2014), respectively. Both parametric (Pearson correlation, r_p) and non-parametric (Spearman correlation, r_s) tests were used in this study to evaluate the relationship between two variables.

$$\text{BIT} = (\text{Ia} + \text{IIa} + \text{IIIa} + \text{IIa}' + \text{IIIa}') / (\text{Ia} + \text{IIa} + \text{IIIa} + \text{IIa}' + \text{IIIa}' + \text{Cren}) \quad (1)$$

$$\Sigma\text{IIIa} / \Sigma\text{IIa} = (\text{IIIa} + \text{IIIa}') / (\text{IIa} + \text{IIa}') \quad (2)$$

$$\text{CBT} = -\log[(\text{Ib} + \text{IIb} + \text{IIb}') / (\text{Ia} + \text{IIa} + \text{IIa}')] \quad (3)$$

$$\text{MBT}' = (\text{Ia} + \text{Ib} + \text{Ic}) / (\text{Ia} + \text{Ib} + \text{Ic} + \text{IIa} + \text{IIa}' + \text{IIb} + \text{IIb}' + \text{IIc} + \text{IIc}' + \text{IIIa}' + \text{IIIa}') \quad (4)$$

3 Results and discussion

3.1 In-situ production of brGDGTs in global marine sediments

Two indicators, namely $\Sigma\text{IIIa} / \Sigma\text{IIa}$ (Xiao et al., 2016) and $\#\text{rings}_{\text{tetra}}$ (Sinninghe Damsté, 2016), have been commonly used to assess the sources of brGDGTs in marine environments. The $\Sigma\text{IIIa} / \Sigma\text{IIa}$ index is generally < 0.92 and ≥ 0.92 in global marine sediments with and without terrestrial influences, respectively. It has been successfully applied in different marine environments, including the continental shelves, bathyal, abyssal and hadal zones (e.g. Menot et al., 2020; Xiao et al., 2020). Based on brGDGT distributions in shelf systems, Sinninghe Damsté (2016) proposed a threshold of $\#\text{rings}_{\text{tetra}} > 0.7$ as a sign of a predominantly marine-sourced brGDGTs, which later has been widely demonstrated in coastal environments (e.g., Crampton-Flood et al., 2019; Huguet et al., 2019). However, distal marine (Weijers et al., 2014) and Mariana Trench sediments (Xiao et al., 2020), where terrestrial input is insignificant, exhibit low $\#\text{rings}_{\text{tetra}}$ values (0.51 ± 0.23 and 0.26 ± 0.04 , respectively). In the Atlantic Iberian margin (Zell et al., 2015), Brazilian continental margin (Ceccopieri et al., 2019), South China Sea (Zhang et al.,

2020) and global dataset in this study (Fig. S2), the $\#rings_{tetra}$ values increase at a water depth of ca. 0-200 m, and then gradually decrease with the increasing water depth. These results suggest that the $\#rings_{tetra}$ index should be applied with caution in the deep ocean (Zhang et al., 2020). Therefore, only the $\Sigma IIIa/\Sigma IIa$ index was used to distinguish terrestrial vs. marine brGDGT sources in our globally distributed marine sediments.

About 310 samples (ca. 40% of the total samples) have the $\Sigma IIIa/\Sigma IIa$ value ≥ 0.92 , suggesting a source of marine production of brGDGTs. Correspondingly, these samples are from significantly ($p < 0.001$) deeper waters (1085 ± 1770 m) and exhibit significantly ($p < 0.001$) lower BIT values (0.07 ± 0.07) compared to the samples receiving mixing marine/terrestrial brGDGTs ($\Sigma IIIa/\Sigma IIa < 0.92$, water depth 188 ± 527 m, BIT 0.18 ± 0.21) (Fig. 1b, c). Moreover, the marine sediments with a water depth > 4000 m ($n = 19$) all have a $\Sigma IIIa/\Sigma IIa$ value ≥ 0.92 and a BIT value < 0.06 . For the samples with pure marine derived brGDGTs ($\Sigma IIIa/\Sigma IIa \geq 0.92$), their CBT and MBT' values (0.06 ± 0.27 and 0.52 ± 0.11 , respectively) are both significantly ($p < 0.001$) lower than these containing mixing marine/terrestrial brGDGTs (0.23 ± 0.24 and 0.51 ± 0.13 , respectively) (Fig. 1d, e). These results are consistent with previous studies (Liu et al., 2021; Xiao et al., 2016), and reveal that marine brGDGT-producing microbes can change the degree of cyclization and methylation to adapt to the ambient conditions, and marine derived brGDGTs are generally biosynthesized at lower temperature and more alkaline pH conditions compared to soil derived brGDGTs.

The comparison of brGDGTs between the suspended particles and sediments provides a strategy to decipher the source location where sedimentary brGDGTs were produced in marine environments. Yamamoto et al. (2016) analyzed depth variation of brGDGT distributions in sinking particles (water depth of ca. 1300–4800 m) and underlying sediments (water depth of ca.

5300 m) in the western North Pacific. The $\Sigma\text{IIIa}/\Sigma\text{IIa}$ values of sinking particles (1.18 to 2.94 with an average of 1.93) and underlying sediments (2.22 to 3.63 with an average of 3.15) indicate that marine derived brGDGTs can be produced both in the water column and the sediments. However, the $\Sigma\text{IIIa}/\Sigma\text{IIa}$ index of sinking particles is significantly ($p < 0.001$) lower than that of underlying sediments (Fig. 2). Similarly, the BIT index of sinking particles (0.0002 to 0.0014 with an average of 0.0007) has one order of magnitude lower than that of underlying sediments (0.003 to 0.022 with an average of 0.011) (Fig. 2). In addition, the CBT index of sinking particles varies within a narrow range (0.43 ± 0.08), which likely reflects the stable pH of the water column, and is significantly ($p < 0.001$) higher than that of underlying sediments (0.21 ± 0.11). These differences in brGDGT-based proxies between the water column and underlying sediments are hardly explained by selective degradation because 1) sinking particles of different water depths have similar $\Sigma\text{IIIa}/\Sigma\text{IIa}$, BIT and CBT values, and 2) brGDGTs IIIa and IIa having similar chemical structures are expected to have similar degradation rates. Thus, the multiple lines of evidence support that lithic rather than planktonic production contributes to the majority of the brGDGT pools in deep sea sediments, and substantially alters the brGDGT' compositions there.

Globally, the CBT values of marine derived brGDGTs are quite variable from -0.33 to 0.81 (0.06 ± 0.27) (Fig. 3a). Assuming that marine brGDGTs-producing organisms have the same response to environmental factors (pH and temperature) as soil brGDGTs-producing bacteria (Liu et al., 2021), the reconstructed pH will change between 6.3 and 8.6 according to the global soil calibration: $\text{pH} = 7.9 - 1.97 \times \text{CBT}$ (De Jonge et al., 2014). Such amplitude is substantially larger than the pH range of seawater in modern ocean that is 7.9–8.4 in surface water and 7.5–7.8 in deep-sea water, although the anthropogenic CO_2 emission may lower seawater pH by up to 0.7

unit beyond the year of 2100 (Andersson et al., 2004). Even if the uncertainty of the CBT-pH proxy was considered (0.8 unit) (Peterse et al., 2012), the variable amplitude of the reconstructed pH is still larger than the actual range of marine water column. This suggests that marine derived brGDGTs preserved in sediments are significantly produced in situ by benthic microorganisms, since the pH of porewater in marine sediments vary significantly due to complex redox conditions related to the physical and chemical characteristics of bottom water, the nature of solid components of sediments and the dissolution of carbonates (Jorabchi et al., 2005).

We chose three cases to further elaborate the source of brGDGTs in marine sediments. In the Brazilian margin (Ceccopieri et al., 2019), Berau delta (Stinninghe Damsté, 2016) and Svalbard fjord (Peterse et al., 2009) sediments, the $\Sigma\text{IIIa}/\Sigma\text{IIa}$ values show increasing trends seawards, indicating enhanced contribution of marine derived brGDGTs (Fig. 4). Meanwhile, the MBT' index, exhibiting a contrary tendency as the $\Sigma\text{IIIa}/\Sigma\text{IIa}$ index, gradually decreases (lower temperature) seawards. However, no trend was observed along the latitude for both $\Sigma\text{IIIa}/\Sigma\text{IIa}$ and the MBT' index. Despite the mixture of terrestrial and marine signals may complicate brGDGT-based proxies in nearshore areas, these sediments with $\Sigma\text{IIIa}/\Sigma\text{IIa} \geq 0.92$ still do not show a clear trend with latitude for the MBT' index. These results suggest that marine derived brGDGTs in sediments may be produced in deep waters or sediments where the ambient temperature is less variable, and they record the in-situ lower water temperature rather than the higher sea surface temperature (SST). Globally, for mixing terrestrial/marine derived brGDGTs ($\Sigma\text{IIIa}/\Sigma\text{IIa} < 0.92$), there is a strong positive correlation between the MBT' and SST ($n = 207$, $r_p = 0.74$, $p < 0.001$) (Fig. 3b). However, for marine derived brGDGTs ($\Sigma\text{IIIa}/\Sigma\text{IIa} \geq 0.92$), such correlation is absent ($n = 162$, $r_p = 0.25$, $p = 0.001$), again suggesting the surface water temperature is not the controlling factor on the composition of marine derived brGDGTs. On the

contrary, a significant correlation was observed between the MBT' and bottom water temperature (BWT, $n = 162$, $r_p = 0.69$, $p < 0.001$) (Fig. 3c), consistent with a previous report by Crampton-Flood et al. (2018) that marine derived brGDGTs reflect in-situ BWT. Our finding supports the possibility that marine derived brGDGTs in sediments are mainly produced by benthic bacteria, and are influenced by the bottom water temperature. It is worth noting that the linear regression of MBT' vs. BWT for marine derived brGDGTs has a similar slope and intercept (0.015 and 0.279, respectively) as that of MBT' vs. mean annual air temperature (MAAT) for the global soil brGDGTs (0.017 and 0.262, respectively) (Fig. S3). This similarity corroborates our hypothesis that the marine brGDGT-producing bacteria have the similar response to temperature as their terrestrial counterparts. Taken together, the significant correlation between MBT' and BWT of marine derived brGDGTs and large amplitude variability of CBT-reconstructed pH suggest that marine derived brGDGTs preserved in globally distributed sediments are predominately biosynthesized by benthic rather than planktonic bacteria.

Globally, the $\Sigma\text{IIIa}/\Sigma\text{IIa}$ index (with the value ≥ 0.92) exerts a significant positive correlation with the water depth ($n = 306$, $r_s = 0.46$, $p < 0.001$) (Fig. 5), indicating that the composition of marine derived brGDGTs is related to the water depth. This can be explained by three reasons. First, with the increasing water depth, the generally decreasing temperature in bottom waters would result in the higher degree of methylation of brGDGTs, and thus higher $\Sigma\text{IIIa}/\Sigma\text{IIa}$ values, like the cases in the Brazilian margin, Berau delta and Svalbard fjord as discussed above. Note that such effect may be minor when the water depth is beyond 1500 m where the bottom temperature is relatively constant (2–4 °C). Second, sediment redox conditions are dependent on oxygen consumption rates. Compared with shallow seas, the deep seas usually have lower sedimentary respiration rates that are associated with decreased levels of CO_2 in pore

water of sediments (Andersson et al., 2004; Jourabchi et al., 2005), leading to higher pH and thereby higher $\Sigma\text{IIIa}/\Sigma\text{IIa}$ values (Xiao et al., 2016). Third, the bathyal, abyssal and hadal zones characterized by high hydrostatic pressure, low temperature and darkness may have evolved unique brGDGT-producing microbes. For example, the unprecedentedly high $\Sigma\text{IIIa}/\Sigma\text{IIa}$ values (7.13 ± 0.98) was observed in the deepest Mariana Trench (Xiao et al., 2020). No matter which explanation is more important, the close relationship between the $\Sigma\text{IIIa}/\Sigma\text{IIa}$ index (with the value ≥ 0.92) and the water depth supports that, on a global scale, marine derived brGDGTs in sediments are mainly derived from in-situ production in the sediments rather than the overlying water column. At current stage, the weighed contributions of brGDGTs from water column and sediment are unable to be quantified, which needs the investigations for intact polar lipids (like IPL-brGDGTs) and microbial community of sinking particles and sediments.

3.2 Implications for brGDGT-based proxies in marine sediments

The benthic production of brGDGTs in global marine sediments complicates the application of brGDGT-based proxies for on earth surface conditions like soil pH and air temperature (Sinninghe Damsté, 2016; Weijers et al., 2014; Zhang et al., 2020). However, if sedimentary brGDGTs are primarily produced by benthic bacteria (particularly from deep seas), MBT', CBT and other brGDGT-based proxies may record the ocean bottom environmental conditions (Crampton-Flood et al., 2018).

A good example is from Dong et al. (2015b) who provided high resolution profiles of brGDGT distributions in a sediment core from the South China Sea (MD05-2896/7, water depth 1658 m), spanning the period of marine isotope stage 6 (MIS 6) to present (the last 175 ka) (Fig. 6). The MBT' index showed an anti-phase pattern as the glacial-interglacial cycle of SST reflected by $U_{37}^{K'}$ and sea level (Fig. 6a, b), with higher methylation degree (lower temperature)

during interglacial periods (MIS 1, 5) and lower methylation degree (higher temperature) during glacial periods (MIS 2-4, 6) (Fig. 6d). This unexpected pattern suggests that the MBT' index in the core MD05-2896/7 does not record air or surface water temperature. Since $\Sigma\text{IIIa}/\Sigma\text{IIa}$ ratio is constantly ≥ 0.92 (Fig. 6f), a contribution of terrestrial brGDGTs can be almost excluded (Xiao et al., 2016). Based on the globally distributed soils, the MBT' index is dependent on temperature and to a lesser extent on pH, whereas the CBT index is solely dependent on pH (Peterse et al., 2012). Our global dataset has suggested that marine bacteria have a similar response as their terrestrial counterparts in producing brGDGTs with changing temperature and pH. Thus, brGDGTs in the core MD05-2896/7 are not mainly derived from surface waters where temperature changes in phase with glacial/interglacial cycles. Furthermore, the CBT index displays a large amplitude variability between 0.17 and 1.04 (0.60 ± 0.18), converting into pH from 6.0 to 8.3 (7.2 ± 0.5). The reconstructed pH values are lower than that of the bottom water in South China Sea, indicating that the majority of brGDGTs in core MD05-2896/7 is not derived from deep water, but is produced by bacteria dwelling in the sediments where the pH of porewater is more variable. Since the in-situ temperature of the sediment core is relatively constant (water depth 1658 m), the variation of the MBT' index is not determined by the temperature but by the pH of porewater in marine sediments. This is confirmed by a strong positive correlation between the MBT' and CBT indexes ($n = 676$, $r_p = 0.93$, $p < 0.001$; Fig. 6d, e, S4).

The intrabasin oceanic processes such as vigorous diapycnal mixing and deep upwelling play an important role in releasing excessive CO_2 from the deep ocean to the sea surface in the southern South China Sea (Dai et al., 2013). Under the low sea level stand during glacial periods (Fig. 5b), the reduced deep vertical mixing and advection weakened intrabasin water renewal and

lowered bottom pH relative to the interglacial periods. Under this condition, the CBT index decreased (higher pH) during interglacial periods and increased (lower pH) during glacial periods, as observed in the core MD05-2896/7. The glacial/interglacial variability in the CBT index is also consistent with that in the Mn/Al ratio (a redox sensitive indicator) (Fig. 6c, e). These results support that brGDGTs in deep sea sediments are potential tools for reflecting environmental conditions of the ocean bottom.

4 Conclusions

The compiling of this global dataset reveals apparent variability in the degree of cyclization of marine derived brGDGTs, and their corresponding pH has a larger amplitude than actual seawater pH in ocean. This suggests that the majority of marine derived brGDGTs is most likely produced in sediments where the pH of pore water is more variable. This is further supported by the significant relationship between the bottom water temperature and the MBT', and between the water depth and the $\Sigma\text{IIIa}/\Sigma\text{IIa}$ of marine derived brGDGTs. Our results show that marine derived brGDGTs are predominately biosynthesized by bacteria dwelling in the sediments, and are influenced by the ambient conditions, such as the porewater pH and bottom temperature (Fig. 7). Thus, marine derived brGDGTs preserved in sediments are valuable tools to study ocean bottom environmental changes.

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Competing interests

The authors declare no competing financial interests.

Supplementary data

Supplementary material

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Figure 1. a) World map showing the location of marine sediments included in the dataset of this study. Samples are color coded based on their $\Sigma\text{IIIa}/\Sigma\text{IIa}$ values. Distribution of parameters including b) water depth, c) BIT, d) MBT' and e) CBT of marine sediments in the dataset.

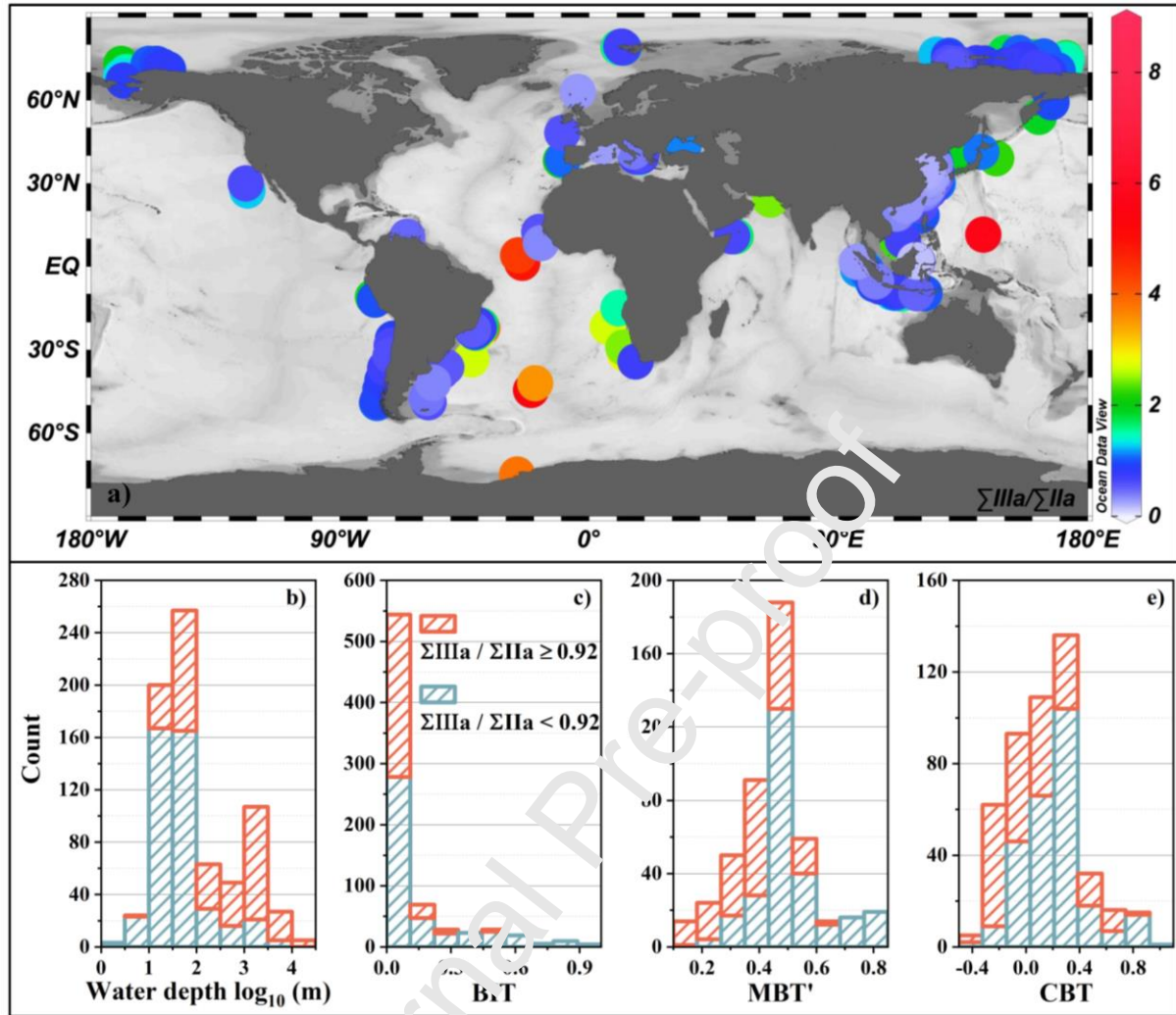


Figure 2. Box plots showing range of the $\Sigma_{IIIa}/\Sigma_{IIa}$, BIT and CBT indexes in the sinking particles (water depth of ca. 1350, 2500, 3000 and 4800 m) and the underlying sediments (water depth of ca. 5300 m) in the western North Pacific. The data are derived from Yamamoto et al. (2016).

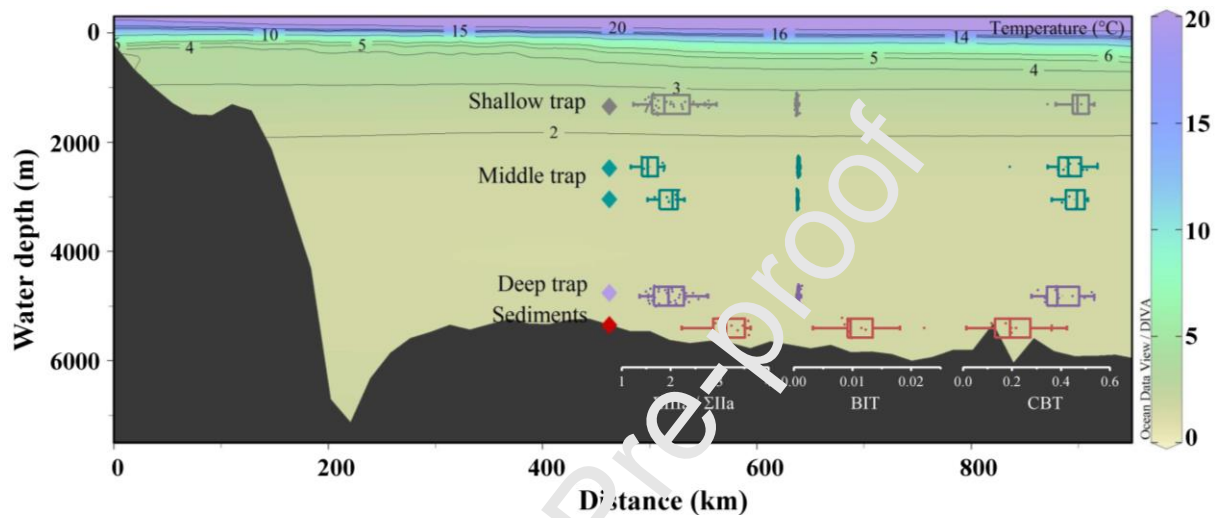


Figure 3. a) Box plot of the CBT index of marine derived brGDGTs ($\Sigma\text{IIIa}/\Sigma\text{IIa} \geq 0.92$) and mixing terrestrial/marine derived brGDGTs ($\Sigma\text{IIIa}/\Sigma\text{IIa} < 0.92$). Relationship between the MBT' index and b) annual sea surface temperature (SST) and c) annual bottom water temperature (BWT) of globally distributed marine sediments. R_p indicates the correlation coefficient of Pearson test.

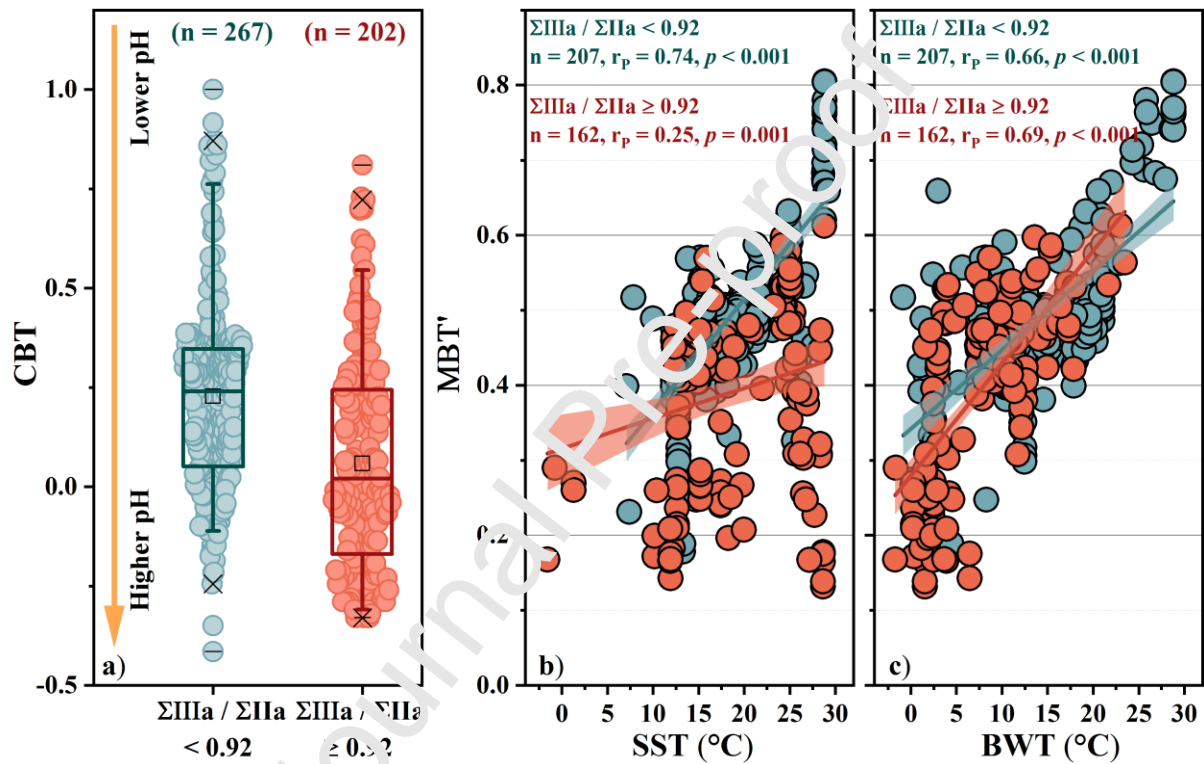


Figure 4. Isosurface plots of the $\Sigma_{IIIa}/\Sigma_{IIa}$ and MBT' indexes for surface sediments of the a, b) Brazilian margin (Ceccopieri et al., 2019), c, d) Berau delta (Sinninghe Damsté, 2016) and e, f) Svalbard fjord (Peterse et al., 2009).

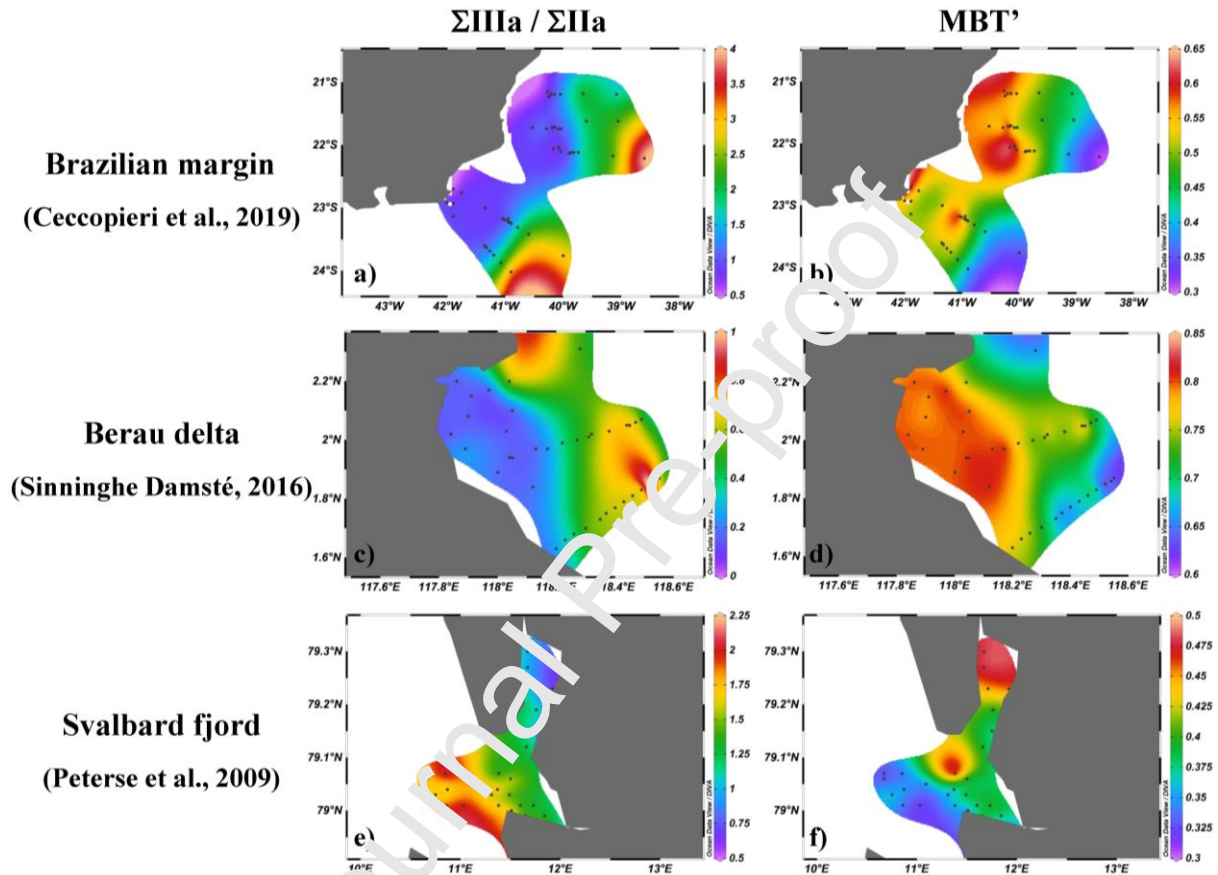


Figure 5. Relationship between the $\Sigma IIIa / \Sigma IIa$ index and the water depth of globally distributed marine sediments. R_s indicates the correlation coefficient of Spearman test.

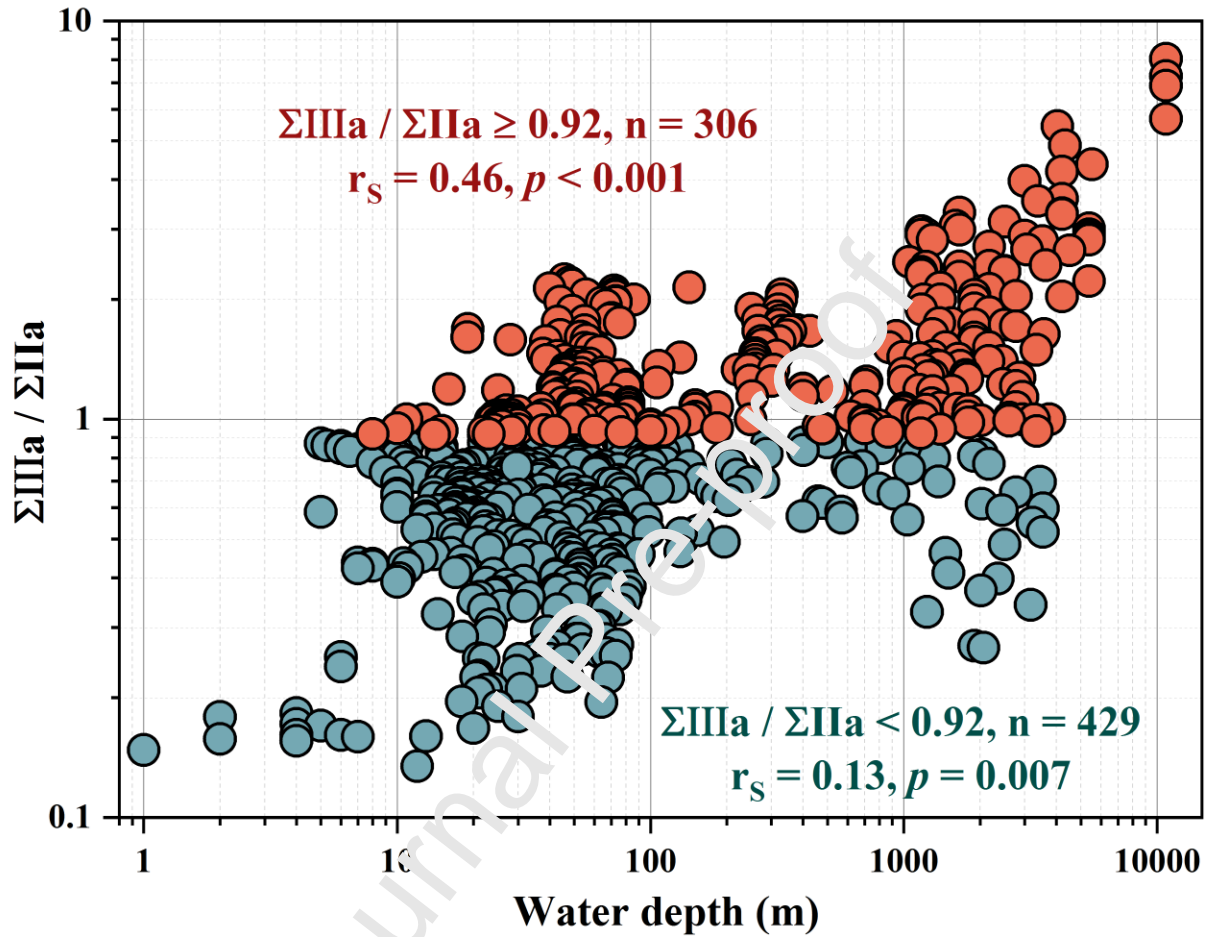


Figure 6. Records of a) sea surface temperature (SST) derived from the $U_{37}^{K'}$ index, d) the MBT index, e) the CBT index and f) the $\Sigma IIIa / \Sigma IIa$ index over the past 175 ka from core MD05-2896/7, South China Sea (Dong et al., 2015a; Dong et al., 2015b). Thin line and coarse line represent the original data and the 10-point moving average, respectively. b) Global relative sea level (Waelbroeck et al., 2002). c) Mn/Al ratio from core MD972142, South China Sea (Li et al., 2017).

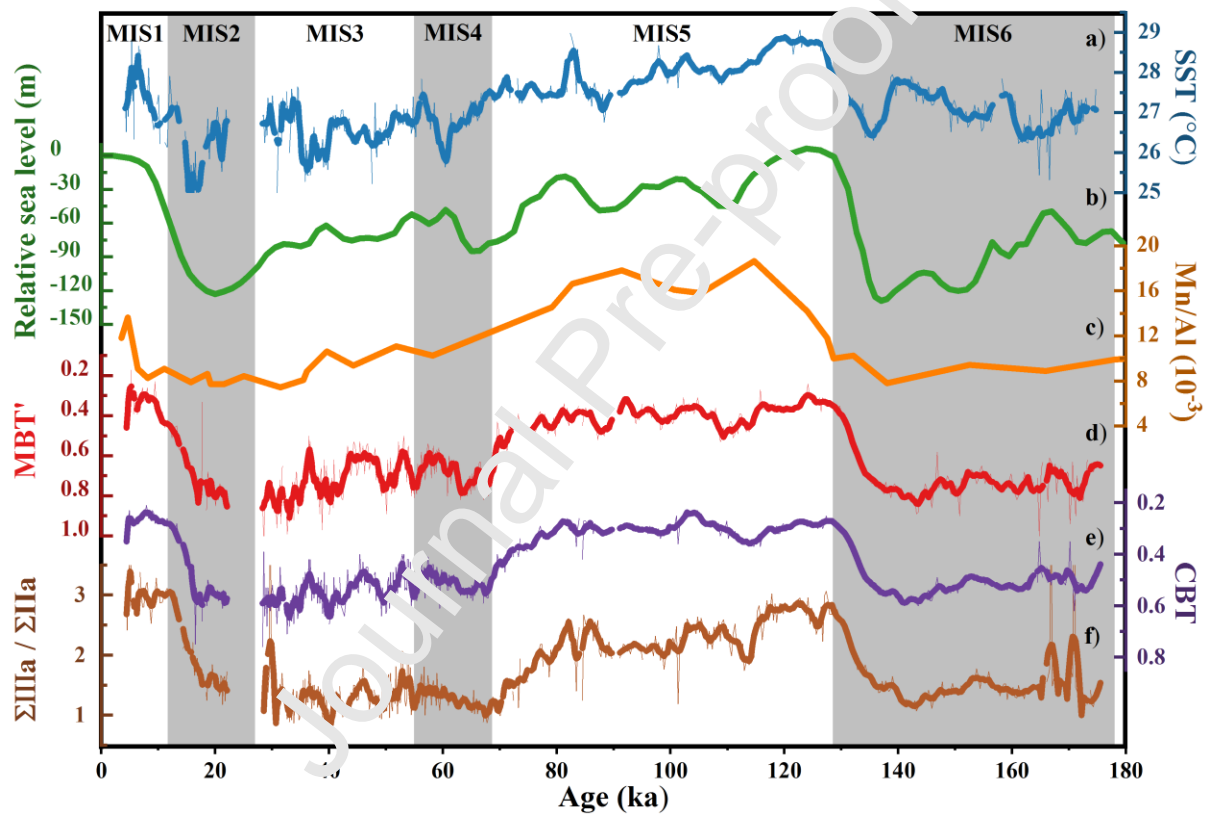
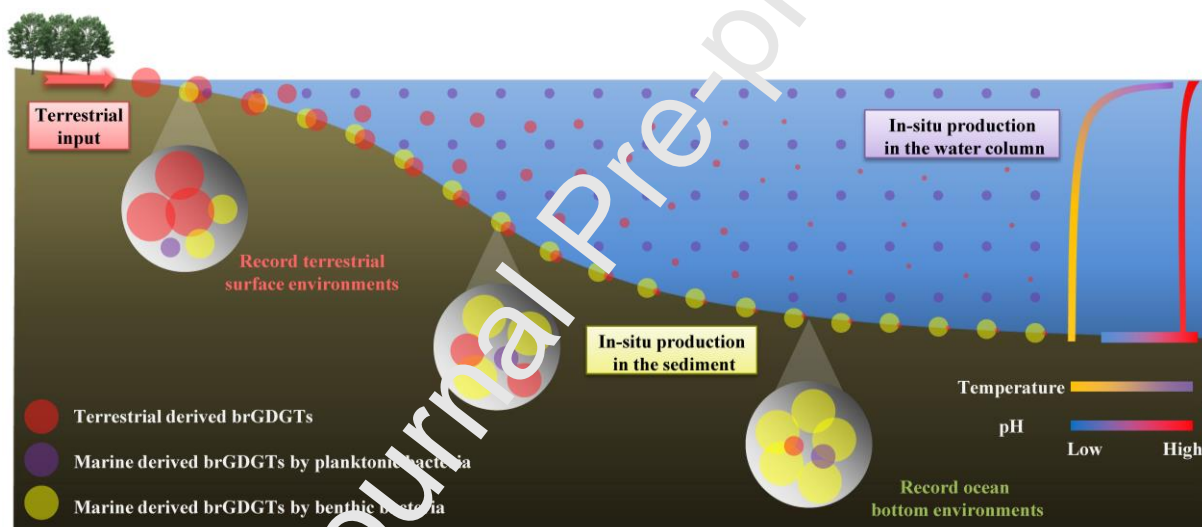


Figure 7. Schematic displaying the distribution of brGDGTs in marine environments. The brGDGTs in marine environments are derived from terrestrial input and marine in-situ production in the sediments and the water column. The contribution of benthic production to the brGDGTs' pool becomes dominant towards the deep-sea sediments. In the shallow area greatly influenced by terrestrial input, sedimentary brGDGTs record the signatures of the continental temperature and soil pH where the brGDGTs were produced. While in the deep ocean dominated by marine in-situ production, sedimentary brGDGTs are potential tools for recording environmental conditions of the ocean bottom.



Highlights

- A dataset of brGDGTs in global marine sediments was compiled.
- Benthic production of brGDGTs in marine sediments is ubiquitous on a global scale.
- Marine derived brGDGTs in sediments reflect ocean bottom environmental conditions