



# Seasonality of archaeal lipid flux and GDGT-based thermometry in sinking particles of high-latitude oceans: Fram Strait (79° N) and Antarctic Polar Front (50° S)

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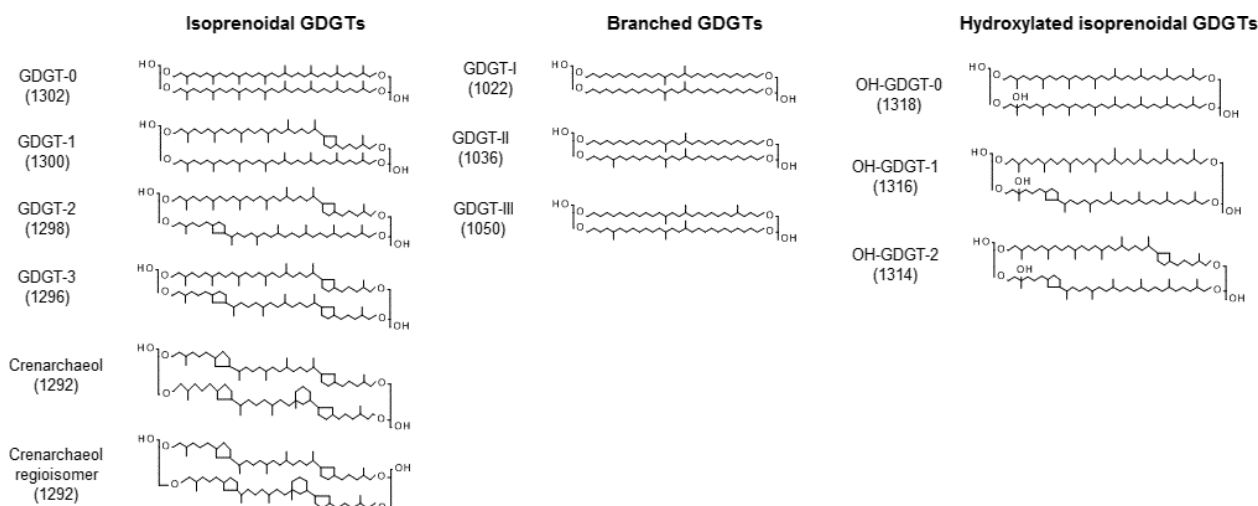
**Abstract.** The relative abundance of individual archaeal membrane lipids, namely of glycerol dialkyl glycerol tetraethers (GDGTs) with different numbers of cyclopentane rings, varies with temperature, which enables their use as a paleotemperature proxy index. The first GDGT-based index in marine sediments called TEX<sub>86</sub> is believed to reflect mean annual sea surface temperature (maSST). The TEX<sub>86</sub><sup>L</sup> is an alternative temperature proxy for “low-temperature” regions (< 15 °C), where the original TEX<sub>86</sub> proxy calibration shows a larger scatter. However, TEX<sub>86</sub><sup>L</sup>-derived temperatures still display anomalous estimates in polar regions. In order to elucidate the potential cause of the disagreement between the TEX<sub>86</sub><sup>L</sup> estimate and SST, we analyzed GDGT fluxes and TEX<sub>86</sub><sup>L</sup>-derived temperatures in sinking particles collected with time-series sediment traps in high-northern- and high-southern-latitude regions. At 1296 m depth in the eastern Fram Strait (79° N), a combination of various transporting mechanisms for GDGTs might result in seasonally different sinking velocities for particles carrying these lipids, resulting in strong variability in the TEX<sub>86</sub><sup>L</sup> signal. The similarity of flux-weighted TEX<sub>86</sub><sup>L</sup> temperatures from sinking particles and surface sediments implies an export of GDGTs without alteration in the Fram Strait. The estimated temperatures correspond to temperatures in water depths of 30–80 m, where nitrification might occur, indicating the favorable depth habitat of Thaumarchaeota. In the Antarctic Polar Front of the Atlantic sector (50° S), TEX<sub>86</sub><sup>L</sup>-derived temperatures displayed warm and cold biases compared to satellite-derived SSTs at 614 m depth, and its flux-weighted mean signal differs from

the deep signal at 3196 m. TEX<sub>86</sub><sup>L</sup>-derived temperatures at 3196 m depth and the surface sediment showed up to 7 °C warmer temperatures relative to satellite-derived SST. Such a warm anomaly might be caused by GDGT contributions from Euryarchaeota, which are known to dominate archaeal communities in the circumpolar deep water of the Antarctic Polar Front. The other reason might be that a linear calibration is not appropriate for this frontal region. Of the newly suggested SST proxies based on hydroxylated GDGTs (OH-GDGTs), only those with OH-GDGT-0 and crenarchaeol or the ring index (RI) of OH-GDGTs yield realistic temperature estimates in our study regions, suggesting that OH-GDGTs could be applied as a potential temperature proxy in high-latitude oceans.

## 1 Introduction

The knowledge that Thaumarchaeota, one phylum of Archaea, regulate the composition of their membrane lipids according to the surrounding water temperatures enabled the development of the paleothermometer TEX<sub>86</sub> (Kim et al., 2010; Schouten et al., 2002). TEX<sub>86</sub> is calculated based on the relative abundance of GDGT containing zero to three cyclopentane (GDGT-0 ~ 3) or four cyclopentane and one cyclohexane (crenarchaeol) ring (Fig. 1).

The ubiquity of Thaumarchaeota, even in the polar oceans where the widely applied U<sub>37</sub><sup>k</sup> proxy for sea surface temperature (SST) reconstructions is often problematic, supports the



**Figure 1.** Chemical structures and molecular ion  $m/z$  values of the isoprenoid glycerol dialkyl glycerol tetraethers (GDGTs), branched GDGTs, and hydroxylated GDGTs.

use of  $\text{TEX}_{86}$  in high-latitude regions (Bendle and Rosell-Melé, 2004; Ho et al., 2014). Nonetheless, the SST dependence of  $\text{TEX}_{86}$  in warm regions is stronger than in colder regions where SSTs are below  $5^\circ\text{C}$  (Kim et al., 2010). Thus, a logarithmic calibration of  $\text{TEX}_{86}^{\text{L}}$ , excluding the crenarchaeol regio-isomer, was suggested for regions where maSSTs are below  $15^\circ\text{C}$  (see Eq. 2; Kim et al., 2010). The authors speculated that the lack of correlation between the crenarchaeol regio-isomer and SST at low temperatures might be caused by genetically different GDGT producers. Moreover, all  $\text{TEX}_{86}^{\text{L}}$  calibrations for temperature include a rather large scatter, resulting in a calibration error of  $\pm 4^\circ\text{C}$  for the  $\text{TEX}_{86}^{\text{L}}$  calibration (Kim et al., 2010).

It has been questioned whether temperature is the only factor influencing the lipid composition (i.e.,  $\text{TEX}_{86}$ ), particularly in the regions where strong cold or warm biases were observed between  $\text{TEX}_{86}$  reconstructed and measured maSSTs. The biases of  $\text{TEX}_{86}$  calibrations have been attributed to enhanced production of GDGTs during seasons with favorable growth conditions for Thaumarchaeota (Pitcher et al., 2011; Wuchter et al., 2006b), a contribution of GDGTs from deep-dwelling communities (Kim et al., 2015; Lee et al., 2008; Taylor et al., 2013), terrestrial input (Weijers et al., 2006), influences of different archaeal communities (Lincoln et al., 2014; Turich et al., 2007), and/or other environmental factors (Huguet et al., 2006; Mollenhauer et al., 2015; Park et al., 2018; Turich et al., 2007).

To circumvent these problems, especially in cold regions, a handful of studies have developed dedicated calibrations on regional scales (Meyer et al., 2016; Seki et al., 2014; Shevenell et al., 2011). For the temperature estimate in the Eocene Arctic Ocean, a modified version of  $\text{TEX}_{86}$ , the index termed  $\text{TEX}'_{86}$ , was proposed (Sluijs et al., 2006). It excludes GDGT-3 to eliminated contribution from terrestrial

input, leading to significantly higher temperature estimates compared to the original  $\text{TEX}_{86}$  calibration. The  $\text{TEX}'_{86}$  values of 104 marine surface sediments showed a good linear correlation with maSSTs ( $R^2 = 0.93$ ) (Sluijs et al., 2006). Following the addition of seven  $\text{TEX}_{86}$  values from surface sediments collected at the continental margin of the western Antarctic Peninsula, Shevenell et al. (2011) modified the  $\text{TEX}_{86}$  calibration of Kim et al. (2008) for this region ( $\text{TEX}_{86} = 0.0125 \times \text{temp} + 0.3038$ ,  $R^2 = 0.82$ ) and estimated water temperatures for the Holocene.

Ho et al. (2014) evaluated  $\text{TEX}_{86}$  and the different calibrations in an extended set of surface sediments from the polar regions of both hemispheres and found that the global  $\text{TEX}_{86}$  calibration is suitable for subpolar regions (e.g., the Pacific sector of the Southern Ocean and the Subarctic Front in the North Pacific). Additionally, the  $\text{TEX}_{86}^{\text{L}}$  yields reasonable temperature estimates if regional calibrations are developed for high-latitude regions. Using a regression of water temperatures (at 20 m depth in August) versus  $\text{TEX}_{86}^{\text{L}}$  values, Seki et al. (2014) obtained plausible paleotemperature estimates for the Sea of Okhotsk and the subpolar North Pacific region. Meyer et al. (2016) confirmed later that this regional  $\text{TEX}_{86}^{\text{L}}$  calibration can also be applied to the subarctic northwest Pacific and the western Bering Sea.

More recently, hydroxylated GDGTs (OH-GDGTs), which contain an additional hydroxyl group in one of the biphytanyl moieties of GDGT-0, GDGT-1, and GDGT-2, have been recognized in marine sediments (Liu et al., 2012; Fig. 1). Based on the finding that the abundance of OH-GDGTs relative to the total isoprenoid GDGTs increases towards cold regions (Huguet et al., 2013), OH-GDGT-based SST calibrations have been proposed for high-latitude oceans (Fietz et al., 2013; Huguet et al., 2013).

In addition to the biological, phylogenetic, and statistical studies of the temperature proxy, sinking particles collected in specific regions can provide new perspectives on the distribution of GDGTs and TEX<sub>86</sub>-based temperature reconstructions. The study using time-series sediment traps can give insights into the seasonal variability of lipid flux and temperature signals, and allows the comparison of the lipid signal to those in underlying sediments. Only one TEX<sub>86</sub> study using a time-series sediment trap in high-latitude regions has been conducted near Iceland (Rodrigo-Gámiz et al., 2015), regardless of the interest in polar regions, where temperature reconstructions with lipid proxies are troublesome. Here, we examine the seasonal GDGT production and TEX<sub>86</sub><sup>L</sup>-derived temperatures in comparison to measured SSTs in order to better understand the drivers influencing TEX<sub>86</sub><sup>L</sup> values in sinking particles collected in two high-latitude oceans (eastern Fram Strait, 79° N, and Antarctic Polar Front of the Atlantic sector, 50° S). The temperatures derived from OH-GDGT proxies were also calculated to evaluate the applicability of these novel proxies for high-latitude regions.

## 2 Study area

### 2.1 Fram Strait

The Fram Strait is located west of Spitsbergen, where the heat exchange occurs between the Arctic Ocean and the North Atlantic (Fig. 2). Relatively warm and nutrient-rich Atlantic water (AW) is transported by the West Spitsbergen Current (WSC) to the central Arctic Ocean, while colder and less saline Arctic water is transported along the East Greenland Current (EGC) to the Nordic Seas (Fig. 2). Part of the WSC is separated from the northward flow and recirculated within the region (Manley, 1995; Soltwedel et al., 2016). Eddies, mixing, and recirculating Atlantic water contribute to the hydrographic complexity observed in the Fram Strait (Walczowski, 2013). The volume transport of the WSC and AW within the WSC displays strong seasonality with maxima in March and minima in July for the WSC and maxima in late autumn and winter and minima in June for the AW (Beszczynska-Möller et al., 2012b). The occurrence of sea ice and its variability has an effect on the particle fluxes as well as on benthic ecosystems (Bauerfeind et al., 2009; Hebbeln and Wefer, 1991; Soltwedel et al., 2016). Sinking particles in the Fram Strait, including organic and terrigenous material, are well known to originate not only from the photic zone in the upper water column, but also from the Svalbard archipelago and Siberian shelf, from where they are transported by sea ice (Hebbeln, 2000; Lalande et al., 2016). Phytoplankton blooms enhance the vertical flux of biogenic components (organic carbon, carbonate, and opal) in spring–summer, but the downward flux of particles is likely affected by the environmental conditions (e.g., hydrographic changes, sea ice extent, and atmospheric low pressure) on an annual

timescale (Wassmann et al., 2006). Based on oceanographic measurements from mooring arrays at multiple depths across the Fram Strait (from 7° W to 9° E, 78.3° N), Beszczynska-Möller et al. (2012a) found two warm anomalies in the Atlantic water through the Fram Strait in 1999–2000 and 2005–2007. During the second warm anomaly, community structures of phytoplankton and zooplankton were affected by the decreased sea ice in this region (Lalande et al., 2013; Nöthig et al., 2015) and had a profound influence on biogenic sediment fluxes.

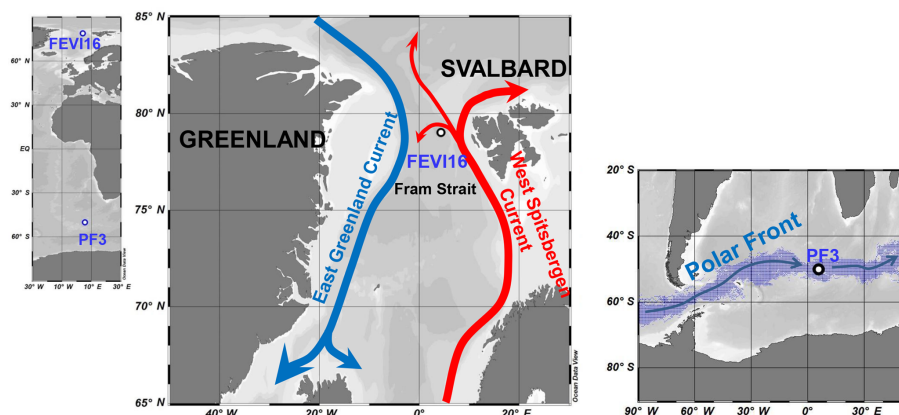
### 2.2 Antarctic Polar Front

The Antarctic Polar Front (APF) region is one of the major frontal zones within the westerly wind-driven Antarctic Circumpolar Current (ACC) (Fig. 2). Spatial and temporal variability of the APF is strongly affected by seafloor topography. The APF has an average width of 43 km (1987–1993), which moves southward during the austral summer and reaches its northernmost position during the austral winter (Moore et al., 1999). The ACC is well known for its meandering jet flow and eddy formation (Moore et al., 1999; Orsi et al., 1995), which likely stimulate phytoplankton blooms and primary production in this region by supplying growth-limiting nutrients, and/or bringing deeper-dwelling phytoplankton into the photic zone (Abbott et al., 2001; Moore and Abbott, 2000, 2002). Thus, the hydrographic structure is an important factor controlling the distribution of phytoplankton and small zooplankton in this region (Read et al., 2002). The highest chlorophyll-*a* levels of up to 3.5 mg m<sup>-3</sup> were recorded in the APF during the early austral summer of 1990–1991, and it co-occurred with elevated silicate levels as well as water column stratification (Laubscher et al., 1993). The current speed close to the PF3 trap was recorded to be up to 8 cm s<sup>-1</sup> (Walter et al., 2001). Read et al. (2002) reported the vertical profile of currents measured between the surface and 400 m depth in the polar frontal regions in December 1995. The currents recorded at the surface and subsurface of the Polar Front were much stronger (30–50 cm s<sup>-1</sup>) than the ones measured at 700 m depth (1–8 cm s<sup>-1</sup>) by Walter et al. (2001).

## 3 Materials and methods

### 3.1 Sediment traps

A time-series sediment trap system was moored to collect sinking particles at the eastern Fram Strait (FEVI16) between July 2007 and July 2008 at 1296 m water depth (Table 1, Fig. 2). Two traps (PF3) were deployed in the permanent ice-free area of the APF in the Atlantic sector at 614 and 3196 m water depth from November 1989 to December 1990 (Table 1, Fig. 2). The cone-shaped funnel of the Kiel trap systems had a 0.5 m<sup>2</sup> collection area, and the collection periods of each individual sample cup were programmed de-



**Figure 2.** Map of sediment trap locations in high northern latitudes (FEVI16) and in Antarctic Polar Front (PF3). In the Fram Strait, the northward red arrow represents the West Spitsbergen Current bringing warm and saline Atlantic waters to the Arctic and the southward blue arrow displays the East Greenland Current transporting cold and fresh water to the Norwegian Sea. Around the Antarctic, the digitized blue shadow exhibits the location of the Antarctic Polar Front for the years 2002–2014 at weekly resolution (Freeman and Lovenduski, 2016). The blue arrow indicates the clockwise Antarctic Circumpolar Current. Ocean Data View is used for mapping (Schlitzer, 2017; available at <https://odv.awi.de>, last access: 3 May 2018).

pending on the expected time of ice cover and/or the seasonality of the production. The sampling cups were filled with filtered seawater enriched in sodium chloride (NaCl) to achieve a salinity of 40 psu, and they were poisoned with mercury chloride ( $\text{HgCl}_2$ ; 0.14 % final solution) for sample preservation. For FEVI16 samples, zooplankton “large swimmers” were removed using forceps before splitting each sampling cup into smaller subsamples (Lalande et al., 2016). For PF3 samples, large swimmers were removed using forceps and a sieve (mesh size: 1 mm mesh) in the laboratory (Fischer et al., 2002). Afterwards, the samples were split for different purposes. A current meter (RCM 9/11) was attached to both trap moorings, and the datasets are available on PANGAEA (<https://doi.org/10.1594/PANGAEA.845610>; FEVI16) and in Walter et al. (2001; PF3).

### 3.2 Mass flux

Split FEVI16 samples were filtered onto GF/F filters (pre-combusted at 500 °C for 4 h) for organic carbon, organic nitrogen (PON), and carbonate and onto cellulose acetate filters for biogenic opal quantification. Organic carbon (POC) and organic nitrogen (PON) were measured with a CHN elemental analyzer after 0.1 N HCl treatment. Carbonate was calculated by subtracting the weight from total mass after 0.1 N HCl treatments and rechecked according to the aragonite contents. The detailed methods used for biogenic silica can be found in Lalande et al. (2016). Split PF3 samples were freeze-dried for further processing. Decalcified samples (6 N HCl) were analyzed for POC using a CHN elemental analyzer. Total nitrogen (TN) was also determined. Carbonate was calculated by subtracting POC from total carbon (TC), which was directly measured without decalcification ( $\text{carbonate} = 8.33 \times (\text{TC} - \text{POC})$ ). The methodology for

the quantification of biogenic silica is described in Fischer et al. (2002).

### 3.3 GDGT analyses

For GDGT analysis, total lipids were extracted with a solvent mixture (9 : 1 *v/v* dichloromethane (DCM) : methanol (MeOH)) using an ultrasonic bath for 10 min and a centrifuge for 5 min, after which the supernatant was decanted. This process was repeated three times and the supernatants were combined. Before the extraction process, a known amount of C<sub>46</sub>-GDGT (internal standard) was added to each sample for GDGT quantification.

Following saponification of total lipids with 1 mL of 0.1 M potassium hydroxide (KOH) in a mixture of MeOH : purified water 9 : 1 (*v/v*) at 80 °C for 2 h, neutral lipids (NLs) were recovered with 1 mL of hexane (three times). NLs were separated into F1 (apolar), F2 (ketone), and F3 (polar) polarity fractions eluted in 2 mL of hexane, 4 mL of DCM : hexane (2 : 1 *v/v*), and 4 mL of DCM : MeOH (1 : 1 *v/v*), respectively, using deactivated silica-gel chromatography (mesh size: 70–230  $\mu\text{m}$ ).

The F3 polar fraction containing GDGTs was redissolved in 500  $\mu\text{L}$  of hexane : isopropanol 99 : 1 (*v/v*) and filtered through a polytetrafluoroethylene (PTFE) filter (pore size: 0.45  $\mu\text{m}$ ) into a glass insert of a 2 mL vial according to Hopmans et al. (2000). The filtered polar fraction was diluted with hexane : isopropanol 99 : 1 (*v/v*) to a concentration of approximately 2  $\text{mg mL}^{-1}$  before the instrumental analysis.

GDGTs were analyzed using high-performance liquid chromatography/atmospheric pressure chemical ionization mass spectrometry (HPLC/APCI-MS) according to Chen et al. (2014). Molecular ions *m/z* 1302, 1300, 1298, 1296, and 1292 for isoprenoid GDGTs and 1050, 1036, and 1022 for

**Table 1.** Information on FEVI16 and PF3 trap.

Trap name	FEVI16	PF3
Region	Eastern Fram Strait	Antarctic Polar Front
Location		
Latitude (° N)	79.02	−50.13
Longitude (° E)	4.35	5.83
Water depth (m)	2580	3785
Trap depth (m)	1296	614 3196
Deployment period		
Start	23 Jul 2007	10 Nov 1989
End	30 Jun 2008	23 Dec 1990
Sampling interval (d)	10–31	21, 42*
Cruise reports	ARK-XXII/1c (Klages and Participants, 2007) ARK-XXIII/2 (Kattner and Participants, 2009)	ANT-VIII/3 (Gersonde and Participants, 1990) ANT-IX/2 (Fahrbach and Cruise Participants, 1992)

\* The exact sampling interval of each sample at FEVI and PF3 can be found on PANGAEA (<https://doi.pangaea.de/10.1594/PANGAEA.897268>).

branched GDGTs were determined and quantified in relation to the molecular ion  $m/z$  744 of the C<sub>46</sub>-GDGT. The late eluting peaks of OH-GDGT-0, OH-GDGT-1, and OH-GDGT-2 with  $m/z$  1318, 1316, and 1314 were also determined in the  $m/z$  1300, 1298, and 1296 scan, as described by Fietz et al. (2013).

A lab-internal standard sediment was repeatedly analyzed along with the samples to assess the analytical error. The standard deviation of replicate analyses was 0.1 units of TEX<sub>86</sub><sup>L</sup> and 6 % for isoprenoid GDGT concentrations.

### 3.4 GDGT flux and indices

TEX<sub>86</sub><sup>L</sup> values were calculated according to Kim et al. (2010).

$$\text{TEX}_{86}^L = \log_{10} \frac{[\text{GDGT} - 2]}{[\text{GDGT} - 1] + [\text{GDGT} - 2] + [\text{GDGT} - 3]}, \quad (1)$$

where the numbers represent the number of cyclopentane moieties in the isoprenoid GDGTs.

TEX<sub>86</sub><sup>L</sup> values were converted into temperatures using the following equation (Kim et al., 2010):

$$\text{SST (}^\circ\text{C)} = 67.5 \times \text{TEX}_{86}^L + 46.9$$

$$(R^2 = 0.86, n = 396, \text{ calibration error: } \pm 4^\circ\text{C}). \quad (2)$$

GDGT flux represents the sum of fluxes of individual GDGTs, which are used for calculating TEX<sub>86</sub><sup>L</sup>.

$$\text{GDGT flux} = [\text{GDGT} - 1] + [\text{GDGT} - 2] + [\text{GDGT} - 3] \quad (3)$$

OH-GDGT-based indices and estimated temperatures were calculated using the following equations (Eq. 4 by Fietz et

al., 2013; Eqs. 5 and 6 by Lü et al., 2015):

$$\frac{[\text{OH-GDGT} - 0]}{[\text{Cren}]} = 0.25 - 0.025 \times \text{SST (}^\circ\text{C)}$$

$$(R^2 = 0.58, n = 13, \text{ calibration error: } \pm 1.5^\circ\text{C}), \quad (4)$$

$$\text{RI-OH}' = \frac{[\text{OH-GDGT} - 1] + 2 \times [\text{OH-GDGT} - 2]}{[\text{OH-GDGT} - 0] + [\text{OH-GDGT} - 1] + [\text{OH-GDGT} - 2]}, \quad (5)$$

$$\text{RI-OH}' = 0.0382 \times \text{SST (}^\circ\text{C)} + 0.1$$

$$(R^2 = 0.75, n = 107, \text{ calibration error: } \pm 6^\circ\text{C}). \quad (6)$$

BIT, an index of relative contribution of terrestrial versus marine input, was calculated according to Hopmans et al. (2004).

$$\text{BIT} = \frac{[\text{I} + \text{II} + \text{III}]}{[\text{I} + \text{II} + \text{III} + \text{Cren}]} \quad (7)$$

The Roman numerals I, II, and III refer to the branched GDGTs with four, five, and six methyl moieties, and Cren represents crenarchaeol containing four cyclopentane moieties and one cyclohexane ring (Fig. 1). A terrestrial effect on TEX<sub>86</sub> can be significant when the BIT value is > 0.3 (Weijers et al., 2006).

MI, the methane index indicating the relative contribution of GDGTs derived from methanotrophic Archaea to those from planktonic Thaumarchaeota, was calculated as follows (Zhang et al., 2011):

$$\text{MI} = \frac{[\text{GDGT} - 1] + [\text{GDGT} - 2] + [\text{GDGT} - 3]}{[\text{GDGT} - 1] + [\text{GDGT} - 2] + [\text{GDGT} - 3] + [\text{Cren}] + [\text{Cren}']}. \quad (8)$$

Cren' represents the regio-isomer of crenarchaeol. Contributions from methanotrophic Archaea are considered to be significant when MI > 3.

%GDGT-0, an indicator of a methanogenic source of GDGTs with a %GDGT value > 67 %, was calculated as follows (Inglis et al., 2015):

$$\%GDGT-0 = \frac{[GDGT-0]}{[GDGT-0] + [Cren]} \times 100\%. \quad (9)$$

RI, the ring index, is a tool for identifying a potential non-thermal influence on GDGT distributions. A sample's ring index is defined as follows (Zhang et al., 2016):

$$RI_{\text{sample}} = 0 \times [GDGT-0] + 1 \times [GDGT-1] + 2 \times [GDGT-2] + 3 \times [GDGT-3] + 4 \times [Cren] + 4 \times [Cren']. \quad (10)$$

$|\Delta RI|$  indicates the residual of a sample's RI ( $RI_{\text{sample}}$ ) from a calculated RI ( $RI_{\text{calculated}}$ ) based on the global  $TEX_{86}$  – RI regression.  $RI_{\text{calculated}}$  and  $|\Delta RI|$  are defined as follows:

$$RI_{\text{calculated}} = -0.77(\pm 0.38) \times TEX_{86} + 3.32(\pm 0.34) \times (TEX_{86})^2 + 1.59(\pm 0.10) \quad (R^2 = 0.87, n = 531), \quad (11)$$

$$|\Delta RI| = RI_{\text{calculated}} - RI_{\text{sample}}. \quad (12)$$

Potential nonthermal influences on  $TEX_{86}$  can be recognized when  $|\Delta RI|$  is > 0.3 (Zhang et al., 2016).

### 3.5 Environmental parameters

$TEX_{86}^L$ -derived temperatures were compared to satellite-derived SSTs obtained by the Advanced Very High Resolution Radiometer (AVHRR), which has a spatial grid resolution of 0.25° and temporal resolution of 1 d (Reynolds et al., 2007). The satellite-derived SSTs were averaged over the collection period of each sample cup. The depth profiles of mean annual water temperature and nitrate concentration were obtained from the World Ocean Atlas 2013 (WOA13) representing averaged values for the years 1955–2012 (Garcia et al., 2013; Locarnini et al., 2013).

## 4 Results

### 4.1 Eastern Fram Strait: FEVI16

#### 4.1.1 Mass and GDGT fluxes

The mass flux data of the FEVI16 trap have been published previously by Lalande et al. (2016). In summary, the mass flux (opal + POC + PON + lithogenic fluxes) measured at 1296 m water depth varied in between 18.5 and 652.9 mg m<sup>-2</sup> d<sup>-1</sup>, with minima in the winter season (November 2007–February 2008; Fig. 3a). Lithogenic flux is presented separately in Fig. 3a due to its significantly higher contribution to total mass flux. The composition of

sinking particles changed between seasons. Lithogenic material flux showed sudden pulses in spring (March and April–May 2008). POC was predominant in mid-July 2007. Carbonate flux was elevated in March, early May, and mid-June 2008. Opal was dominant in early September 2007 and mid-May to June 2008.

GDGT fluxes varied between 7.2 and 85.9 ng m<sup>-2</sup> d<sup>-1</sup> (Fig. 3b). Peaks of GDGT flux occurred in August–September 2007 and March and May–June 2008, while minima were observed in the winter season (November 2007–February 2008). The GDGT flux was strongly correlated with opal flux ( $R^2 = 0.82$ ,  $p < 0.0001$ ) when two GDGT maxima in late September 2007 and March 2008 were excluded and with carbonate flux ( $R^2 = 0.86$ ,  $p < 0.0001$ ) except for the last 2 months in May–June 2008 (Fig. 3a and b).

#### 4.1.2 $TEX_{86}^L$ thermometry

$TEX_{86}^L$  values varied between −0.70 and −0.64 and their  $TEX_{86}^L$ -derived temperatures (Kim et al., 2010, calibration) ranged between 0.6 and 3.7 °C (Fig. 3b and f). The flux-weighted mean  $TEX_{86}^L$  temperature during the trap deployment period was 2.8 °C. Two surface sediment  $TEX_{86}^L$  values (−0.66 at PS68-251/2, 79.1° N, 4.6° E; −0.65 at PS68-271/2, 79.3° N, 4.3° E) are available near the FEVI16 trap (Ho et al., 2014). Most index values of BIT, MI, %GDGT-0, and  $|\Delta RI|$  for samples from FEVI16 were below the critical values of each index (> 0.3 for BIT and MI, > 67 % for %GDGT-0, > 0.3 for  $|\Delta RI|$ ). One sample with a  $|\Delta RI|$  value > 0.3 was found, indicating a nonthermal effect on  $TEX_{86}$  (Table A1 in the Appendix).

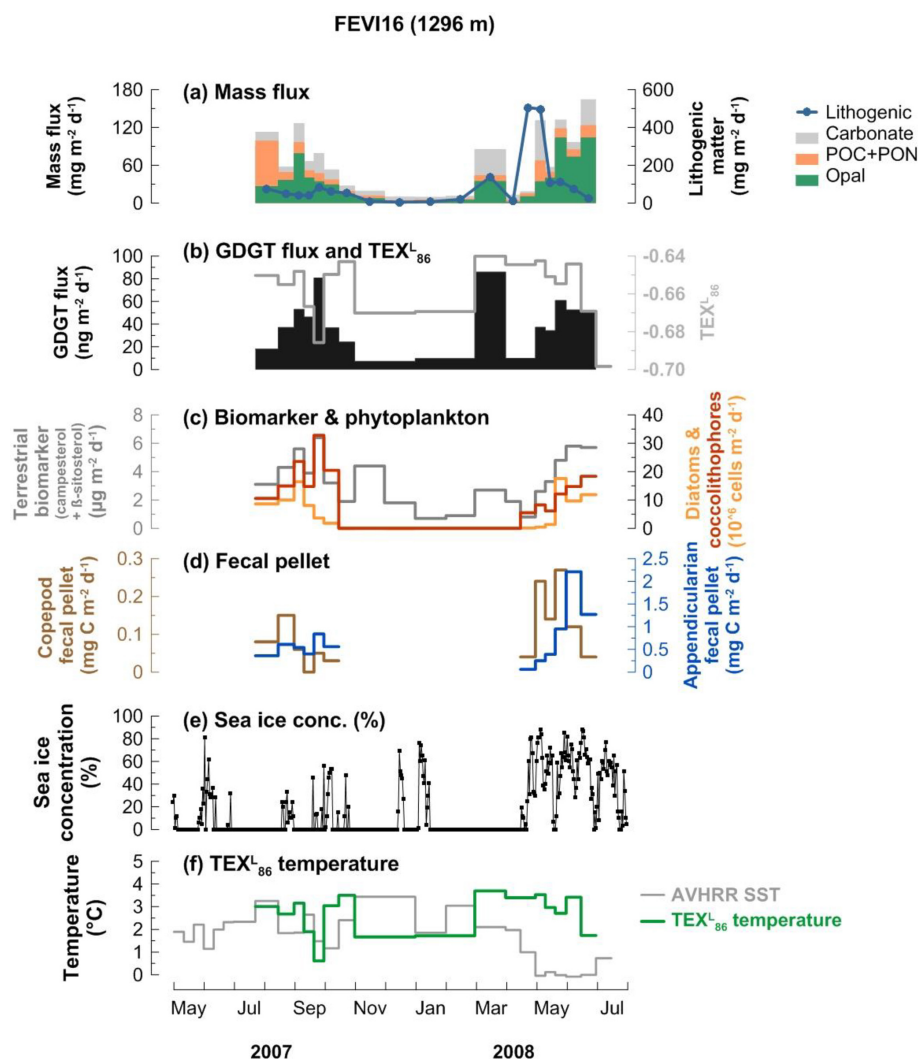
#### 4.1.3 Fractional abundance of OH-GDGTs

All OH-GDGTs were clearly present in FEVI16 samples. OH-GDGT-0 was predominant in the OH-GDGT pool, accounting for 87 %–95 % (Fig. 4a). The proportion of OH-GDGTs in the sum of iso- and OH-GDGTs ranged between 7 % and 11 % during the trap deployment period (Fig. 4a). Estimated temperatures calculated using the calibrations as described in Eqs. (4) and (6) (Fietz et al., 2013; Lü et al., 2015) ranged between −0.3 and 2.5 °C and between −0.9 and 1.4 °C, respectively. The last two data points for the trap time series were unusually low when using Eq. (4) (Fig. 4b).

### 4.2 Antarctic Polar Front: PF3

#### 4.2.1 Mass and GDGT fluxes

The mass flux data of the PF3 trap have been published previously by Fischer et al. (2002). The mass flux at the shallow trap ranged between 19.7 and 592.1 mg m<sup>-2</sup> d<sup>-1</sup> with a distinct seasonal variability (Fig. 5a). Flux maxima occurred in November 1989 to early March 1990 and mid-October to November 1990, while it stayed relatively low



**Figure 3.** Changes in mass flux (a), GDGT flux (b), terrestrial biomarker flux and diatom and coccolithophore abundances (c), fecal pellet flux (d), sea ice concentration (e), and  $\text{TEX}_{86}^L$ -derived temperatures and satellite-derived SSTs obtained at  $79.125^{\circ} \text{N}$ ,  $4.375^{\circ} \text{E}$  near the trap site (f) at 1296 m water depth at the eastern Fram Strait site, FEVI16, during the deployment period July 2007–July 2008. Panels (a), (c), (d), and (e) were previously reported by Lalande et al. (2016).

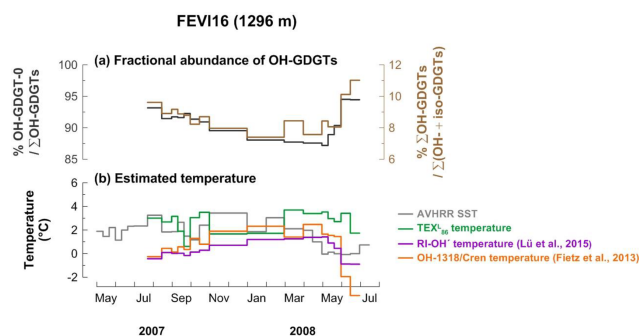
from mid-March to mid-October 1990. At the deep trap, the mass flux varied between 0 and  $516.6 \text{ mg m}^{-2} \text{d}^{-1}$  (Fig. 5d). The mass flux peaked in mid-December 1989 and January and March 1990 but stayed low for the remaining sampling period except for mid-May 1990.

There was no clear correlation between organic material and GDGT fluxes at either depth. The fluxes in GDGTs ranged from  $10.5$  to  $73.9 \text{ ng m}^{-2} \text{d}^{-1}$  at 614 m and from  $0.5$  to  $153.9 \text{ ng m}^{-2} \text{d}^{-1}$  at 3196 m depth (Fig. 5b and e). GDGT fluxes were mostly enhanced during the low-mass-flux season at the shallow trap (Fig. 5b). GDGT flux maxima occurred in November and December 1989 and late May and early November 1990 at the deep trap (Fig. 5e). GDGT analysis was not possible between late July and October 1990 in

the deep trap since very little material was collected during this time period.

#### 4.2.2 $\text{TEX}_{86}^L$ thermometry

$\text{TEX}_{86}^L$  values varied between  $-0.58$  and  $-0.67$  and  $-0.55$  and  $-0.59$  at the shallow and deep traps, respectively (Fig. 5b and e). The  $\text{TEX}_{86}^L$ -derived temperatures ranged between  $1.3$  and  $7.8^{\circ}\text{C}$  for the shallow trap and  $7.4$  and  $9.8^{\circ}\text{C}$  for the deep trap (Fig. 5c and f). The flux-weighted mean  $\text{TEX}_{86}^L$  temperatures were  $4.6$  and  $8.5^{\circ}\text{C}$  at the shallow and deep traps, respectively. In some samples, GDGT-3 was below the detection limit. In these cases,  $\text{TEX}_{86}^L$  values and estimated temperatures were not included in the figures, but GDGT flux was calculated. A surface sediment  $\text{TEX}_{86}^L$  value of  $-0.56$  (CHN-115-4-34;  $51.00^{\circ} \text{S}$ ,  $5.33^{\circ} \text{E}$ ) in the vicinity of the PF3



**Figure 4.** Changes in fractional abundance of OH-GDGTs (a) and estimated temperatures based on OH-related calibrations (b) at site FEVI16. The purple and orange lines indicate the calculated temperatures using the calibrations by Fietz et al. (2013) (Eq. 4) and Lü et al. (2015) (Eq. 6), respectively. The green line indicates the  $\text{TEX}_{86}^L$ -derived temperatures.

trap site (50.13° S, 5.83° E) can be found in the  $\text{TEX}_{86}$  global calibration dataset (Kim et al., 2010), and its  $\text{TEX}_{86}^L$ -derived temperature estimate is 9.1 °C. Most samples did not exceed the index values indicating nonthermal impacts on  $\text{TEX}_{86}$  (see Sect. 3.4). Two samples from the deep trap between early November and late December 1989, however, showed MI values between 0.3 and 0.5 (Table A1).

#### 4.2.3 Fractional abundance of OH-GDGTs

All OH-GDGTs were clearly present in samples from both depths. OH-GDGT-0 was predominant in the OH-GDGT pool (76 %–89 % for the shallow trap, 77 %–83 % for the deep trap; Fig. 6a and c). The proportion of OH-GDGTs in the sum of iso- and OH-GDGTs ranged from 4 % to 7 % and 5 % to 6 % at the shallow and deep traps, respectively, (Fig. 6a and c). Estimated temperatures calculated according to Eqs. (4) and (6) ranged between 3.0–6.1 and 1.1–5.1 °C in the shallow trap (Fig. 6b). With the same calibrations, estimated temperatures varied between 3.6–5.8 and 2.8–5.2 °C in the deep trap (Fig. 6d).

## 5 Discussion

### 5.1 Eastern Fram Strait (79° N)

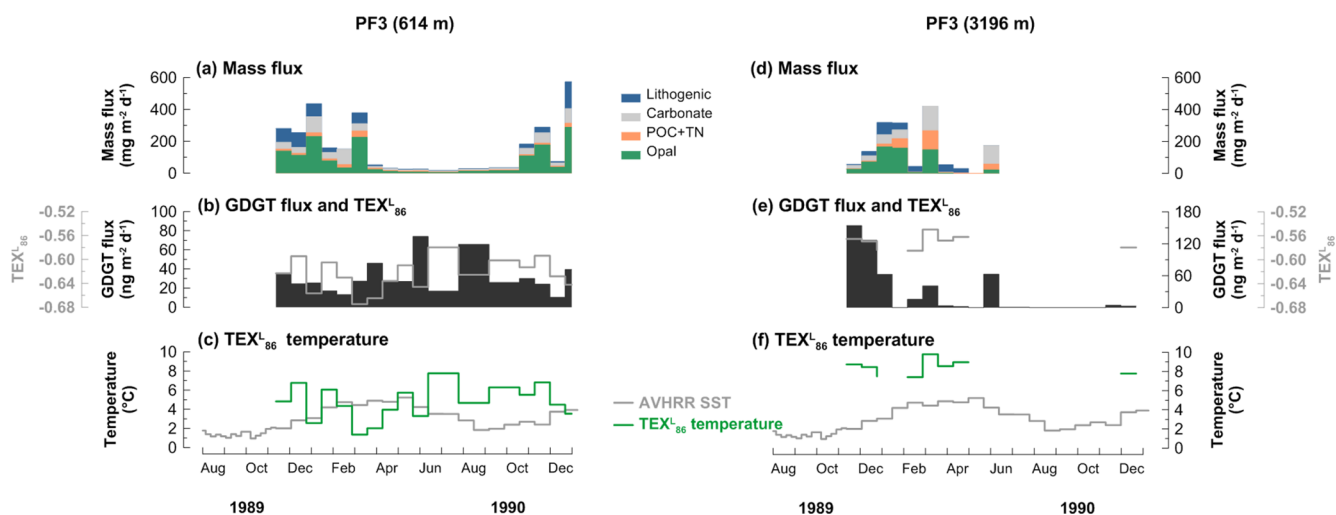
#### 5.1.1 GDGT flux and particle export

Organic matter (including GDGTs), formed in the upper ocean, is exported to the bathypelagic mainly as zooplankton fecal pellets or marine snow aggregates (Fischer and Karakaş, 2009; Wuchter et al., 2006b). The velocities of the sinking aggregates vary depending on their composition (Fischer and Karakaş, 2009; Iversen and Ploug, 2010) and physical characteristics. For instance, aggregates formed from coccolithophores are ballasted by carbonate and may

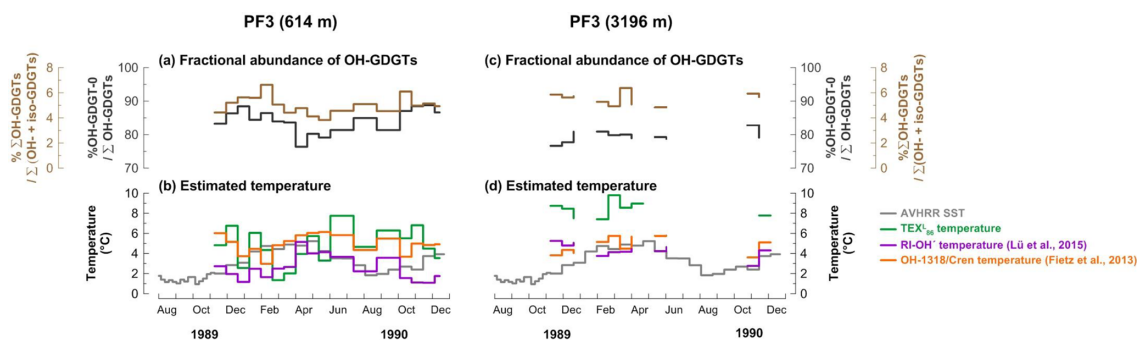
sink faster than opal-ballasted diatom aggregates (Iversen and Ploug, 2010). Still, fecal pellets formed from either coccolithophores or diatoms sink faster than fecal pellets formed from non-ballasted flagellates (Ploug et al., 2008a, b). GDGTs have been suggested to be preferentially incorporated into opal-dominated particles (Mollenhauer et al., 2015). This is in agreement with observed  $\text{TEX}_{86}$ -derived temperatures in sediment trap samples off Cape Blanc being delayed relative to the SST signal, and this time delay was longer than for  $\text{U}_{37}^K$ -derived temperatures (Mollenhauer et al., 2015).

GDGT fluxes covaried with fluxes of biogenic and non-biogenic components in the eastern Fram Strait (Fig. 3). GDGT fluxes were enhanced in summer 2007 and spring 2008 with two clear maxima in late September 2007 and March 2008 (Fig. 3b). Without those two distinct GDGT flux maxima, GDGT flux showed a good correlation with opal ( $R^2 = 0.82$ ,  $p < 0.0001$ ) throughout the deployment period. Those two episodic GDGT pulses occurred when carbonate, coccolithophore, and terrestrial biomarker fluxes were enhanced, potentially resulting in the enhanced GDGT flux (Chen et al., 2016; Yamamoto et al., 2012). GDGT fluxes also covaried with carbonate from the beginning of the deployment until April 2008 ( $R^2 = 0.8$ ,  $p < 0.0001$ ) (Fig. 3a and b). This agrees with the concept that GDGTs are transported by particles mainly containing opal and carbonate as previously shown in sediment trap studies (Chen et al., 2016; Huguet et al., 2007; Mollenhauer et al., 2015; Park et al., 2018; Yamamoto et al., 2012). This observation suggests that GDGTs are exported together with diatoms (opal) and coccolithophores (carbonate). Supporting this, diatom and coccolithophore fluxes both, previously reported by Lalande et al. (2016), showed good correlations with GDGT fluxes ( $R^2 = 0.64$ ,  $p < 0.001$  for diatoms and  $R^2 = 0.68$ ,  $p < 0.0001$  for coccolithophores) when excluding two GDGT flux maxima in the correlations with opal fluxes. The flux of terrestrial biomarkers (campesterol +  $\beta$ -sitosterol), previously analyzed by Lalande et al. (2016), also showed a positive correlation with GDGT flux ( $R^2 = 0.64$ ,  $p < 0.001$ ). The sterols (campesterol and  $\beta$ -sitosterol), used frequently to assess the plant-derived organic matter input to aquatic systems (Moreau et al., 2002), reflect the input of terrestrial matter transported by sea ice in the Fram Strait (Lalande et al., 2016). We thus assume that terrestrial material is aggregated into particles which carry GDGTs. However, the lithogenic flux, also representing terrestrial input, displayed a different trend with maxima only in mid-April–mid-May 2008, when the sea ice concentration abruptly increased (Fig. 3e). The lithogenic material seems to be mainly supplied by downslope export from the nearby Svalbard archipelago (Lalande et al., 2016), with a significant input when the sea ice is present (Fig. 3e).

GDGTs might also be exported with fecal pellets after grazing of Archaea by zooplankton. Indeed, GDGTs have been found in decapod guts and intestines, and their



**Figure 5.** Changes in mass flux (a, d), GDGT flux (b, e), and TEX<sub>86</sub><sup>L</sup>-derived temperatures and satellite-derived SSTs obtained at 50.125° S, 5.875° E near the trap site (c, f) at the 614 m (a, b, c) and 3196 m (d, e, f) water depths at the Antarctic Polar Front site (PF3) during the deployment period (November 1989–December 1990).



**Figure 6.** Changes in fractional abundance of OH-GDGTs (a, c) and estimated temperatures based on OH-related calibrations (b, d) at site PF3. The purple and orange lines indicate the calculated temperatures using the calibrations by Fietz et al. (2013) (Eq. 4) and Lü et al. (2015) (Eq. 6), respectively. The green lines indicate the TEX<sub>86</sub><sup>L</sup>-derived temperatures.

abundance ratios appear to be unaltered during gut passage (Huguet et al., 2006). The fecal pellet carbon (FPC) flux from appendicularians contributed more to the total carbon flux than copepod FPC flux. Both appendicularian and copepod FPC fluxes were higher during spring 2008 than during summer 2007 (Fig. 3d; Lalande et al., 2016). The correlation of GDGT fluxes with appendicularian FPC fluxes ( $R^2 = 0.67$ ,  $p < 0.05$  in mid-July to mid-October 2007) implies grazing of Archaea by the appendicularians. Appendicularians are well known to ingest micro-size particles like Archaea ( $< 1 \mu\text{m}$ ) efficiently through a fine mucus filter (Conley and Sutherland, 2017).

Overall, it is reasonable to assume that seasonal changes in the relative proportion of materials that GDGTs can be aggregated with may result in variable export velocities of particles carrying GDGTs to deeper waters in the eastern Fram Strait.

### 5.1.2 Variability of TEX<sub>86</sub><sup>L</sup>-derived temperature

TEX<sub>86</sub><sup>L</sup> temperatures varied ( $-0.2$  to  $3.7^\circ\text{C}$ ) within a similar range as the satellite-derived SSTs ( $-0.1$  to  $3.4^\circ\text{C}$ ) during the trap deployment period in the eastern Fram Strait. Although it did not display a clear seasonality, the TEX<sub>86</sub><sup>L</sup> signal is most likely to reflect the surface water environments without nonthermal effects influencing the TEX<sub>86</sub><sup>L</sup> (Fig. 3e). First of all, when the error of the TEX<sub>86</sub><sup>L</sup> calibration ( $\pm 4^\circ\text{C}$ ) is considered, the SST estimates are identical to the satellite-derived SSTs. Secondly, the index values (e.g., BIT, MI, %GDGT-0, RI), which could indicate potential nonthermal factors influencing the distribution of GDGTs, suggest that TEX<sub>86</sub><sup>L</sup> temperatures should reflect the upper water column temperature (Table A1). The BIT index defined by Hopmans et al. (2004) as a tracer for the terrestrial organic matter input was very low ( $< 0.02$ ), showing that negligible amounts of soil-derived GDGTs are entrained in sinking particles.

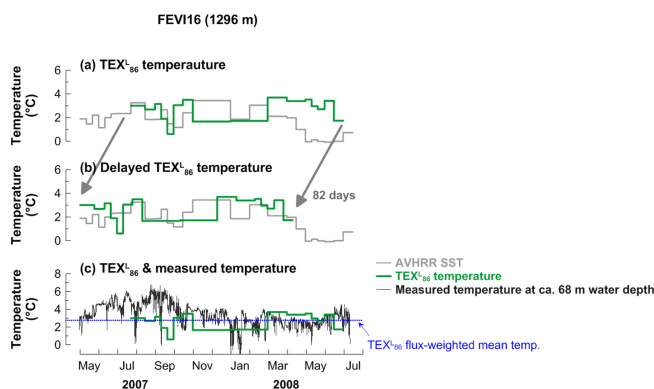
MI (Zhang et al., 2011) and %GDGT-0 (Inglis et al., 2015) have been suggested as indicators for the impact of methanotrophic Archaea even though it was questioned if the latter index can be applied in marine settings (Inglis et al., 2015). Both index values were consistently lower than the respective critical values of 0.3 for MI and 67 % for %GDGT-0.  $|\Delta RI|$  was suggested by Zhang et al. (2016) as an indicator for the integrated nonthermal factors on  $TEX_{86}^L$ . Most  $|\Delta RI|$  values were lower than a value of 0.3, and only three samples collected in late September 2007 and June 2008 satisfied the criteria of being influenced by nonthermal factors. Lastly, although significant portions of particles transported to the mid-depth and deep water in the eastern Fram Strait originated off Svalbard and the Barents Sea (Lalande et al., 2016), it is known that GDGTs are most likely to reflect the local conditions rather than being affected by lateral transport (Kim et al., 2009; Mollenhauer et al., 2008). Therefore, we conclude that GDGTs are mainly transported from upper waters and non-regional sources of particles do not play a significant role in the  $TEX_{86}^L$  estimate of sinking particles of the eastern Fram Strait.

$TEX_{86}^L$  temperatures varied strongly between mid-July and October 2007, with the minimum estimated temperature ( $0.6^\circ\text{C}$ ) when the GDGT and terrestrial biomarker fluxes were highest (Fig. 3b, c, and f). This is the most productive period in the Fram Strait and also the period with the highest export fluxes (e.g., Lalande et al., 2016). The material collected by deep-ocean sediment traps is a mixture of many types of aggregates with different composition and settling velocities. Therefore, fluctuating  $TEX_{86}^L$  temperatures might be due to an average of signals from previous (before mid-July, i.e., slowly sinking particles) and current seasons (mid-July to October, i.e., fast settling aggregates), which were exported via different aggregation and sinking mechanisms with different horizontal displacements during particle descent. Due to the collection of aggregates sinking with different velocities, one sample cup in a sediment trap may collect GDGTs of different ages.

During the low flux period between November 2007 and February 2008,  $TEX_{86}^L$  temperatures were constantly lower than the satellite-derived SSTs (Fig. 3a, b, and f). These signals might be derived from GDGTs synthesized in mid-August to October, which was a late-bloom period dominated by protists that do not produce biominerals, such as *Phaeocystis* sp., dinoflagellates, and nanoflagellates (Nöthig et al., 2015). Aggregates formed without biominerals have low sinking velocities (Iversen and Ploug, 2010; Ploug et al., 2008a, b), which may explain why the satellite-derived SSTs from mid-August to October 2007 were similar to the  $TEX_{86}^L$ -derived temperatures in sinking particles collected during November 2007 and February 2008. In spring 2008,  $TEX_{86}^L$  temperatures stayed relatively warm until mid-April and suddenly dropped in May and June (Fig. 3f). The relatively warm  $TEX_{86}^L$  temperatures in particles collected in March and April 2008 probably reflect the signal transported

by GDGTs produced in November–December 2007. Warm and cold biases of the  $TEX_{86}^L$ -derived temperatures varied within the calibration error ( $\pm 4^\circ\text{C}$ ) throughout the trap deployment period. It shows that the bias of the calibration occurs neither in one direction only nor to the same extent even at a given location; instead the temperature estimate is more affected by other processes discussed above. Like previous seasons, the lack of transporting materials for GDGTs delayed the  $TEX_{86}^L$  signal. The sudden drop in  $TEX_{86}^L$ -derived temperatures in mid-May to June 2008 was matched with the season of the enhanced flux of biogenic materials, GDGTs, phytoplankton, zooplankton fecal pellets, and terrestrial biomarkers at the trap when the sea ice concentration was enhanced (Fig. 3). Eddies around the ice edge create upwelling and downwelling, which breaks the stratified surface water and supplies nutrients to the upper water column, fostering phytoplankton blooms (Lalande et al., 2013). The sea ice-edge bloom probably enhanced the GDGT production followed by zooplankton grazing. Additionally, the terrestrial materials as ballast were more available, and were derived from sea ice as it melted during spring 2008. This process caused the fast export of  $TEX_{86}^L$  signal from the surface waters in mid-May to June 2008. Overall, the particles containing GDGTs captured at 1296 m depth in the eastern Fram Strait were likely delivered by a combination of packaging mechanisms involving biological and nonbiological transport materials with different delay times over the season. Moreover, most GDGTs are exported vertically from the upper waters and reflect a regional  $TEX_{86}^L$  signal in the eastern Fram Strait (Fig. 3f), while significant particle supply from the south via lateral advection has been reported previously (Lalande et al., 2016). The reflection of SSTs based on the  $TEX_{86}^L$  calibration and the good correlation of GDGTs with opal and Appendicularian fecal pellet fluxes agrees with the finding that diatom and fecal pellet fluxes can be traced as the export of local production regardless of the lateral particle supply in the Fram Strait (Lalande et al., 2016).

The average export velocity of particles containing GDGTs can be calculated by dividing the travel distance of particles (i.e., the depth of the sediment trap) by the temporal offset between  $TEX_{86}^L$ -derived temperature and the corresponding satellite-derived SST, greatly simplifying the complexity of sinking mechanisms and range of settling velocities throughout the season (Mollenhauer et al., 2015). When the  $TEX_{86}^L$ -derived temperatures are shifted by 82 d, we found the best fit to the satellite-derived SSTs (Fig. 7b). Therefore, we see that GDGTs synthesized in the eastern Fram Strait likely travel to the trap (1296 m) approximately within 82 d. This translates into an average minimum export velocity of GDGT-containing particles of approximately  $15\text{ m d}^{-1}$  when assuming export from 30 to 80 m water depth (see Sect. 5.1.3). We have assumed that the export velocity includes the time of GDGT synthesis and transport time to the trap. The estimated sinking velocity of  $15\text{ m d}^{-1}$  is within the range ( $9\text{--}17\text{ m d}^{-1}$ ) estimated at a similar water depth



**Figure 7.** Changes in  $\text{TEX}_{86}^L$ -derived temperatures and satellite-derived SSTs (a), time-delayed  $\text{TEX}_{86}^L$  temperatures (b), and sensor-measured temperatures (c) at approximately 68 m water depth at site FEVI16.

( $\sim 1300$  m) in the filamentous upwelling zone off northwest Africa (Mollenhauer et al., 2015) and only slightly lower than settling velocities found previously for aggregates from the Fram Strait (Wekerle et al., 2018). Based on the seasonal succession of peaks in appendicularian fecal pellets in the same mooring system, their sinking velocity was estimated to be 5- to 11-fold higher (15–35 d to 2400 m depth; Lalande et al., 2016) compared to GDGTs. This is a realistic scenario because of the fast sinking velocity of a fecal pellet, being rather big and dense, and generally faster average export velocities to deeper ocean depths (Fischer and Karakaş, 2009; Iversen et al., 2017). Moreover, the average export velocity of GDGTs includes the time it takes to be incorporated into aggregates. A more rapid export of GDGTs to depth resulting in smaller temporal offsets of  $\text{TEX}_{86}^L$ -derived temperature to water temperature changes occurred in mid-May–June 2008 when the FPC flux of appendicularians was enhanced (Fig. 3d). It has to be noted that the export velocity we calculated here may represent an averaged velocity of all the aggregates collected in the trap, as the export velocity of GDGTs can vary depending on the type of sinking materials GDGTs are incorporated into.

The flux-weighted mean  $\text{TEX}_{86}^L$  temperature over the mooring period was  $2.8^\circ\text{C}$ . In the vicinity of the site FEVI16 ( $79.03^\circ\text{N}$ ,  $4.35^\circ\text{E}$ ), two  $\text{TEX}_{86}^L$  values from surface sediments are available in the dataset published by Ho et al. (2014). The estimated temperatures based on  $\text{TEX}_{86}^L$  are  $2.8$  and  $2.3^\circ\text{C}$  in PS68-251/2 ( $79.30^\circ\text{N}$ ,  $4.30^\circ\text{E}$ ) and PS68-271/2 ( $79.10^\circ\text{N}$ ,  $4.60^\circ\text{E}$ ), respectively. Considering the error of the  $\text{TEX}_{86}^L$  calibration (Kim et al., 2010),  $\text{TEX}_{86}^L$  estimates are almost identical between sinking particles and surface sediments. This suggests that GDGTs synthesized in the upper waters are propagated through the water column into the sediment without significant alteration even though the sinking process of GDGTs in the eastern Fram Strait seems quite complicated as discussed above. Iversen et al. (2010) and

Jackson and Checkley Jr. (2011) also suggested that most biological activities resulting in aggregate alteration and degradation occur around the base of the photic zone. Our observations agree with previous studies, as  $\text{TEX}_{86}$  displays consistent values at multiple depths and/or in the underlying surface sediments in various environmental regimes such as in the Arabian Sea (Wuchter et al., 2006b), in the northwestern Pacific (Yamamoto et al., 2012), off Cape Blanc (Mollenhauer et al., 2015), near Iceland (Rodrigo-Gámiz et al., 2015), off southern Java (Chen et al., 2016), and in the northern Gulf of Mexico (Richey and Tierney, 2016).

In the eastern Fram Strait, the changes in  $\text{TEX}_{86}^L$ -derived temperature are largely controlled by the depth and time of GDGT production and temporally variable sinking materials aggregated with GDGTs. Additionally, the absolute estimated temperatures varied within the  $\text{TEX}_{86}^L$  calibration error ( $\pm 4^\circ\text{C}$ ). In this case, the temperature error inherited from the calibration is less important than other relative changes.

### 5.1.3 Potential depth of $\text{TEX}_{86}^L$ signal origin

To determine the water depth where the  $\text{TEX}_{86}^L$  signal originated, the flux-weighted mean  $\text{TEX}_{86}^L$  temperature ( $2.8^\circ\text{C}$ ) was compared to the depth profile of nutrient concentrations ( $\text{NH}_4^+$ ,  $\text{NO}_2^-$ , and  $\text{NO}_3^-$ ) measured in late June to early July 2010, 2011, and 2013 and water temperature extracted from WOA13 ( $79.125^\circ\text{N}$ ,  $4.375^\circ\text{E}$ ) in the eastern Fram Strait (Fig. 8). Unfortunately, ammonia data are unavailable in 2007 and 2008. The mean estimated temperature ( $2.8^\circ\text{C}$ ) in sinking particles corresponds to the surface water in July–September or of 30–80 m subsurface waters in April–June (Fig. 8). Major production of GDGTs in both seasons is a possible scenario because GDGT flux peaked in both seasons (Fig. 3b). However, the latter period (April–June) is the more plausible season dominantly supplying GDGTs to the sediment if we consider the delay time of the GDGT signal (approximately 82 d; see Sect. 5.1.2), which illustrates the initial time of GDGT production (Fig. 3b). Furthermore, Thaumarchaeota, the primary synthesizers of GDGTs in the ocean, are aerobic ammonia oxidizers (Könneke et al., 2005) and maximum GDGT concentrations have been found near the  $\text{NO}_2^-$  maximum, where maximum rates of ammonia oxidation and nitrification occur (Beman et al., 2008; Hurley et al., 2016). The depths of consumption of ammonia and production of nitrite as well as nitrate accumulation as a result of the nitrification process in the subsurface can be deduced from the nutrient profile (Fig. 8). Assuming that maximum thaumarchaeotal abundance occurs at the depth of highest substrate availability, we, therefore, infer that Thaumarchaeota mainly record the subsurface water temperature (30–80 m) during the warm season when the spring bloom may occur (April–June) in the eastern Fram Strait. Similar observations were made in the Sea of Okhotsk and the northwest Pacific (Seki et al., 2014) and in the western Bering Sea (Meyer et al., 2016), where the  $\text{TEX}_{86}^L$ -derived temperature is attributable

to the regional season and water depth of GDGT production. The temperatures measured by a temperature sensor attached to the mooring array at approximately 68 m depth (Beszczynska-Möller et al., 2012a), which is approximately in the middle of the depth interval (30–80 m) corresponding to the  $\text{TEX}_{86}^L$  temperature (Fig. 8), support our interpretation. The flux-weighted mean  $\text{TEX}_{86}^L$  temperature is within the range of measured temperatures at this depth and it reflects well the temperatures in March–June as deduced from the nutrient depth profile well (Fig. 7c).

## 5.2 Antarctic Polar Front (50° S)

### 5.2.1 GDGT flux

At the shallow trap (614 m water depth) of site PF3, there was no clear correlation between fluxes of organic matter and GDGTs, which displayed their respective maxima in different seasons (Fig. 5a and b). Peaks of mass flux occurred in November 1989–early March 1990 and mid-October–December 1990, while GDGT fluxes were highest in mid-May–June and late July–August 1990 (late austral autumn and winter) (Fig. 5a and b). Similar observations were made in the North Sea, where the abundance of Thaumarchaeota (previously known as marine group I Crenarchaeota) and GDGT concentrations were high in wintertime, supported by the seasonality of Thaumarchaeotal 16S rRNA and *amoA* gene abundances in that region (Herfort et al., 2007; Pitcher et al., 2011; Wuchter et al., 2006a). An austral “winter bloom” of planktonic Archaea was also found near the Antarctic Peninsula (Church et al., 2003; Murray et al., 1999; Tolar et al., 2016). Photoautotrophic phytoplankton, which use ammonium as a N source, would outcompete Archaea for ammonia in spring and summer. In contrast, in wintertime when phytoplankton productivity is limited due to the lack of light, ammonia availability for Archaea is higher (Pitcher et al., 2011; Wuchter et al., 2006b). This explains the winter bloom of ammonia-oxidizing Archaea. Laubscher et al. (1993) found that ammonia was highly depleted at the chlorophyll-*a* maximum across the Antarctic Polar Front in early austral summer, which might create less favorable conditions for ammonia-oxidizing planktonic Archaea. Additionally, Thaumarchaeota are known to be sensitive to photoinhibition (Horak et al., 2017). Therefore, the austral winter maxima in GDGT flux at the shallow trap of PF3 might be a consequence of the higher production of planktonic Archaea in surface waters of the APF during the time when ammonia is more available and photoautotrophs cannot compete due to light limitation.

At the deep trap (3196 m water depth) of site PF3, GDGT flux peaked in November 1989 while mass flux was most pronounced in March 1990 (Fig. 5d and e). Due to the lack of sinking particles captured in the deep trap in June–November 1990, it is unclear if a potential winter bloom of GDGTs was also exported to deeper waters. The trapping

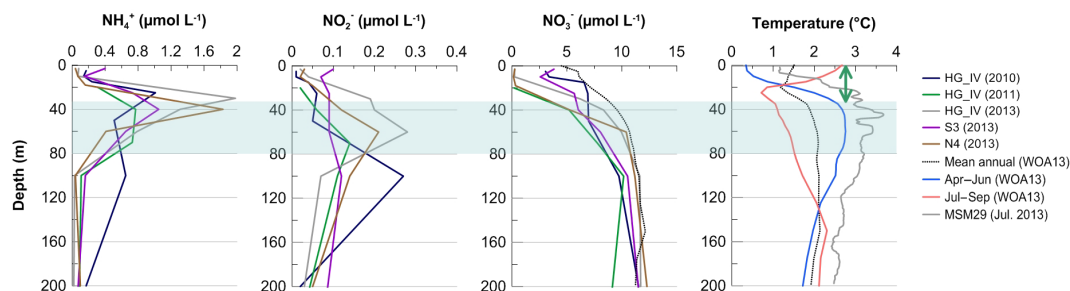
efficiency of PF3 was found to be below 50 % at the deep trap using  $^{230}\text{Th}$  as flux proxy (Walter et al., 2001). The negligible mass flux could thus be caused by low trapping efficiency. However, Fischer et al. (2002) observed a similar seasonal flux pattern in the following years measured almost at the same depths and at the same location as site PF3. At site BO (54.50° S, 3.33° W), which was located further south than site PF3, the authors also found a period of almost no flux in July–December for 4 years approximately at 2200 m depth. Thus, it is assumed that a low trapping efficiency or a failure of the trap system did not account for the lack of particle samples at the deep trap of PF3.

### 5.2.2 Variability of $\text{TEX}_{86}^L$ -derived temperature

$\text{TEX}_{86}^L$ -derived temperatures at the shallow trap did not display a clear seasonal variability. Only one-third of the data points were similar to the SSTs, while the remaining samples showed warm- or cold-biased temperatures relative to the satellite-derived SSTs during the sampling period (Fig. 5c). All samples in the deep trap displayed warmer  $\text{TEX}_{86}^L$  temperatures by up to 7 °C relative to the satellite-derived SSTs (Fig. 5f). As discussed above, the mismatch between lipid proxy-based temperature estimates and satellite-derived temperature could be explained by the delay time of the proxy signal, due to the time needed between lipid synthesis and incorporation into sinking aggregates plus the sinking time (Mollenhauer et al., 2015; Müller and Fischer, 2003; Park et al., 2018). However, without a marked seasonality of the estimated temperature, it is difficult to determine a delay time of the  $\text{TEX}_{86}$  signal. The other explanation for the absence of covariance between the  $\text{TEX}_{86}$  signal and the satellite-derived SST is that we observe a mixed signal of Thaumarchaeotal GDGTs derived from surface and deep ocean. The warm biases have a tendency to occur during periods of lower GDGT flux and thus may include higher proportions of material from different sources (early December, mid-June–July, September–November), which may be more dominant in the deep trap (see discussion below).

Temperature residuals ( $\sim 7^\circ\text{C}$ ) in the deep trap, which are larger than the calibration error ( $\pm 4^\circ\text{C}$ ), suggest significant nonthermal effects on GDGT compositions or the unreliability of the global calibration in this region.

Warm-biased  $\text{TEX}_{86}^L$ -derived temperatures have been attributed to the contribution of different archaeal communities or GDGT input from terrestrial sources, which may alter the composition of pelagic GDGTs, leading to unusual  $\text{TEX}_{86}^L$ -derived temperature estimates (Hopmans et al., 2004; Inglis et al., 2015; Zhang et al., 2011, 2016). For example, several studies showed that anomalous  $\text{TEX}_{86}$  based temperature estimates can be caused by isoprenoid GDGTs produced by Group II Euryarchaeota as significant contributors to the archaeal tetraether lipid pool and, thus,  $\text{TEX}_{86}$  (Lincoln et al., 2014; Schouten et al., 2008, 2014; Turich et al., 2007), a suggestion that is controversial. A prevalence of ma-



**Figure 8.** Depth profiles of nutrient concentration ( $\text{NH}_4^+$ ,  $\text{NO}_2^-$ ,  $\text{NO}_3^-$ ) and temperature in the Fram Strait. The navy, green, and gray lines represent the central station of the Long-Term Ecological Research (LTER) observatory HAUSGARTEN (HG-IV), close to the FEVI16 trap location. The purple and brown lines represent the southern and northern stations of HG-IV. Black dotted lines for  $\text{NO}_3^-$  and temperature and blue and orange lines for temperature represent the datasets extracted from WOA13. Only two seasonal profiles (April to June and July to September) from WOA13 are available at the same location ( $79.125^\circ \text{N}$ ,  $4.375^\circ \text{E}$ ) near the site FEVI16. Measured nutrient concentrations are reported on PANGAEA (Bauerfeind et al., 2014).

rine Group II Euryarchaeota has been reported in Circumpolar Deep Water (Alonso-Sáez et al., 2011) and in deep waters of the Antarctic Polar Front (López-García et al., 2001; Martin-Cuadrado et al., 2008; Moreira et al., 2004; Murray et al., 1999). Therefore, we would assume that GDGTs produced by deep-dwelling Euryarchaeota might have caused the warm-biased  $\text{TEX}_{86}^{\text{L}}$  signal at depth in the region. Alternatively, advection of particles from warmer ocean regions could potentially lead to warm biases. However, it is known that the impact of lateral transport of GDGTs on the local signal is insignificant (Kim et al., 2009; Mollenhauer et al., 2008).

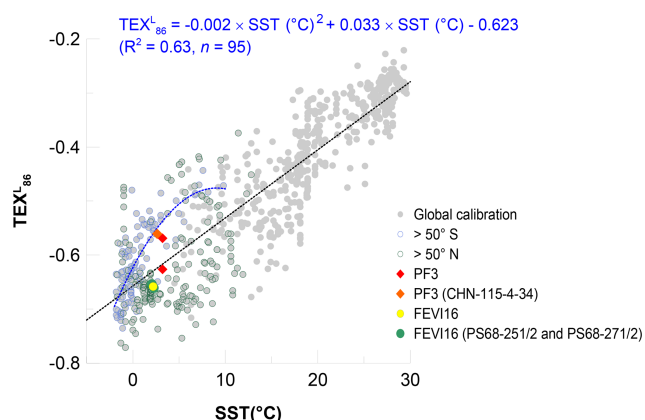
BIT, MI, %GDGT-0, and  $|\Delta \text{RI}|$  were examined to evaluate nonthermal factors on GDGT composition and  $\text{TEX}_{86}$  values at both PF3 traps. However, none of the values for BIT, %GDGT-0, and  $|\Delta \text{RI}|$  exceeded the defined critical values (Table A1). MI values were higher at the deep trap than at the shallow one. The first two samples at the deep trap reached values of 0.3–0.5 MI, suggesting that GDGTs are derived from a mixture of non-methanotrophic and methanotrophic communities (Zhang et al., 2011). However, these two samples alone cannot fully explain the episodic and continuous warm-biased  $\text{TEX}_{86}^{\text{L}}$  estimates at the shallow and deep traps. Therefore, those indices cannot explain the warm anomaly of  $\text{TEX}_{86}^{\text{L}}$  estimates in the APF. Like in the Gulf of Mexico (Richey and Tierney, 2016), a longer time series of samples would be helpful to investigate inter-annual variability, paired with a direct assessment of the archaeal community in the region.

The flux-weighted mean  $\text{TEX}_{86}^{\text{L}}$  temperatures averaged over the trap deployment period were 4.6 and 8.5 °C at the shallow and deep traps, respectively. The  $\text{TEX}_{86}^{\text{L}}$  temperature in surface sediment (CHN 115-4-34;  $51.00^\circ \text{S}$ ,  $5.33^\circ \text{E}$ ) from the vicinity of the PF3 trap was 9.1 °C, which is 4.5 °C warmer than the  $\text{TEX}_{86}^{\text{L}}$  estimate temperature in the shallow trap (4.6 °C) but is similar to the one in the deep trap (8.5 °C). This situation is different from the Fram Strait and

other time-series trap studies which showed that  $\text{TEX}_{86}$  values in different depths are almost identical (Chen et al., 2016; Mollenhauer et al., 2015; Wuchter et al., 2006b). This implies that there is a clearly different origin of GDGTs in particles collected in the upper ( $< \sim 600 \text{ m}$ ) and deeper ocean, yet the main explanation of the warm-biased  $\text{TEX}_{86}^{\text{L}}$  is still speculative.

### 5.3 Re-evaluation of $\text{TEX}_{86}^{\text{L}}$ calibration in polar oceans

In the two previous sections, the  $\text{TEX}_{86}^{\text{L}}$  calibration developed by Kim et al. (2010) for regions where maSSTs are below 15 °C was applied. This calibration is based on data from 396 surface sediments in the global ocean. Several studies have expanded the  $\text{TEX}_{86}$  and  $\text{TEX}_{86}^{\text{L}}$  surface sediment dataset. We revisited the latest global surface sediment dataset containing 1095 surface sediment measurements (Tierney and Tingley, 2015, and references therein) to recalculate the  $\text{TEX}_{86}^{\text{L}}$  calibration and to examine the agreement of our results with the new calibration. From the dataset, the data points for which  $\text{TEX}_{86}^{\text{L}}$  values cannot be calculated or which were pointed out as problematic by Ho et al. (2014) (see  $\text{BIT} > 0.3$  or  $\sim 1.0$  and GDGT concentration below detection limit) were excluded to avoid potential biases. With approximately 2 times more data points for the  $\text{TEX}_{86}^{\text{L}}$  calibration than the original one (Kim et al., 2010;  $n = 396$ ), the new calibration again shows a linear correlation with maSSTs even though the correlation coefficient is slightly lower (Fig. 9;  $\text{TEX}_{86}^{\text{L}} = 0.013 \times \text{SST} - 0.657$ ;  $R^2 = 0.80$ ,  $n = 744$ ) and the residual standard error ( $\pm 5.0^\circ \text{C}$ ) is higher than those of the calibration of Kim et al. (2010) ( $R^2 = 0.86$ ,  $\pm 4.0^\circ \text{C}$ ). It should be noted that our new linear calibration has SST on the  $x$  axis as the control variable and the  $\text{TEX}_{86}^{\text{L}}$  value on the  $y$  axis as the dependent variable because the composition of GDGTs and membrane lipids of Thaumarchaeota (i.e.,  $\text{TEX}_{86}/\text{TEX}_{86}^{\text{L}}$  value) is affected by water temperature. This different regression might explain parts of the discrepancy between our and the original cali-



**Figure 9.** Correlation of  $\text{TEX}_{86}^L$  values with mean annual SSTs in the global ocean (gray symbols; Tierney and Tingley (2015) and reference therein). Blue and black round symbols represent the high-latitude regions higher than  $50^\circ\text{S}$  and  $50^\circ\text{N}$ , respectively. In the southern regions ( $> 50^\circ\text{S}$ ),  $\text{TEX}_{86}^L$  values have a polynomial correlation with maSSTs ( $R^2 = 0.63$ ,  $n = 95$ ). Red and orange diamonds represent the flux-weighted mean  $\text{TEX}_{86}^L$  of the two traps at the site PF3 and  $\text{TEX}_{86}^L$  on the surface sediment (CHN-115-4-34), respectively. Yellow and green round symbols represent the flux-weighted mean  $\text{TEX}_{86}^L$  at the site FEV116 and  $\text{TEX}_{86}^L$  on the surface sediments (PS68-251/2 and PS68-271/2), respectively.

brations of Kim et al. (2010). Larger scatter towards colder temperatures appears in the new calibration as shown in previous studies (Ho et al., 2014; Kim et al., 2010) (Fig. 9).

It is obvious that the relationship of maSSTs and  $\text{TEX}_{86}^L$  values in high-latitude northern ( $> 50^\circ\text{N}$ ) and southern ( $> 50^\circ\text{S}$ ) oceans is significantly different when both regions are plotted separately (Fig. 9). The data from the north ( $> 50^\circ\text{N}$ ) are scattered on both the right- and left-hand sides of the regression line, resulting in potential warm and cold anomalies, respectively (Fig. 9). This may reflect various sources of GDGTs in the high-latitude northern oceans, which have direct geographical connections to Eurasia and North America. Warm-biased  $\text{TEX}_{86}^L$  estimates in these regions might be caused by input of soil-derived GDGTs, which are picked up by the sea ice in the Arctic marginal seas (Laptev Sea, Kara Sea, eastern Greenland), and released while the sea ice melts. Alternatively,  $\text{TEX}_{86}^L$ -derived temperatures might reflect warmer subsurface temperatures in the warm season as shown in the subarctic North Pacific region with the regional calibration by Seki et al. (2014) and Meyer et al. (2016). Cold-biased  $\text{TEX}_{86}^L$  estimates are mainly found in the Barents Sea, North Sea, and the Norwegian Sea. In the North Sea, a significantly enhanced abundance of planktonic Archaeal cells and high GDGT concentration in the winter-time could account for colder  $\text{TEX}_{86}^L$ -derived temperature compared to SST (Herfort et al., 2007; Pitcher et al., 2011; Wuchter et al., 2006a). Hence, regional calibrations in the high-latitude northern oceans seem to be a valid approach.

By contrast, most of the data from the south ( $> 50^\circ\text{S}$ ) show higher  $\text{TEX}_{86}^L$  values than predicted by the regression line ( $n = 95$ ). This explains the observed warm anomalies of  $\text{TEX}_{86}^L$ -derived temperature when using the original linear regression calibration. Instead of the global linear calibration, a polynomial one seems to be a better option in these regions (Fig. 9). When excluding one data point in the winter sea-ice-covered Southern Ocean, the correlation coefficient of the polynomial correlation is encouraging ( $R^2 = 0.63$ ).

The data points of the flux-weighted mean  $\text{TEX}_{86}^L$  in the FEV116 trap and  $\text{TEX}_{86}^L$  in the underlying surface sediment against maSSTs are both closely located to the linear regression line. It illustrates the applicability of the linear  $\text{TEX}_{86}^L$  calibration in the eastern Fram Strait (Fig. 9). At the site PF3, the  $\text{TEX}_{86}^L$ -derived temperatures based on the polynomial calibration yield  $-0.1$ ,  $1.9$ , and  $2.2^\circ\text{C}$  at the shallow and deep traps and in the underlying surface sediment, respectively (Fig. 9), which are colder than the estimates based on the linear calibration. The polynomial  $\text{TEX}_{86}^L$  temperatures at the deep trap and on the sediment are slightly colder than maSST ( $2.4^\circ\text{C}$ ; the data can be found in Tierney and Tingley, 2015), but less biased compared to the reconstructed temperatures based on the linear  $\text{TEX}_{86}^L$  calibration (see Sect. 5.2.2). To test this polynomial calibration in the Southern Ocean ( $> 50^\circ\text{S}$ ), a further assessment needs to be made with down-core sediments in similar regions, where the original  $\text{TEX}_{86}^L$  estimate displays a warm bias.

#### 5.4 Applicability of OH-GDGT-related calibrations for SST estimates in the high-latitude Atlantic Ocean

OH-GDGTs were determined in sinking particles of the eastern Fram Strait and of the APF. The proportions of OH-GDGTs to total GDGTs (sum of OH- and iso-GDGTs) ranged between 7 % and 14 % in the eastern Fram Strait and 4 % and 7 % in the APF (Figs. 4a, 6a, and c). This is a relatively smaller portion than in the Nordic Seas ( $\sim 16\%$ ; Fietz et al., 2013).

Several OH-GDGT-based calibrations for SST have been suggested for the global ocean and the Nordic Seas (Fietz et al., 2013; Huguet et al., 2013; Lü et al., 2015). We applied those calibrations suggested by Fietz et al. (2013) and Lü et al. (2015). Only two calibrations (Eqs. 4 and 6) yield realistic temperature estimates, which varied within a similar range as satellite-derived SSTs or  $\text{TEX}_{86}^L$ -derived temperatures (Figs. 4b, 6b and d). The former calibration considers relative abundances of OH-GDGT-0 and crenarchaeol (Eq. 4). The latter is based on the RI-OH-GDGTs (Eqs. 5 and 6). In the eastern Fram Strait, both OH-GDGT-based temperatures showed similar changes with an increasing trend until late April 2008 and decreasing temperatures in May 2008 except for the last two samples (Fig. 4b). In the APF, both OH-GDGT-based temperatures were also close to the satellite-derived SSTs and the  $\text{TEX}_{86}^L$ -derived temperatures at the shallow trap (Fig. 6b). Warm biases such as the  $\text{TEX}_{86}^L$  cal-

**Table 2.** Summary of TEX<sub>86</sub><sup>L</sup> thermometry in FEVI16 and PF3 site.

Trap name	FEVI16	PF3
Oceanographic setting	Seasonal ice cover	Winter ice edge
Main GDGTs producer	Thaumarchaeota	Thaumarchaeota + Euryarchaeota
Surface ocean temperature	<sup>a</sup> Satellite SST: −0.1 to 3.4 °C <sup>b</sup> Ave. SST: 1.9 °C	<sup>a</sup> Satellite-SST: 1.8 to 5.2 °C <sup>b</sup> Ave. SST: 3.5 °C
Shallow trap	<sup>c</sup> TEX <sub>86</sub> <sup>L</sup> <i>T</i> : 2.8 °C (30–80 m depth signal)	<sup>c</sup> TEX <sub>86</sub> <sup>L</sup> <i>T</i> : 4.6 °C (Thaumarchaeota + Euryarchaeota)
Deep trap		<sup>c</sup> TEX <sub>86</sub> <sup>L</sup> <i>T</i> : 8.5 °C (dominant Euryarchaeota)
Surface sediment	TEX <sub>86</sub> <sup>L</sup> <i>T</i> : 2.3/2.8 °C at 2400 m	TEX <sub>86</sub> <sup>L</sup> <i>T</i> : 9.1 °C at 3800 m
Relevant processes for GDGTs	– Export of upper ocean signal by fast settling particles – Highly ballasted with opal and carbonate <sup>d</sup> s.v.: 15 m d <sup>−1</sup>	– Contribution of Euryarchaeota in °CDW causing warm biases
Conclusions	– Linear calibration (TEX <sub>86</sub> <sup>L</sup> ) applicable – Temporal offset due to changing ballast materials and s.v. – OH-GDGT-based calibrations applicable	– Linear calibration (TEX <sub>86</sub> <sup>L</sup> ) unreliable – Nonlinear relationship between TEX <sub>86</sub> <sup>L</sup> and SST (> 50° N) – OH-GDGT-based calibrations applicable

<sup>a</sup> Satellite-derived sea surface temperature. <sup>b</sup> Averaged surface temperature over the trap deployment period. <sup>c</sup> Flux-weighted average temperature over the trap deployment period. <sup>d</sup> s.v.: sinking velocity. <sup>e</sup> CDW: Circumpolar Deep Water.

ibration did not occur with those OH-GDGT calibrations at the deep trap (Fig. 6d). We speculated in Sect. 5.2.2 that the warm-biased TEX<sub>86</sub><sup>L</sup> temperatures relative to SSTs may be caused by GDGTs synthesized by Euryarchaeota dwelling in deep waters of the APF. The methanogenic Euryarchaeota are known to produce GDGT-1, GDGT-2, and GDGT-3 and may alter the TEX<sub>86</sub> signal derived from pelagic Thaumarchaeota (Weijers et al., 2011). This might explain why only calibrations (OH-GDGT/Cren and RI-OH'), which do not contain GDGTs potentially originating from Euryarchaeota, show a correspondence to regional SSTs. At this stage, however, this has to be considered speculative. Moreover, it is not clear yet if OH-GDGTs are exclusively produced by Thaumarchaeota since OH-GDGTs were also detected in a culture of Euryarchaeota (*Methanothermococcus thermolithotrophicus*) (Liu et al., 2012). Therefore, further research is needed to clarify various aspects of distribution, production, and modification of OH-GDGTs in response to physicochemical changes in the global ocean. Nonetheless, OH-GDGTs appear to be a potential temperature proxy in our two high-latitude regions.

## 6 Summary and conclusions

Sinking particles collected using time-series sediment traps allowed us to determine the variability of the downward

GDGT export and the environmental influence on TEX<sub>86</sub><sup>L</sup> thermometry in northern and southern high-latitude regions of the Atlantic Ocean. We observed fundamentally different patterns between the eastern Fram Strait and the Antarctic Polar Front (Table 2).

In the eastern Fram Strait, the seasonally different composition of sinking materials resulted in different sinking velocities of GDGTs, and thus the temporal offsets between TEX<sub>86</sub><sup>L</sup>-derived temperatures and SSTs may vary by season. Although increased flux of terrestrial matter transported by sea ice in intermediate and deep waters is well known in the region (Lalande et al., 2016), TEX<sub>86</sub><sup>L</sup> thermometry does not seem to be affected by lateral advection of particles. The flux-weighted mean TEX<sub>86</sub><sup>L</sup> temperature in sinking particles was similar to the one in the underlying surface sediment, indicating that the TEX<sub>86</sub><sup>L</sup> signal did not experience substantial changes while sinking. The flux-weighted mean TEX<sub>86</sub><sup>L</sup> temperature corresponded to temperatures in water depths ranging between 30 and 80 m, where nutrient profiles suggest favorable conditions for Thaumarchaeota.

In the Antarctic Polar Front, changes in GDGT fluxes and TEX<sub>86</sub><sup>L</sup> temperatures are different between the two traps moored at different depths. At the shallow trap, TEX<sub>86</sub><sup>L</sup> estimates do not covary with satellite-derived SSTs, and its flux-weighted mean temperature is 4.6 °C. The warm-biased TEX<sub>86</sub><sup>L</sup> at the deep trap and in the underlying surface sed-

iment may be caused by GDGT contributions from Eur-archaeota, which is dominant in the deeper part of the water column of CDW at the Antarctic Polar Front. Alternatively, a systematic warm bias of the linear calibration in the high-latitude Southern Ocean could explain the discrepancy between  $\text{TEX}_{86}^L$  reconstructed and observed temperatures. The discrepancies can be reduced by using a nonlinear  $\text{TEX}_{86}^L$  calibration developed for high-latitude samples from the Southern Hemisphere.

Our findings offer insights into the potential factors governing  $\text{TEX}_{86}^L$  thermometry such as nutrient availability or archaeal community composition in high-latitude regions. In the high-latitude North Atlantic, regional  $\text{TEX}_{86}^L$  calibrations can be an additional option next to the global calibration, while in the high-latitude Southern Ocean, the benefit of a polynomial  $\text{TEX}_{86}^L$  calibration needs to be further tested. Our studies suggest that OH-GDGT-based calibrations are also worth further investigation in the polar oceans. Accordingly, our study highlights that multiple approaches to global versus regional calibrations, polynomial relationships, or OH-GDGT-based calibrations are beneficial to overcome the limitations of a single global  $\text{TEX}_{86}^L$  calibration in high-latitude ocean regions.

*Data availability.* The data presented here are available in the PANGAEA database (<https://doi.pangaea.de/10.1594/PANGAEA.897268>; Park et al., 2019).

## Appendix A

**Table A1.** Index values for assessing the terrestrial and/or methanogen GDGT inputs at FEVI16 and PF3 sediment traps. The exact sampling interval of each sample number at FEVI16 and PF3 can be found in PANGAEA (<https://doi.pangaea.de/10.1594/PANGAEA.897268>).

Cup no.	FEVI16				PF3							
	(1296 m)				(614 m)				(3196 m)			
	BIT <sup>a</sup>	MI <sup>b</sup>	%GDGT-0 <sup>c</sup>	$\Delta$ RI  <sup>d</sup>	BIT	MI	%GDGT-0	$\Delta$ RI	BIT	MI	%GDGT-0	$\Delta$ RI
1	0.01	0.09	60	0.15	0.00	0.15	58	0.05	0.00	0.34	62	0.23
2	0.01	0.11	60	0.22	0.02	0.14	58	0.08	0.00	0.30	61	0.17
3	0.01	0.11	60	0.21	0.00	0.14	64	0.24	0.00	0.21	58	0.09
4	0.01	0.10	60	0.20	0.00	0.16	61	0.16	–	–	–	–
5	0.01	0.11	61	0.30	0.00	0.19	62	0.17	0.00	0.22	58	0.06
6	0.01	0.10	60	0.23	0.00	0.15	63	0.19	0.00	0.21	56	0.05
7	0.01	0.11	60	0.23	0.00	0.20	65	0.24	0.00	0.19	55	0.08
8	0.01	0.11	60	0.09	0.00	0.19	60	0.08	0.00	0.21	56	0.01
9					0.00	0.19	62	0.16				
10	0.01	0.13	61	0.14	0.00	0.23	64	0.20	–	–	–	–
11					0.00	0.20	59	0.06	–	–	–	–
12	0.01	0.15	59	0.19	0.00	0.21	64	0.26	–	–	–	–
13	0.01	0.13	59	0.10	0.00	0.21	62	0.19	–	–	–	–
14					0.00	0.16	61	0.16	–	–	–	–
15	0.02	0.14	59	0.13	0.00	0.15	60	0.15	–	–	–	–
16	0.02	0.13	61	0.22	0.00	0.14	60	0.11	0.00	0.21	56	0.04
17	0.01	0.11	61	0.20	0.00	0.16	62	0.16	–	–	–	–
18	0.01	0.09	63	0.26								
19	0.01	0.10	64	0.31								
20	–	–	–	–								
Total 20 samples					Total 17 samples							

<sup>a</sup> BIT (Hopmans et al., 2004; Weijers et al., 2006) for terrestrial input when > 0.3. <sup>b</sup> MI (Zhang et al., 2011) for methanotrophic archaeal input when > 0.5 and normal oceanic signal when < 0.3. <sup>c</sup> %GDGT-0 (Blaga et al., 2009; Inglis et al., 2015) for methanogenic archaeal input when > 67 %. <sup>d</sup> | $\Delta$ RI| (Zhang et al., 2016) for both methanotrophic archaeal and terrestrial input > 0.3.

**Author contributions.** EP and GM designed the study. EP measured the samples with the help of JH, analyzed the data, prepared figures and tables, and wrote the paper. All the authors provided feedback on the paper and GM reviewed the paper.

**Competing interests.** The authors declare that they have no conflict of interest.

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