



Paleoclimate Records of the Middle Okinawa Trough Since the Middle Holocene: Modulation of the Low-Latitude Climate

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The ubiquity of glycerol dibiphytanyl glycerol tetraethers (GDGTs) and their temperature sensitivity make them one of the most effective tools for paleoclimate reconstruction. High- and low-latitude climates influence the Okinawa Trough (OT). It receives diverse inputs from the East China Sea, the western Pacific, and the Kuroshio Current, providing good conditions for paleoclimate studies. Here, isoprenoid GDGTs (isoGDGTs), branched GDGTs, and hydroxylated GDGTs (OH-GDGTs) were studied to reconstruct the sea surface temperature (SST) of the central OT for the past 8.2 kyr using the tetraether index of 86 carbon atoms at low latitudes (TEX_{86}^H) and the ring index of OH-GDGTs (RI-OH). The GDGT-0/crenarchaeol ratios ranged from 0.39 to 0.98. The branched and isoprenoid tetraether index and the methane index values were lower than 0.1 and 0.5, respectively, indicating that the isoGDGTs were mainly derived from marine *Thaumarchaeota* and that TEX_{86}^H could be used to reconstruct the paleotemperatures. The TEX_{86}^H SSTs ranged from 21.6 to 27.2°C during 8.2 kyr. The overall range of TEX_{86}^H SSTs is close to the U_{37}^K SST of the middle OT and reflects the mean annual SST. In contrast, RI-OH temperatures varied from 17.4 to 26.0°C, showing a lower trend than TEX_{86}^H SSTs. The core top RI-OH temperature is 24.1°C, in line with the mean annual seawater temperature at 40 m (24.2°C) in the study area, which likely reflects the subsurface temperature in this case. The small overall warming trend of TEX_{86}^H SSTs agrees with the increasing intensity of the Kuroshio Current during the last 8.2 kyr, indicating that the SST evolution is governed by the Kuroshio Current that transports heat from the western tropical Pacific. The decreasing temperature differences between TEX_{86}^H and RI-OH and between U_{37}^K and RI-OH showed increased mixing of the upper water column, which was in good accordance with the increasing low-latitude winter insolation decoupling from the East Asian summer monsoon. The cold event that occurred at 7.4–6.6 kyr was magnified (~5°C) at the TEX_{86}^H and RI-OH temperatures and possibly caused by tephra's significant input (~7.3 kyr).

Keywords: isoprenoid glycerol dibiphytanyl glycerol tetraethers, hydroxylated glycerol dibiphytanyl glycerol tetraethers, TEX_{86}^H , sea surface temperature, okinawa trough

INTRODUCTION

The Okinawa Trough, located in the western North Pacific, was influenced by the East Asian monsoon (ESM). Paleoclimate records indicated that correlations between temperature variation and East Asian summer monsoon (EASM) precipitation have become weak since the middle Holocene (Liew et al., 2006; Peterse et al., 2011; Park et al., 2014). However, such records in marine sediments are sparse (Xu et al., 2018 and references therein). The Okinawa Trough (OT) has been regarded as an ideal location for paleoclimatic, paleohydrogeological, and paleoceanographic studies because it is within the influence of the climate of both the high-latitude North Atlantic and the low-latitude western Pacific Ocean (Sun et al., 2005; Kubota et al., 2010; Ruan et al., 2015; Xu et al., 2018). The seasonal variations in EAM are driven by Northern Hemisphere summer and winter solar insolation, reflecting teleconnection with the high-latitude North Atlantic (An et al., 2001; Wang et al., 2005; Ruan et al., 2015; Huang and Sarnthein, 2021). The Kuroshio Current (KC) originates from the western Pacific warm pool and transports massive amounts of warm and salty water into the OT (Hu et al., 2015; Zheng et al., 2016). Paleoclimate indicators have demonstrated the influences of both the EAM and KC on the OT paleoclimate, such as oxygen isotopes (Kubota et al., 2010), Mg/Ca ratios of planktonic foraminifera (Sun et al., 2005), alkenone unsaturation index (U_{37}^K ; Ruan et al., 2015), and the tetraether index of 86 carbon atoms (TEX_{86} ; Xu et al., 2018). Sea surface temperatures (SSTs) from different locations of the OT tend to reveal different controlling factors on the paleoclimate (Ruan et al., 2015; Zhao et al., 2015; Xu et al., 2018). The cold anomalies recorded in the northern Atlantic, such as the 4 kyr event, have affected the northern and southern OT paleoclimate (Sun et al., 2005; Ruan et al., 2015; Zhao et al., 2015). The U_{37}^K SST of the southern OT has been observed to have decreased since 7.4 kyr BP (Ruan et al., 2015), which was attributed to the reduction in Northern Hemisphere summer solar insolation and the weakening of EASM intensity (Ruan et al., 2015 and references therein). However, in the southern OT, Xu et al. (2018) found that paleotemperatures from U_{37}^K/TEX_{86} decoupled from the EASM during the last 13.3 kyr BP. In contrast, the western tropical Pacific and Northern Hemisphere winter solar insolation were demonstrated to be the controlling factors of SSTs. Whether EAM changes in the OT are in phase

with temperature changes in the EAM region during the Holocene remains unsolved (Liew et al., 2006; Peterse et al., 2011; Park et al., 2014). Therefore, further investigations are needed to elucidate the OT paleotemperature evolution and the controlling factors.

The TEX_{86} index (**Table 1**) was proposed by Schouten et al. (2002) based on the increasing number of cyclopentane rings in isoprenoid glycerol dibiphytanyl glycerol tetraethers (isoGDGTs) with increasing temperature (Gliozzi et al., 1983; Uda et al., 2001; Sinninghe Damsté et al., 2002). The export depth of the TEX_{86} signal in the water column is still under debate (Schouten et al., 2002, 2013; Ingalls et al., 2006; Rueda et al., 2009; Jia et al., 2012; Zhang and Liu, 2018). Most researchers now regard TEX_{86} as a better reflection of subsurface temperatures (Huguet et al., 2007; Jia et al., 2012). However, some studies have shown that TEX_{86} corresponds well with the annual mean SST (Ingalls et al., 2006; Rueda et al., 2009). Consequently, the use of TEX_{86} for temperature reconstruction in specific areas needs to be evaluated.

Marine Group I (MG-I) *Thaumarchaeota* were proposed to produce isoGDGTs used in the TEX_{86} equation (**Table 1**; **Supplementary Figure S1**; Brochier-Armanet et al., 2008; Spang et al., 2010). To achieve better accuracy, Kim et al. (2010) proposed TEX_{86}^L for high latitudes (<15°C) and TEX_{86}^H for low latitudes (>15°C) according to studies on global ocean samples (**Table 1**). To further assess the proxy, the Bayesian regression approach, which offers several advantages, was introduced to the TEX_{86} -SST calibration to provide a comprehensive estimation of past changes in SST from TEX_{86} in both modern and ancient environments (Tierney and Tingley, 2014). However, many environmental factors, except for SSTs, are included in the Bayesian regression approach for TEX_{86} -SST calibrations, making it more challenging to apply in geological sites than TEX_{86}^L/TEX_{86}^H (Kim et al., 2010; Tierney and Tingley, 2014). Since isoprenoid tetraethers produced by marine *Thaumarchaeota* can be mixed with those from *Euryarchaeota* thriving in seawater and *Thaumarchaeota* in soils, indicators such as branched and isoprenoid tetraether index (BIT; Hopmans et al., 2004) and methane index (MI; Zhang et al., 2011) are used to assess isoGDGTs contributed from soil origin and *Euryarchaeota*. For example, an MI less than 0.5 is used to preclude the contribution of methanotrophic archaea (Zhang et al., 2011). In addition to isoGDGTs, hydroxylated GDGTs (OH-GDGTs) are widespread in marine environments (Liu et al., 2012; Fietz et al., 2013; Huguet et al., 2013; Lü et al., 2015). In culture experiments, archaea affiliated with *Thaumarchaeota* Group I.1a have produced OH-GDGTs (Liu et al., 2012; Elling et al., 2014), which were not observed in *Thaumarchaeota* Group I.1b (Sinninghe Damsté et al., 2012b). The ratio of sum OH-GDGTs relative to sum isoGDGTs was related to SSTs and increased with increasing latitude (Huguet et al., 2013). In addition, a positive correlation between the number of cyclopentane rings of OH-GDGTs and SSTs was reported in the surface sediments from the subpolar and polar areas (Fietz et al., 2013). Although the mechanism for OH-GDGTs in response to SST is unclear, the ring index based on OH-GDGTs (RI-OH) is a potential approach for temperature reconstruction (**Table 1**; Lü et al., 2015; Yang et al., 2018).

TABLE 1 | Initial definitions of various proxies are used in this article.

Index Definitions	authors
$TEX_{86} = \frac{GDGT-2+GDGT-3+crenarchaeol\ isomer}{GDGT-1+GDGT-2+GDGT-3+crenarchaeol\ isomer}$	Schouten et al. (2002)
$TEX_{86}^L = \log\left(\frac{GDGT-2}{GDGT-1+GDGT-2+GDGT-3}\right)$	Kim et al. (2010)
$RI - OH = \frac{10H-GDGT-1+2 \times [OH-GDGT-2]}{[OH-GDGT-1]+[OH-GDGT-2]}$	Lü et al. (2015)
$BIT = \frac{([GDGT-1a]+[GDGT-1b]+[GDGT-11a])}{([GDGT-1a]+[GDGT-1b]+[GDGT-11a]+[Crenarchaeol])}$	Hopmans et al. (2004)
$MI = \frac{[GDGT-1]+[GDGT-2]+[GDGT-3]}{[GDGT-1]+[GDGT-2]+[GDGT-3]+[Crenarchaeol\ isomer]}$	Zhang et al. (2011)
$\%GDGT - 2 = \frac{GDGT-2}{[GDGT-1]+[GDGT-2]+[GDGT-3]+[Crenarchaeol\ isomer]}$	Sinninghe Damsté et al. (2012a)

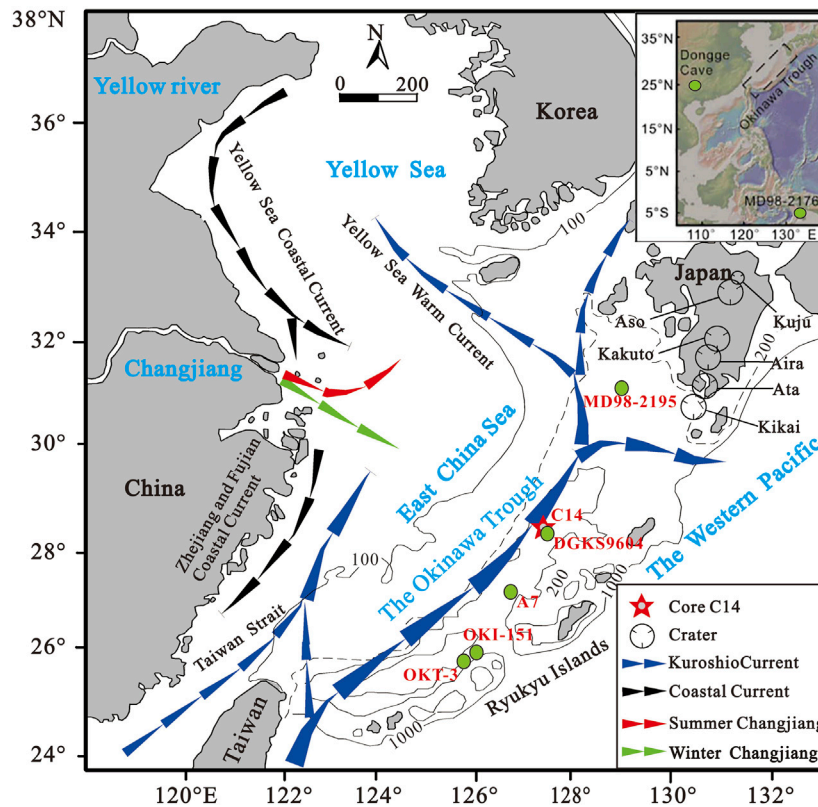


FIGURE 1 | Location of core C14 in the middle Okinawa Trough (OT) and selected paleoenvironmental settings, including Dongge Cave in southern China (Dykoski et al., 2005), core MD98-2176 in the western tropical Pacific (Stott et al., 2004), cores A7 (Sun et al., 2005) and DGKS9604 (Yu et al., 2009) in the middle OT, cores OKT-3 (Zhao et al., 2015) and OKI-151 (Xu et al., 2018) in the southern OT, and core MD98-2195 (Yamamoto et al., 2013) in the northern OT. The map was revised from Dou et al. (2010). The Contour unit is meter. Volcano locations around Japan were modified according to Machida (1999).

TEX_{86}^H has been used to reconstruct the paleotemperature records of the southern and northern OT, and a gradual warming trend has been identified since the Holocene (Yamamoto et al., 2013; Zhao et al., 2015; Xu et al., 2018). Generally, shifts in the main axis and changes in the KC strength, variations in EAM, sea-level changes, and freshwater inputs are potential factors for paleoclimate changes in the OT (Jian et al., 2000; Kubota et al., 2010; Zhao et al., 2015). The influence of the EAM on SST variations was identified in the middle and northern OT (Yu et al., 2009; Kubota et al., 2010). However, low-latitude western tropical Pacific dominance via KC variations has been demonstrated in the southern OT (Xu et al., 2018). Therefore, factors controlling the paleotemperature in specific areas of the OT need to be clarified. Here, we used TEX_{86}^H temperatures and temperatures calibrated from RI-OH as an approach to reconstruct the paleoclimate and hydrological evolution of the middle OT since the middle Holocene.

OCEANOGRAPHIC SETTING

The OT, a back-arc basin, formed from the subduction of the Philippine Sea Plate beneath the Eurasian Plate at the Ryukyu

Trench (Figure 1; Sibuet et al., 1998; Guo et al., 2022). The strike direction of the middle OT is approximately NE–SW, E–W to the south, and NNE–SSW to the north (Wu et al., 2014). The EAM and KC have been reported to dominate the modern oceanography of the middle OT (Ruan et al., 2015; Zhao et al., 2015). Two distinct seasons are present due to the seasonal reversals of the EAM, driven by the annual cycle of insolation and its effect on the land-sea thermal contrast (Dykoski et al., 2005; Sun et al., 2005; Wang et al., 2005; Huang and Sarthain, 2021). In the boreal summer, a warm, wet season caused by the northward migration of the intertropical convergence zone and the maximum monsoonal convective rainfall dominates the OT (Wang et al., 2005; Zheng et al., 2014). In contrast, northeasterly winds lead to a cool, dry season in the boreal winter (Diekmann et al., 2008; Zheng et al., 2014). The KC, originating from the Western Pacific warm pool, flows northeast above the OT and returns to the North Pacific through the Tokara Strait (Yu et al., 2009; Hu et al., 2015), taking warm, saline water into the OT (Hu et al., 2015).

The sea level has been roughly constant for the study area during the last 8.2 kyr BP (Liu et al., 2004). The sediment core C14 is far from the coastline and the shelf of the East China Sea (ECS; Figure 1). At present, the mean annual SST is 24.8°C, with

TABLE 2 | Planktonic foraminifera ¹⁴C age data.

series	Depth (cmbstf)	Planktonic foraminifera	AMS ¹⁴ C age (yr BP)	Calibrated age (yr BP)
498910	35–37.5 ^a	<i>G.ruber</i> + <i>G.sacculifer</i>	660±30	616–730
498911	65–67.5	<i>G.ruber</i> + <i>G.sacculifer</i>	930±30	994–820
498912	155–157.5 ^a	<i>G.ruber</i> + <i>G.sacculifer</i>	1890±30	1858–2045
498913	245–247.5	<i>G.ruber</i> + <i>G.sacculifer</i>	2800±30	3172–2957
498914	365–367.5 ^a	<i>G.ruber</i> + <i>G.sacculifer</i>	4350±30	4943–5227
498915	455–457.5	<i>G.ruber</i> + <i>G.sacculifer</i>	5840±30	6831–6646
498916	545–547.5	<i>G.ruber</i> + <i>G.sacculifer</i>	6500±30	7520–7385
498917	575–577.5 ^a	<i>G.ruber</i> + <i>G.sacculifer</i>	7340±30	8170–8327

^aData from Xu et al. (2020).

summer and winter seasonal temperatures of approximately 27.8 and 22.1°C, respectively. The maximum SST near the core site is 28.7°C in August, and the minimum is 21.5°C in February (Locarnini et al., 2013).

METHODS

Materials and Age Model

Sediment core C14 (580 cm in length) was retrieved from the middle OT at a water depth of approximately 1100 m (Figure 1; 28°39.587'N, 127°19.194'E). The top part of the core was lost during sampling. To make up the lost part, the upper 10 cm of core C10 (28°38.8'N, 127°21.283'E; water depth of 960 m) was utilized to represent the missing top part. After sampling, the sediment core was segmented, transported to the laboratory, and kept at –20°C until analysis. A tephra layer was found between depths of 532.5 and 442.5 cm.

To obtain AMS ¹⁴C dating for the core, eight samples of the planktonic foraminifer *Globigerinoides ruber* (*G. ruber*) and *G. sacculifer* were selected at equal intervals and analyzed by Beta Analytic Inc., United States (Table 2). The raw ¹⁴C dates were calibrated using the Marine13 dataset (Reimer et al., 2013) and expressed in calibrated years BP (years before AD 1950). We adopted a ΔR value of 30 ± 41 years in the calibration.

Lipid Extraction

37 samples were collected at 15–30 cm intervals from core C14 (36 samples from core C14 and 1 from core C10). They were freeze-dried and homogenized. Twenty to 30 g of powder (dry weight) were Soxhlet-extracted for 72 h with a mixture of dichloromethane and methanol (9:1; v:v) to obtain the total lipid extracts (TLEs). The TLE was separated into neutral (n-hexane) and polar (methanol) fractions with a silica gel column. The polar fractions were filtered through a 0.45 μm polytetrafluoroethylene filter membrane (PTFE) to remove particulate matter and stored at –20°C.

Instrumental Analysis

The dried polar fractions were redissolved in 300 μl n-hexane and isopropanol (98.2:1.8, v/v). No internal standard was

added. The analysis was performed on an Agilent 1200 series high-performance liquid chromatography–atmospheric pressure chemical ionization–6460 triple, quadrupole mass spectrometer (HPLC–APCI–MS²) following the analytical protocol of Hopmans et al. (2016). Separation was achieved on two HPLC silica columns (BEH HILIC columns, 2.1 × 150 mm, 1.7 μm) in series, maintained at 30°C. The GDGTs were eluted isocratically for 25 min with 18% B, followed by a linear gradient to 35% B in 25 min and then a linear gradient to 100% B in 30 min and kept in for 10 min to clean the column with a constant flow rate of 0.2 ml/min. Finally, the gradient of B was back at 18% for 20 min to re-equilibrate the column. Solvent A was n-hexane, and B was n-hexane: isopropanol (9:1, v/v). The GDGTs were ionized in an atmospheric pressure chemical ionization chamber and detected using single ion monitoring (SIM) mode. The relative abundance of compounds was determined by integrating the areas of the (M + H)⁺ (protonated molecular ion) peaks at 1302, 1300, 1298, 1296, and 1292 for isoGDGTs and OH-GDGTs (Liu et al., 2012) and 1050, 1048, 1046, 1036, 1034, 1032, 1022, 1020, and 1018 for branched GDGTs (brGDGTs). The analytical error for duplicate measurements was better than ±0.008. The positive ion APCI settings were set as follows (Hopmans et al., 2016): probe heater temperature, 400°C; sheath gas (N₂) pressure, 50 AU (arbitrary units); auxiliary gas (N₂) pressure, 5 AU; spray current, 5 μA; capillary temperature, 350°C; S-lens, 100 V.

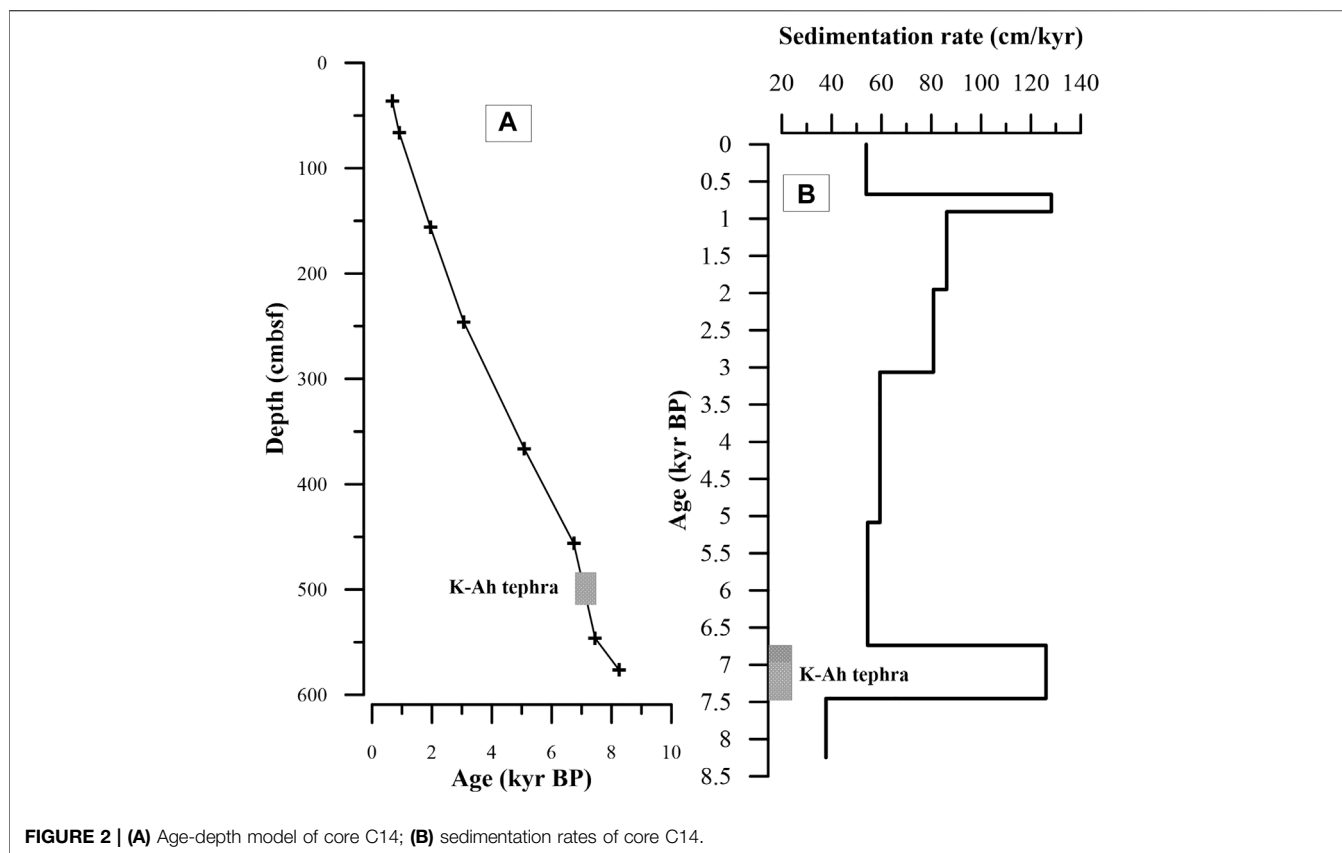
TEX₈₆^H, RI-OH and BIT

In this study, the SST calibration of the TEX₈₆^H proxy (Kim et al., 2010) was used to reconstruct the paleoclimate evolution due to the high temperatures of over 15°C (low latitude) in the OT. Cren' represents the crenarchaeol isomer (see Supplementary Figure S1 for details).

$$\text{TEX}_{86}^{\text{H}} = \log\left(\frac{\text{GDGT} - 2 + \text{GDGT} - 3 + \text{Cren}'}{\text{GDGT} - 1 + \text{GDGT} - 2 + \text{GDGT} - 3 + \text{Cren}'}\right);$$

$$\text{SST} = 68.4 \times (\text{TEX}_{86}^{\text{H}}) + 38.6 \quad (1)$$

The empirical equation of RI-OH (Lü et al., 2015) was used to supplement the SST reconstruction.



$$RI - OH = \frac{[OH - GDGT - 1] + 2 \times [OH - GDGT - 2]}{[OH - GDGT - 1] + [OH - GDGT - 2]}$$

$$SST = 35.71 \times RI - OH - 32.86 \quad (2)$$

Based on the analytical error (± 0.008), we calculated the corresponding errors, which are approximately $\pm 0.4^\circ\text{C}$ and $\pm 0.3^\circ\text{C}$ for calibrations 1) and (2), respectively.

As brGDGT regio isomers were detected (Supplementary Figure S1; De Jonge et al., 2014), the BIT formula used in this article was modified as follows:

$$BIT = \frac{GDGT - Ia + GDGT - IIa + GDGT - IIIa' + GDGT - IIIa + GDGT - IIIa'}{Crenarchaeol + GDGT - Ia + GDGT - IIa + GDGT - IIa + GDGT - IIIa + GDGT - IIIa' + GDGT - IIIa'} \quad (3)$$

RESULTS

AMS ^{14}C Dating

The AMS ^{14}C dating results are listed in Table 2. The time scale was established by linear interpolation and extrapolation. An age model was set for core C14 after calibration, and the mean sedimentation rate of core C14 was ~ 70 cm/kyr during the last 8.2 kyr BP (Figures 2A,B).

For the tephra layer at 532.5–442.5 cm (7.4–6.6 kyr BP), an extremely high sedimentation rate of ~ 126 cm/kyr was identified (Figure 2B).

Distribution of GDGTs

The relative contributions of the isoGDGTs, brGDGTs, and OH-GDGTs are listed in Table 3. The isoGDGTs dominated all GDGTs, with contributions ranging from 88 to 98%, while brGDGTs accounted for 1.4–10.3%, and OH-GDGTs accounted for less than 6.0% of the total GDGTs. Crenarchaeol was the most abundant component among the isoGDGTs in all of the samples, accounting for 32–50% of all of the isoGDGTs, followed by GDGT-0 (range 16–32%), whereas only small amounts of GDGT-1, -2, -3, and the crenarchaeol isomer were detected in all of the samples.

Past Temperatures Reconstructed by isoGDGT- and OH-GDGT-Based Proxies

For the past 8.2 kyr BP, the TEX_{86}^H values in core C14 ranged from -0.25 to -0.17 , corresponding to temperatures ranging from 21.6 to 27.2°C (Equation 2; Table 4; Figure 3C). The index of RI-OH in core C14 ranged from 1.41 to 1.65, revealing temperatures ranging from 17.4 to 26.0°C (Equation 3; Table 4; Figure 3C). To better understand TEX_{86} and RI-OH temperatures, the mean seasonal and annual temperatures and monthly mean water

TABLE 3 | The contribution of specific GDGTs with depth in core C14.

Sample	Depth (cmbsf)	%isoGDGTs							% (b)	% (OH)		
		% [0]	% [1]	% [2]	% [3]	% [4]	%Cren	%cren'		% [OH-0]	% [OH-1]	% [OH-2]
C10_01	7.5	21.45	5.54	5.82	1.26	12.48	47.67	2.78	2.09	0.25	0.27	0.39
C14_01	37.5	21.68	5.10	5.75	1.21	12.22	47.81	3.16	1.96	0.31	0.33	0.47
C14_02	67.5	20.82	5.45	5.54	1.22	12.00	48.40	3.61	1.93	0.24	0.29	0.50
C14_03	83.75	18.28	9.52	9.78	3.10	3.93	46.79	5.66	2.94	-	-	-
C14_04	97.5	20.79	5.14	5.58	1.20	12.78	48.82	3.13	1.71	0.22	0.25	0.38
C14_05	127.5	20.54	5.14	6.13	1.31	12.46	47.98	3.56	1.87	0.23	0.31	0.47
C14_06	143.75	17.33	7.18	8.69	2.48	8.18	44.51	4.20	7.43	-	-	-
C14_07	157.5	20.96	5.29	5.93	1.26	11.62	48.91	3.41	1.80	0.20	0.27	0.35
C14_08	173.75	25.23	9.14	11.31	1.69	5.60	36.62	4.46	3.33	1.15	0.58	0.89
C14_09	187.5	21.57	5.26	6.07	1.24	11.53	48.52	3.11	1.85	0.20	0.23	0.42
C14_10	203.75	16.29	10.37	12.02	3.03	5.76	36.25	4.91	6.71	1.68	1.19	1.79
C14_11	212.5	20.34	5.45	6.14	1.29	11.86	49.36	3.15	1.57	0.21	0.24	0.39
C14_12	217.5	19.74	5.47	5.55	1.31	11.77	50.44	3.26	1.56	0.22	0.26	0.42
C14_13	228.75	29.09	8.98	9.72	2.30	5.86	33.58	4.74	5.73	-	-	-
C14_14	247.5	20.34	5.31	5.54	1.23	12.97	48.41	3.61	1.76	0.19	0.23	0.41
C14_15	263.75	23.73	9.36	9.82	2.80	4.97	35.58	4.34	5.77	1.74	0.75	1.14
C14_16	277.5	21.19	5.13	5.47	1.18	11.98	49.71	2.95	1.50	0.24	0.25	0.40
C14_17	293.75	27.12	9.35	8.92	2.26	6.14	34.96	5.35	5.90	-	-	-
C14_18	307.5	21.13	5.67	5.65	1.30	12.54	48.32	3.04	1.47	0.24	0.26	0.38
C14_19	323.75	21.95	9.35	10.71	2.11	5.16	34.73	5.66	10.33	-	-	-
C14_20	337.5	19.93	5.24	5.61	1.26	12.36	49.93	3.44	1.42	0.18	0.24	0.39
C14_21	353.75	19.68	9.25	10.39	2.93	6.70	35.68	5.49	5.94	1.80	0.84	1.30
C14_22	367.5	19.90	5.12	5.59	1.17	12.51	50.20	3.22	1.47	0.21	0.25	0.36
C14_23	383.75	18.78	12.18	13.76	3.63	6.19	37.11	5.63	2.72	-	-	-
C14_24	397.5	21.86	5.26	5.80	1.20	12.42	48.27	2.91	1.43	0.23	0.25	0.37
C14_25	413.75	24.83	7.63	9.14	2.24	6.38	37.73	3.98	4.58	1.23	0.94	1.32
C14_26	427.5	22.82	5.83	5.80	1.19	11.66	47.11	2.90	1.62	0.32	0.32	0.43
C14_27	443.75	29.44	7.63	7.50	1.92	4.85	38.14	4.21	3.63	1.24	0.73	0.71
C14_28	457.5	28.62	4.67	3.72	0.86	10.68	46.05	1.90	1.93	0.66	0.53	0.38
C14_29	473.75	23.81	8.90	7.59	1.77	6.13	38.92	2.74	5.37	2.50	1.31	0.96
C14_30	487.5	29.24	4.81	3.62	0.91	11.43	44.48	1.74	2.06	0.73	0.57	0.41
C14_31	503.75	26.67	7.06	6.04	1.46	7.27	40.36	2.16	4.53	2.60	1.10	0.75
C14_32	517.5	29.25	5.17	4.02	0.90	11.48	43.30	1.79	2.18	0.88	0.58	0.45
C14_33	533.75	31.63	6.41	6.54	1.48	8.39	32.20	2.37	5.32	2.32	1.66	1.68
C14_34	547.5	20.13	5.60	5.71	1.14	12.45	49.15	3.09	1.79	0.25	0.31	0.38
C14_35	563.75	23.21	8.50	9.94	2.38	5.47	33.10	5.71	8.29	1.27	1.01	1.12
C14_36	577.5	21.56	5.34	5.96	1.10	12.81	47.57	3.12	1.68	0.25	0.25	0.36

(x), GDGT-x; Cren, crenarchaeol; Cren', crenarchaeol isomer; (b), brGDGTs; (OH-x), OH-GDGT-x

temperatures at different depths are summarized in **Figures 3A,B**.

DISCUSSION

Constrains of GDGT Sources

The isoGDGTs used in TEX₈₆ are derived from marine *Thaumarchaeota* (Brochier-Armanet et al., 2008; Spang et al., 2010; Schouten et al., 2013). However, GDGTs-1, -2, -3, and the crenarchaeol isomer can also be synthesized by additional archaea (Sinninghe Damsté et al., 2002; Schouten et al., 2008; Pitcher et al., 2011). For example, marine group II (MG-II) *Euryarchaeota* thriving in shallow water can synthesize GDGTs-1, -2, and -3 (Turich et al., 2007; Schouten et al., 2008). IsoGDGTs from soil and brGDGTs produced *in situ* in marine environments may contribute to the GDGT pool (Hopmans et al., 2004; Peterse et al., 2009; Schouten et al., 2013). Such inputs may affect the application of TEX₈₆ in

paleotemperature reconstruction. Therefore, indicators such as BIT (Hopmans et al., 2004; De Jonge et al., 2014), GDGT-0/crenarchaeol (Blaga et al., 2009), MI (Zhang et al., 2011), GDGT-2/crenarchaeol (Weijers et al., 2011), %GDGT-2 (Sinninghe Damsté et al., 2012a), and GDGT-2/GDGT-3 (Taylor et al., 2013) were introduced to assess the biological precursors of the isoGDGTs (**Table 1**).

In this study, crenarchaeol was the most abundant component among isoGDGTs, with GDGT-0/crenarchaeol ratios ranging from 0.4 to 1.0 (between 0.2 and 2), suggesting that the isoGDGTs were predominantly produced by MG-I *Thaumarchaeota* in core 14 (Schouten et al., 2008, 2013; Spang et al., 2010). IsoGDGTs introduced by soil and rivers were negligible, as indicated by the low BIT values of 0.01–0.10 (<0.10; **Table 4**; Hopmans et al., 2004; Kim et al., 2010; De Jonge et al., 2014). Apart from terrestrial input, methanotrophic *Euryarchaeota* prevailing in cold seeps can contribute to GDGT-1, -2, and -3 and modify the distribution of isoGDGTs (Elvert et al., 2005; Guan et al., 2019). In this study, the ratios of GDGT-2/crenarchaeol varied from 0.1 to 0.4 (<0.4), GDGT-

TABLE 4 | Various indices were used to evaluate the application of TEX₈₆^H and reconstruct paleotemperatures in core C14.

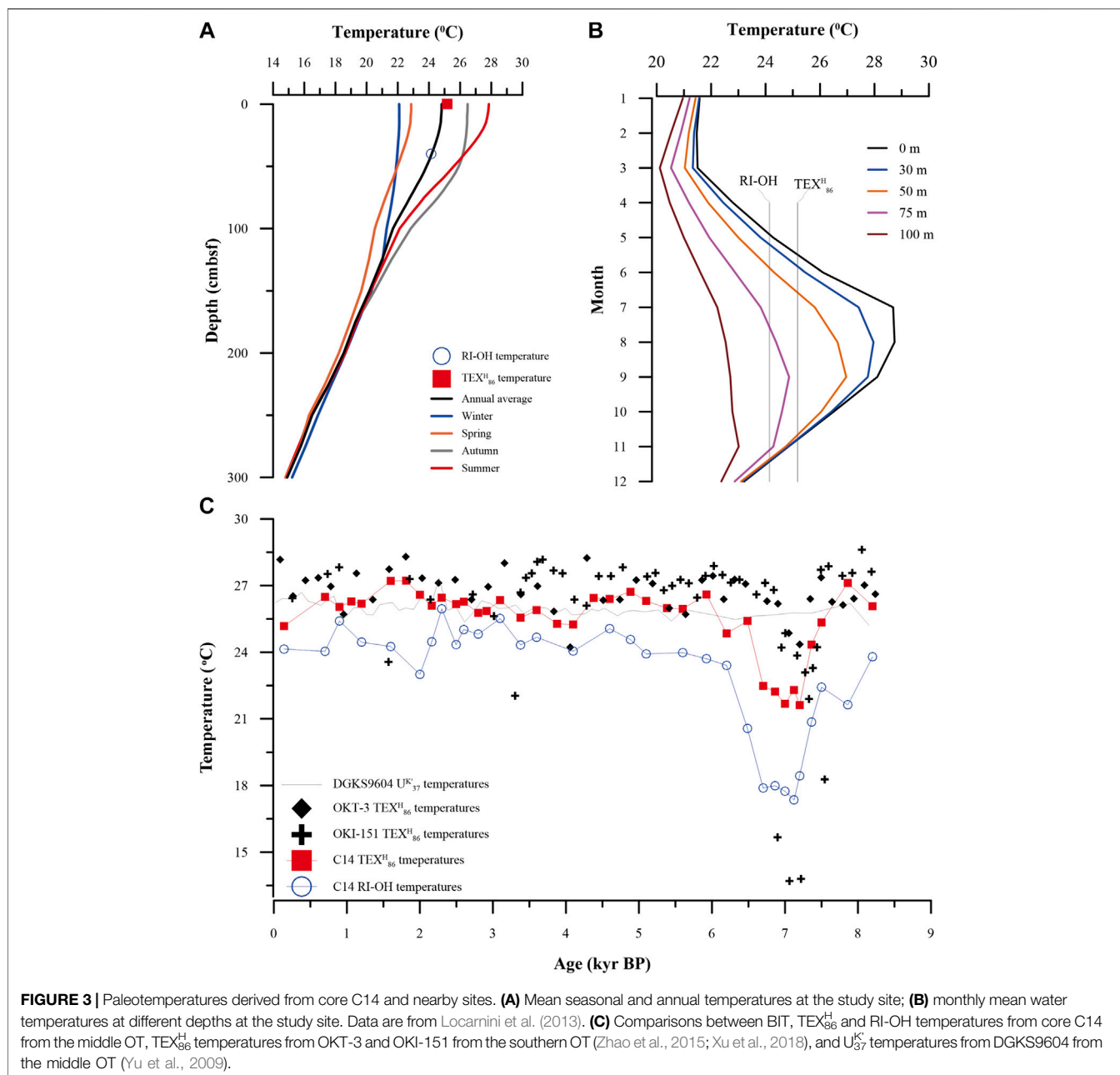
Sample	Depth (cmbsf)	Age (kyr)	%GDGT-2	GDGT-0/Cren	GDGT-2/Cren	GDGT-2/GDGT-3	MI	BIT	TEX ₈₆ ^H		RI-OH	
									Values	SST (°C)	Values	SST (°C)
C10_01	7.5	0.1	38	0.5	0.1	4.6	0.2	0.02	-0.196	25.2	1.596	24.1
C14_01	37.5	0.7	38	0.5	0.1	4.7	0.2	0.02	-0.177	26.5	1.593	24
C14_02	67.5	0.9	35	0.4	0.1	4.6	0.2	0.01	-0.184	26	1.631	25.4
C14_03	83.75	1.1	35	0.4	0.2	3.2	0.3	0.02	-0.18	26.3	-	-
C14_04	97.5	1.2	37	0.4	0.1	4.7	0.2	0.01	-0.182	26.2	1.605	24.5
C14_05	127.5	1.6	38	0.4	0.1	4.7	0.2	0.02	-0.167	27.2	1.599	24.3
C14_06	143.75	1.8	39	0.4	0.2	3.5	0.3	0.1	-0.166	27.2	-	-
C14_07	157.5	2.0	37	0.4	0.1	4.7	0.2	0.01	-0.176	26.6	1.564	23
C14_08	173.75	2.2	43	0.7	0.3	6.7	0.4	0.06	-0.183	26.1	1.605	24.5
C14_09	187.5	2.3	39	0.4	0.1	4.9	0.2	0.01	-0.177	26.5	1.647	26
C14_10	203.75	2.5	40	0.5	0.3	4.0	0.4	0.05	-0.182	26.2	1.601	24.3
C14_11	212.5	2.6	38	0.4	0.1	4.8	0.2	0.01	-0.18	26.3	1.621	25
C14_12	217.5	2.8	36	0.4	0.1	4.3	0.2	0.01	-0.188	25.8	1.615	24.8
C14_13	228.75	2.9	38	0.9	0.3	4.2	0.4	0.06	-0.186	25.8	-	-
C14_14	247.5	3.1	35	0.4	0.1	4.5	0.2	0.01	-0.179	26.3	1.634	25.5
C14_15	263.75	3.4	37	0.7	0.3	3.5	0.4	0.06	-0.191	25.5	1.601	24.3
C14_16	277.5	3.6	37	0.4	0.1	4.6	0.2	0.01	-0.186	25.9	1.611	24.7
C14_17	293.75	3.9	35	0.8	0.3	4.0	0.3	0.07	-0.195	25.3	-	-
C14_18	307.5	4.1	36	0.4	0.1	4.3	0.2	0.01	-0.195	25.2	1.593	24
C14_19	323.75	4.4	39	0.6	0.3	5.1	0.4	0.1	-0.178	26.4	-	-
C14_20	337.5	4.6	36	0.4	0.1	4.5	0.2	0.01	-0.178	26.4	1.622	25.1
C14_21	353.75	4.9	37	0.6	0.3	3.5	0.4	0.06	-0.174	26.7	1.608	24.6
C14_22	367.5	5.1	37	0.4	0.1	4.8	0.2	0.01	-0.18	26.3	1.59	23.9
C14_23	383.75	5.4	39	0.5	0.4	3.8	0.4	0.03	-0.184	26	-	-
C14_24	397.5	5.6	38	0.5	0.1	4.8	0.2	0.01	-0.185	26	1.591	24
C14_25	413.75	5.9	40	0.7	0.2	4.1	0.3	0.04	-0.175	26.6	1.584	23.7
C14_26	427.5	6.2	37	0.5	0.1	4.9	0.2	0.01	-0.201	24.8	1.575	23.4
C14_27	443.75	6.5	35	0.8	0.2	3.9	0.3	0.03	-0.193	25.4	1.496	20.6
C14_28	457.5	6.7	33	0.6	0.1	4.3	0.2	0.02	-0.236	22.5	1.421	17.9
C14_29	473.75	6.9	36	0.6	0.2	4.3	0.3	0.04	-0.239	22.2	1.424	18
C14_30	487.5	7.0	33	0.7	0.1	4.0	0.2	0.02	-0.247	21.7	1.417	17.7
C14_31	503.75	7.1	36	0.7	0.2	4.1	0.3	0.04	-0.238	22.3	1.406	17.4
C14_32	517.5	7.2	34	0.7	0.1	4.5	0.2	0.02	-0.248	21.6	1.436	18.4
C14_33	533.75	7.4	39	1.0	0.2	4.4	0.3	0.04	-0.209	24.3	1.504	20.9
C14_34	547.5	7.5	37	0.4	0.1	5.0	0.2	0.01	-0.194	25.3	1.548	22.4
C14_35	563.75	7.9	38	0.7	0.3	4.2	0.4	0.08	-0.168	27.1	1.526	21.6
C14_36	577.5	8.2	38	0.5	0.1	5.4	0.2	0.01	-0.183	26.1	1.586	23.8

2/GDGT-3 showed a constant trend below 5.0, the MI index ranged between 0.2 and 0.4 (<0.5), and %GDGT-2 fell in the range of 33–39 (<45), indicating that the contributions of methanotrophic archaea and MG-II *Euryarchaeota* were low (Table 4; Blaga et al., 2009; Weijers et al., 2011; Zhang et al., 2011; Sinninghe Damsté et al., 2012a; Taylor et al., 2013). Consequently, isoGDGTs predominantly originating from MG-I *Thaumarchaeota* and TEX₈₆^H can be used to reconstruct paleotemperatures in core C14.

Implications of TEX₈₆^H Temperatures

TEX₈₆^H studies revealed relatively variable temperature records in the OT and ECS (Nakanishi et al., 2012; Xu et al., 2018; Yuan et al., 2018). Yamamoto et al. (2013) compared temperatures derived from Mg/Ca, U₃₇^K and TEX₈₆^H at site MD98-2195 of the northern OT. Mg/Ca- and U₃₇^K-derived temperatures were assigned to summer and spring seawater temperatures, respectively, because they behaved differently to seasonal variations in the sinking fluxes. Still, the core-top values

yielded similarities to summer and spring SSTs, respectively (Yamamoto et al., 2013 and references therein). Since GDGTs in sinking particles were well mixed, the lower TEX₈₆^H temperature most likely represented the annual subsurface seawater temperature, as assumed by Yamamoto et al. (2013). However, Zhao et al. (2015) showed similarity between modern summer seawater temperature and core-top TEX₈₆^H temperature at the southern OT and concluded that both U₃₇^K and TEX₈₆^H temperatures represented summer SSTs. Similar observations were made by Xu et al. (2018), and both U₃₇^K and TEX₈₆^H temperatures were used to indicate annual SST at the southern OT. Therefore, TEX₈₆^H calibrations for specific locations may be closely related to the specific local hydrology. In this study, the core-top TEX₈₆^H and RI-OH temperatures at the study site were 25.2 and 24.1°C, respectively. This is in line with the mean annual SST (24.8°C) and mean annual seawater temperature at 40 m below the sea surface (24.2°C) (Figure 3A). In addition, the core-top TEX₈₆^H temperature agreed well with the SST in June and November and the average temperature from June to November



at depths of 50–7 m (**Figure 3B**). However, $\text{TEX}_{86}^{\text{H}}$ reflects SST in a specific month, which is unlikely because the GDGTs produced in different seasons have been demonstrated to be suspended and well mixed in the surface water in both the western North Pacific (Yamamoto et al., 2012) and the northern OT (Nakanishi et al., 2012). In this study, the average $\text{TEX}_{86}^{\text{H}}$ SST ($26.1 \pm 1.3^\circ\text{C}$; except for 7.4–6.6 kyr BP) in core C14 is comparable to the $\text{TEX}_{86}^{\text{H}}$ SST of the southern OT from 8.2 kyr BP to the present (**Figure 3C**; Zhao et al., 2015; Xu et al., 2018), which is close to the average U_{37}^{K} SST ($26.0 \pm 1.3^\circ\text{C}$; **Figure 3C**) when compared to a nearby site (core DGKS9604; Yu et al., 2009), indicating that the $\text{TEX}_{86}^{\text{H}}$ temperatures during the last 8.2 kyr BP reflected mean annual SSTs rather than temperatures in a specific short period.

Interestingly, the temperatures derived from RI-OH were generally lower than the $\text{TEX}_{86}^{\text{H}}$ and U_{37}^{K} SSTs with a similar changing pattern and trend (**Table 4**; **Figures 3A,C**). To date, the biological precursors of OH-GDGTs remain unclear. To date, OH-GDGTs have been observed in cultures of archaea affiliated with *Thaumarchaeota* Group I.1a (Sinninghe Damsté et al., 2012b; Elling et al., 2014), whereas TEX_{86} -related isoGDGTs are explicitly ascribed to *Thaumarchaeota* Group I.1b (Brochier-Armanet et al., 2008; Spang et al., 2010). The potentially different origins of OH-GDGTs suggest that the RI-OH index may supplement $\text{TEX}_{86}^{\text{H}}$ in marine environments (Lü et al., 2015). Lü et al. (2015) and Yang et al. (2018) analyzed the surface sediments of the marginal sea in China and revealed a high correlation between RI-OH and the local mean

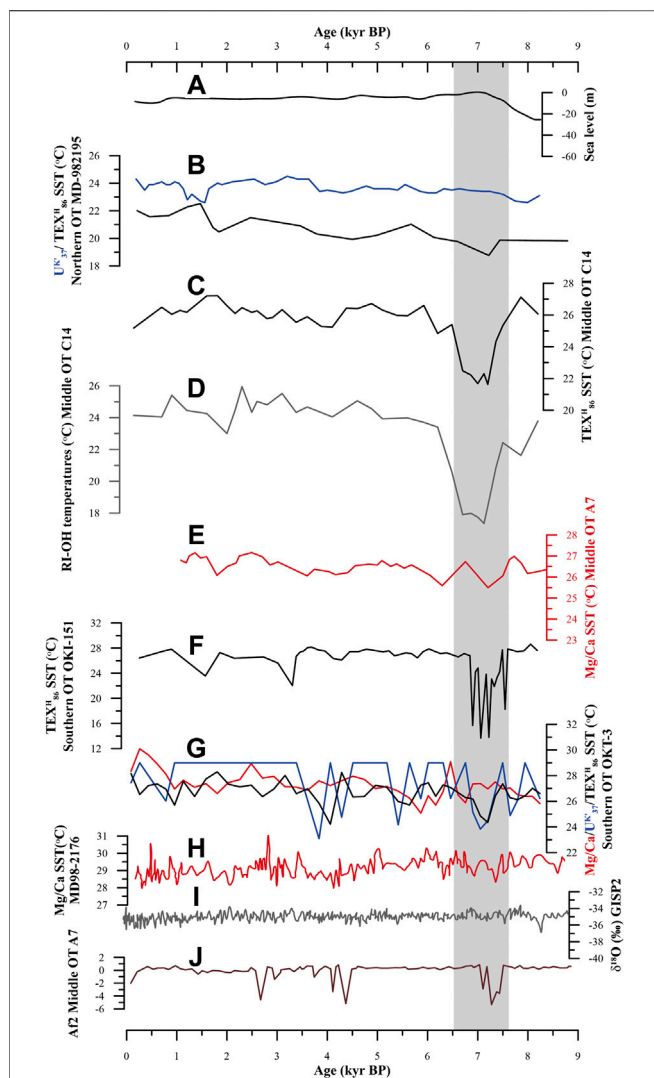


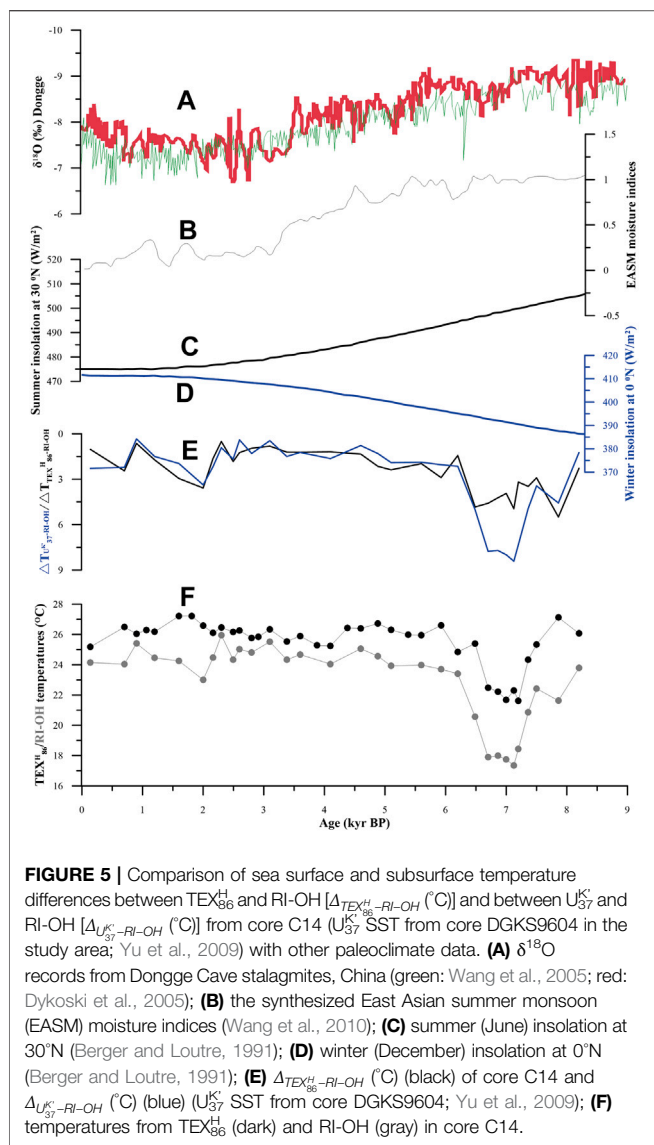
FIGURE 4 | Comparison of paleotemperature variations inferred from core C14 with other paleoclimate data. **(A)** Postglacial sea-level changes in the western Pacific (Liu et al., 2004); **(B)** Temperatures from U_{37}^K (blue) and TEX_{86}^H (dark) in core MD98-2195 (Yamamoto et al., 2013); **(C)** TEX_{86}^H SST of C14 in this article; **(D)** Temperatures from RI-OH in core C14 in this article; **(E)** Mg/Ca SST of A7 (Sun et al., 2005); **(F)** SST from TEX_{86}^H in core OKI-151 (Xu et al., 2018); **(G)** SST from Mg/Ca (red), U_{37}^K (blue) and TEX_{86}^H (black) in core OKT-3 (Zhao et al., 2015); **(H)** Mg/Ca SST data from core MD98-2176 in the western tropical Pacific (Stott et al., 2004); **(I)** $\delta^{18}O$ record from the GISP2 ice core (Stuiver and Grootes, 2000); **(J)** the factor representing the western tropical Pacific (Stott et al., 2004); **(I)** $\delta^{18}O$ record from the GISP2 ice core (Stuiver and Grootes, 2000).

annual SST. Such correlations possibly suggested that OH-GDGTs produced in different seasons have been well mixed in seawater before preservation in sediments (Lü et al., 2015). The core-top RI-OH temperature agreed well with the SSTs in May and December, which most likely reflected subsurface temperatures in the present study (Figure 3B).

Cold Events Identified in the Middle Okinawa Trough

In core 14, a warming trend during the last 8.2 kyr BP with an apparent cold event at 7.4–6.6 kyr BP was found using TEX_{86}^H and RI-OH proxies. Paleoclimate studies have indicated that the cold anomalies in the North Atlantic may have spread signals of rapid climate changes via the westerlies to the Asian monsoon regions (Dykoski et al., 2005; Sun et al., 2005; Wang et al., 2005). However, studies on the Holocene paleoclimate suggested that the tropical Pacific likely controlled the paleotemperature in East Asia via KC variations rather than the North Atlantic (Lim and Fujiki, 2011; Xu et al., 2018). The KC, the northward branch of the North Pacific subtropical gyre, exerts an influence on the exchange of climatic properties of downstream regions in the Pacific Ocean and is a mediator of the Pacific to the OT (Hu et al., 2015). An intensified trend of the KC since the Holocene has been demonstrated (Sun et al., 2005; Xiang et al., 2007; Diekmann et al., 2008; Yamamoto, 2009). These warm and highly saline waters transported by the KC may have significantly increased the SST of the OT (Figure 4J; Zheng et al., 2016). Therefore, the warming of the middle OT may be caused by the enhancement of the Holocene driven by the tropical Pacific during the Holocene. Furthermore, the most remarkable cold event at 7.4–6.6 kyr BP (~7.3 kyr BP) was widely reported in the southern and middle OT (Sun et al., 2005; Zhao et al., 2015; Xu et al., 2018). Since this ~7.3 kyr BP cold event has not been reported in the North Atlantic, factors other than the North Atlantic were likely responsible for the SST decrease in the middle OT (Figure 4I; Stuiver and Grootes, 2000). During the 7.4–6.6 kyr BP period, the KC intensity was weakened (Xiang et al., 2007; Hu et al., 2015), potentially contributing to the decreased temperatures of the middle OT (Figures 4C, J; Zheng et al., 2016).

However, the ~7.3 kyr BP cold event was magnified in the GDGT-related SST records. In core C14, the TEX_{86}^H SST showed a drop of ~5°C at 7.4–6.6 kyr BP (Figure 4C). A similar observation has been reported in the southern OT: the TEX_{86}^H SST dropped approximately 3–14°C during 7.6–6.9 kyr BP, whereas the Mg/Ca and U_{37}^K SST only showed a drop of 1–3°C during this period (Sun et al., 2005; Kubota et al., 2010; Zhao et al., 2015; Xu et al., 2018). Xu et al. (2018) attributed the 7.6–6.9 kyr BP SST drop to an unknown cold event that occurred during KC intensity weakening. However, the cold event during that period may not be solely responsible for such a large amplitude of decline in TEX_{86}^H SST. Global climate change cycles could also have regular influences on the SST of marginal seas (Ho and Laepple, 2015; Ignatov and Gutman, 1999; Debret et al., 2007). Sometimes, the SST of the OT could have suffered from strongly enhanced temperature amplitudes across climate cycles (Debret et al., 2007; Zhao et al., 2014). However, no noticeable long- and short-term climate effect on OT hydrology was reported during the last 7.4–6.6 kyr BP (Bond et al., 1997; Zhao et al., 2014). Factors controlling the abnormal TEX_{86}^H SST drop at ~7.3 kyr BP have not been resolved. In core C14, a tephra layer was found from 532.5 to 442.5 cm (K-Ah tephra; 7.4–6.6 kyr BP). A volcanic eruption that occurred at ~7.3 kyr BP has been identified in southwestern Japan (K-Ah tephra; Kitagawa et al., 1995), suggesting that the core C14 site was likely under the scope of the volcanic eruption (Figure 1). The K-Ah tephra has been



widely reported in sediments from the northern to middle OT (Sun et al., 2005; Kubota et al., 2010; Zheng et al., 2016). Modeling studies on Toba volcano in Sumatra have indicated that volcanic eruptions could result in spreading tephra, blocking sunlight, and thus leading to global or regional temperature cooling (Rampino and Self, 1992; Machida, 1999; Oppenheimer, 2002). For core C14, the sedimentation rate during the last 7.4–6.6 kyr BP is exceptionally high (~126 cm/kyr) compared to a nearby site (~20 cm/kyr; DGKS9604; Yu et al., 2009). Therefore, except for the decrease in KC intensity, the cold event and the extremely high sedimentation rate at ~7.3 kyr in core C14 were probably related to volcanic eruptions. The differences between TEX_{86}^H SST and SST records from the $U_{37}^{K'}$ and Mg/Ca at ~7.3 kyr BP in the OT may result from distinct sensibilities and responses of specific organisms to climate events (Schouten et al., 2013; Steinke et al., 2008 and references therein). However, the specific and detailed mechanism

of the ash impacts on living communities remains unclear and requires further investigation.

Decoupling of the East Asian Summer Monsoon and Paleotemperatures in the Middle OT

The temperature difference between the sea surface and subsurface (ΔT) is sensitive to trace heat and water exchanges, reflecting the depth of the thermocline (DOT) of marginal seas (Jian et al., 2000; Lopes dos Santos et al., 2010; Jia et al., 2012; Yuan et al., 2018). In marginal seas, ΔT has been reported to have a negative relationship with DOT (Jian et al., 2000; Lopes dos Santos et al., 2010; Jia et al., 2012). In the OT, the DOT is mainly controlled by the EAM and KC, which stir the surface of the seawater and cause fluctuations in the DOT. Specifically, the enhancement in the EAM or KC intensities will lead to strengthened mixing of upper water and deepening of the DOT (a reduction in ΔT), and vice versa (Jian et al., 2000; Yamamoto et al., 2013).

In this study, TEX_{86}^H and $U_{37}^{K'}$ temperatures (DGKS9604; Yu et al., 2009) were used as SSTs, whereas RI-OH temperatures tentatively served as subsurface temperatures (Figure 3C). The temperature differences between TEX_{86}^H and RI-OH [$\Delta_{TEX_{86}^H-RI-OH}$ ($^{\circ}C$)] and between $U_{37}^{K'}$ and RI-OH [$\Delta_{U_{37}^{K'}-RI-OH}$ ($^{\circ}C$)] were calculated (Figure 5E). From 8.2 kyr BP to the present, a slight overall decrease was found for $\Delta_{TEX_{86}^H-RI-OH}$ ($^{\circ}C$) and $\Delta_{U_{37}^{K'}-RI-OH}$ ($^{\circ}C$), suggesting an increased mixing of the upper water column in the middle OT (Figure 5E). Studies on the Holocene EASM intensity showed that the EASM intensity reached a maximum at ~7.0 kyr or ~9.0 kyr (Yang et al., 2019; Liu et al., 2015; Wang et al., 2005; Dykoski et al., 2005). However, the EASM is primarily impacted by summer (June) insolation at 30°N, exhibiting a gradually decreasing trend since 8.2 kyr (Figure 5C; Berger and Loutre, 1991). Therefore, the increased mixing of upper water was inconsistent with the EASM intensity, especially over the last 7.0 kyr BP, indicating the decoupling of SSTs in the middle OT with the EASM (Figures 5A–C). In the middle Holocene, a decoupling trend between thermal conditions and precipitation has been revealed by pollen sequences from Taiwan (Liew et al., 2006). For land records, decoupling between temperature warming, which was likely caused by the strong influence of low-latitude warm currents, and the EASM intensities were common during the Holocene (Peterse et al., 2011; Park et al., 2014; Wu et al., 2017). The low-latitude warm currents flowing through the OT, such as the KC, may warm and stir the seawater and overstep the influence of high-latitude climates, causing the decoupling of SSTs in the middle OT with the EASM (Zheng et al., 2014, 2016; Xu et al., 2018).

In the present study, the TEX_{86}^H SST, revealed a gradual warming trend in the middle OT since 8.2 kyr BP (Figure 5F). Modeling of the Holocene climate showed that moderate temperature warming was caused by winter warming that slightly exceeded summer cooling in the tropics (Lorenz et al., 2006). Therefore, the warming trend in the middle OT might be caused by the increased boreal winter insolation at low latitudes (Figures 5D,F).

CONCLUSION

Paleotemperatures were reconstructed for the last 8.2 kyr in the middle Okinawa Trough (OT) using $\text{TEX}_{86}^{\text{H}}$ and RI-OH. IsoGDGTs were mainly derived from marine *Thaumarchaeota*, and $\text{TEX}_{86}^{\text{H}}$ can be used to reconstruct temperatures of the middle OT, as revealed by GDGT-0/crenarchaeol, BIT, %GDGT-2, and MI. $\text{TEX}_{86}^{\text{H}}$ temperatures in this study indicated mean annual SSTs close to the U_{37}^{K} SST of the middle OT. In contrast, RI-OH temperatures were interpreted to represent subsurface temperatures relatively lower than the $\text{TEX}_{86}^{\text{H}}$ and U_{37}^{K} SSTs. The cold event at ~7.3 kyr BP and the general warming trend, as revealed by the $\text{TEX}_{86}^{\text{H}}$ and RI-OH temperatures at 8.2 kyr BP, were attributed to the increasing Kuroshio Current intensity punctuated by a decline at ~7.3 kyr BP. The decreasing temperature differences between $\text{TEX}_{86}^{\text{H}}$ and RI-OH and between U_{37}^{K} and RI-OH indicated the decoupling of the SSTs in the middle OT and the East Asian summer monsoon during 8.2 kyr BP. The magnification of the cold phase in $\text{TEX}_{86}^{\text{H}}$ and the extremely high sedimentation rate at 7.4–6.6 kyr BP were probably partially due to the widespread Kikai-Akahoya tephra (~7.3 kyr).

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**, further inquiries can be directed to the corresponding authors.

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AUTHOR CONTRIBUTIONS

LL and HG was responsible for writing manuscripts. LX and LL conducted tests. ZS and NW provided the studied samples. HG revised texts, and NW and ZS revised texts and provided comments.

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SUPPLEMENTARY MATERIAL

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