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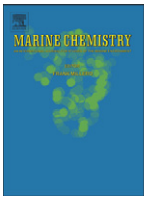

Hydroxylated isoprenoidal GDGTs in the Nordic Seas

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ABSTRACT

Archaea modify their membrane lipid composition to maintain integrity and exchange capabilities across a wide temperature range. The number of rings in the tetraether structure increases with higher ambient temperatures. Recently, novel glycerol dialkyl glycerol tetraethers (GDGTs) with an additional hydroxyl group on the alkyl chain were described (OH-GDGTs). Here we show that at a regional scale in the Nordic Seas (western Barents, Norwegian, Greenland, and Iceland Seas), the OH-GDGTs' contribution to the total archaeal GDGT pool as well as the ratio of specific OH-GDGTs versus crenarchaeol are higher in colder waters, especially in polar water masses. We also observe a shift in the number of rings of the OH-GDGTs with temperature. Thus, both the contribution of the OH-GDGTs to the total isoprenoidal GDGT pool and the number of rings in OH-GDGTs could be used as potential indicators of polar waters and might complement the currently used paleo sea surface temperature proxies in polar regions.

1. Introduction

Archaea are ubiquitously distributed in marine environments (DeLong, 1998; Fuhrman et al., 1992) where they constitute a large part (up to 30%) of the picoplankton (Karner et al., 2001). They are important contributors to the microbial communities in the Arctic pelagic and sea-ice communities (e.g., Bano et al., 2004; Galand et al., 2009; Collins et al., 2010). The isoprenoidal glycerol dialkyl glycerol tetraethers (isoGDGTs) are cell membrane lipids of Archaea that are used in modern microbial surveys to track changes in archaeal presence (e.g. Sinninghe Damsté et al., 2002) and to estimate past water temperatures in paleoclimate research (e.g. Schouten et al., 2002). Crenarchaeol, for example, is used as a marker for pelagic mesophilic Archaea, or more specifically for Crenarchaeota (Sinninghe Damsté et al., 2002) recently re-classified as Thaumarchaeota (Brochier-Armanet et al., 2008). On the other hand, changes in the relative abundance of the membrane tetraether lipids are an essential adaptation to the wide range of environmental conditions Archaea inhabit, e.g. to maintain membrane permeability and fluidity at various temperatures (Gliozzi et al., 2002; Gabriel and Chong, 2000).

The number of cyclopentane moieties of the archaeal GDGTs in marine sediments, expressed as the TEX_{86}-index, has been shown to vary linearly with sea surface temperature (SST) (Schouten et al., 2002; Kim et al., 2010). The linear relation is, however, weak in waters colder than 15 °C and a modified index 'TEX^{L}_{86}' has been proposed to reconstruct past SSTs in lower temperature ranges (Kim et al., 2010). Nonetheless, the scatter of the TEX^{L}_{86} index versus SST and of the standardized residuals is large (Kim et al., 2010), especially in the Arctic (Ho et al., in review). Also, SST estimates derived from both TEX_{86} and TEX^{L}_{86} are anomalously warm in the Arctic (Ho et al., in review) which constrains their use to resolve climatic episodes in cold areas. Parts of the Arctic Ocean are devoid of alkenone-producing Haptophytes, and hence the applicability of the well established organic SST proxy U^{K}_{37} is limited here. This leads to a potential advantage of GDGT-derived proxies, as GDGT-producing Archaea seem to be ubiquitous and GDGTs have been found in all water masses including the Arctic (e.g., Kim et al., 2010; Ho et al., in review).

Recently, Liu et al. (2012a) identified a new type of GDGT, the hydroxylated isoprenoidal GDGTs (OH-GDGTs) and showed their occurrence in temperate to tropical marine sediments. Liu et al. (2012b) pointed out that they may harbor potential as taxonomic biomarkers and geobiological proxies. Huguet et al. (2013) reported an increasing relative abundance of OH-GDGTs in cold regions and a significant correlation between the relative abundance of OH-GDGTs and SST in surface sediments at a global scale. The physiological function of the

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addition of a hydroxy group, however, is not yet certain. Sprott et al. (1990) proposed for diethers that hydroxy groups alter the cell membrane properties, either extending the polar head group region or creating a hydrophilic pocket. Liu et al. (2012a) suggested furthermore that the hydroxylation of the biphytanyl moiety may result in enhanced membrane rigidity.

In here we describe the occurrence of OH-GDGTs in surface water layers and surface sediments from subpolar and polar regions. We

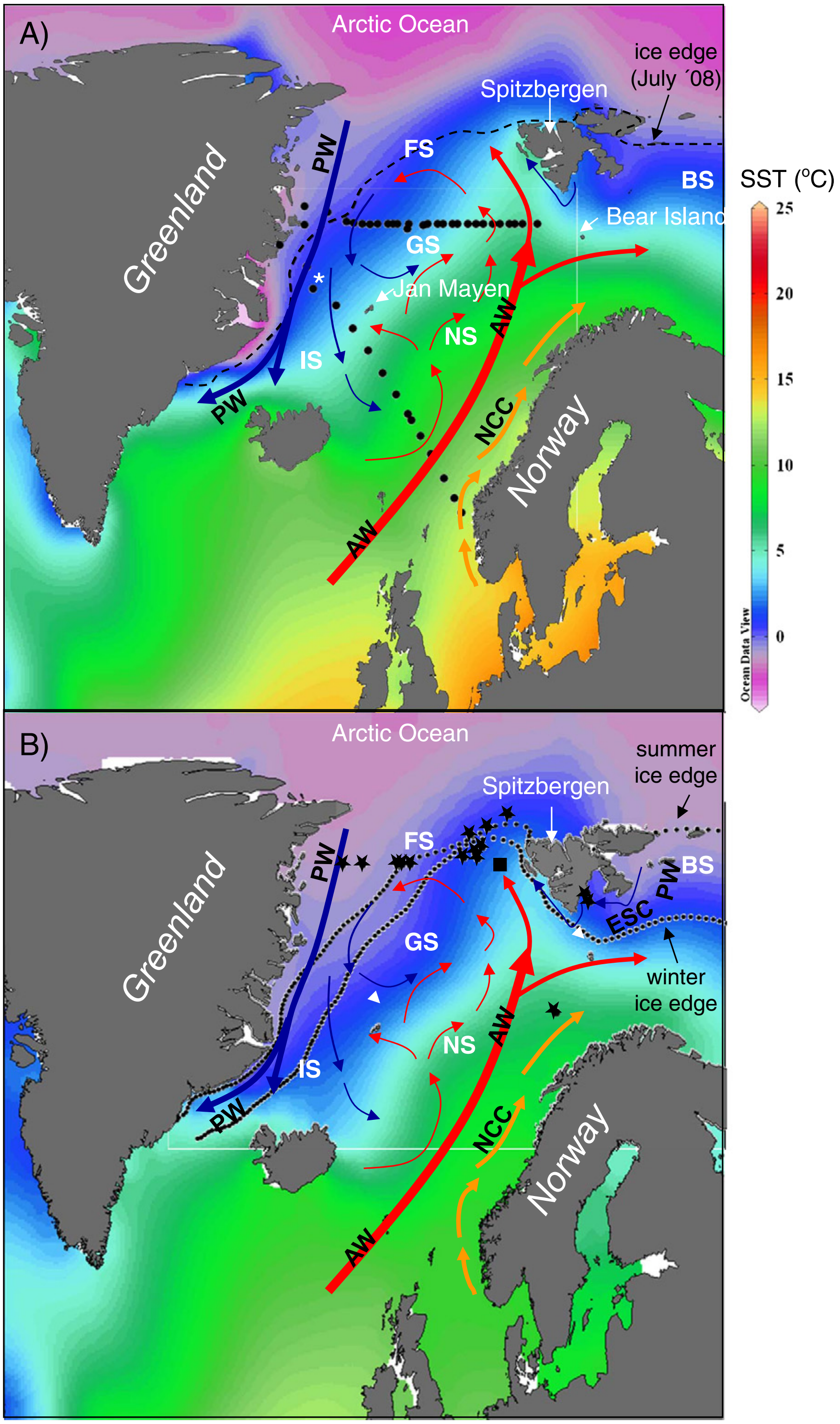

Fig. 1. Maps for (A) water samples and (B) sediments. (A) Map showing July SST (WOA09) and location of major currents. Dashed line indicates approximate sea ice extent at the time of sampling (21st of July 2008). Black dots indicate surface water sampling stations. Asterisk indicates sample location for which the example chromatogram is shown in Fig. 2. (B) Map showing annual SST (WOA09), location of major currents, and summer and winter sea ice extent (dotted lines). Summer and winter ice extent data are multidecadal means for April and August, respectively, from 1870 to 2002 provided by the National Snow & Ice Data Center, USA (Divine and Dick, 2007). Black stars indicate surface sediment sampling stations. Black square indicates location of retrieved sediment core. Abbreviations: BS — Barents Sea, ESC — East Spitsbergen Current, FS — Fram Strait, GS — Greenland Sea, IS — Icelandic Sea, NS — Norwegian Sea, NCC — Norwegian Coastal Current, AW — Atlantic Water, PW — Polar Water. Temperature ranges are −4 to 25 °C for both maps.