Evidence of methane release from Blake Ridge ODP Hole 997A during the Plio-Pleistocene: Benthic foraminifer fauna and total organic carbon

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Methane is a powerful greenhouse gas and may have played a significant role in global climate change in the geological past. Destabilization of gas hydrates, frozen methane stored within the ocean floor sediment and in permafrost, may have provided an important source of methane to the atmosphere. Ocean Drilling Program Hole 997A (water depth 2770 m), situated on the crest of the Blake Outer Ridge, is a potentially large reservoir of gas hydrate. Methane emissions from the Blake Outer Ridge have been reported previously, which has been suggested as a driver for global climate change. Methane at this site is of biogenic origin, produced by the bacterial decomposition of organic matter. We used benthic foraminifer faunal assemblages (>125 μm size fraction) and species diversity, combined with total organic carbon data from Hole 997A, to identify intervals of methane releases during the late Neogene (last 5.4 Ma). We identified a group of benthic foraminifera, which were taken to indicate methane fluxes based on previous work on seep-related benthic foraminifera. We then classified 'seep-related' benthic foraminifera, as well as high organic carbon taxa independent of deep-sea oxygenation. We recognized five intervals of increased abundance of the seep-related benthic foraminifera since last 3.6 Ma representing intervals of methane release, which coincide with intervals of lowered sea level. Changes in benthic foraminifera are more abrupt over the past 3.6 Ma when the northern hemisphere glaciation began to intensify and climate switched to a 41-kyr cycle world.

Keywords: Blake Outer Ridge, benthic foraminifera, total organic carbon, methane release, northern hemisphere glaciation.

THERE is growing realization that methane plays an important role in rapid global warming because of its potential as a greenhouse gas (64 times more than CO_2)[1]. Polar ice provides well-documented records of centennial to millennial scale variability during the Quaternary and Holocene[2,3]. Large amount of methane release from the marine system has been related to the late Palaeocene climate warming[4]. Other workers have related methane release to sea-level changes[5]. There are three major sources for methane in the marine system, viz. microbial, thermogenic and abiogenic. Microbial or biogenic methane production is predominantly found in organic-rich anoxic sediments, especially below the penetration depth of sulphate[6]. Thermogenic methane is produced from the geothermal transformation of organic matter buried in marine sediments[7]. Abiogenic methane is formed at hydrothermal vents by water–rock reactions with hydrogen and CO_2 and during serpentinization on large areas of the seafloor[8].

Gas hydrate, an ice-like solid natural crystal composed of water and methane molecules, may contribute substantial amount of methane to the atmosphere[9,10]. Methane escape from destabilizing gas hydrates has been inferred by highly negative $\delta^{13}C$ values of benthic and planktic foraminifera and organic carbon[4,11–13]. Destabilization of gas hydrate may be triggered by temperature increases in bottom waters, sea-level changes or catastrophic structural collapse of the seafloor or rapid changes in sedimentation rate[4,11,12,14–17]. However, the concept of gas hydrate decomposition as the only explanation for the negative excursions in $\delta^{13}C$ of foraminifera and organic carbon was recently challenged by Milkov *et al.*[18] and Milkov[19] on the basis that the amount of carbon in gas hydrates is likely to be much smaller than previously thought[11].

In this study, we attempt to identify intervals of methane escape at Ocean Drilling Program (ODP) Hole 997A, Blake Ridge, northwestern Atlantic over the past 5.4 m.y. using methane seep- and organic carbon-related benthic foraminifera combined with total organic carbon (TOC) data. The Blake Ridge, the earliest recognized marine gas hydrate province in the northwestern Atlantic having prominent depression probably related to methane seepage associated with mussel/clam beds and warm tubes[20], is an ideal site for the study of methane fluxes at millennial to orbital timescales[4,17,21,22]. Benthic foraminifera are an important component of marine communities, having good potential for reconstructing past climate and oceanic changes owing to their wide distribution, high sensitivity

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to different ecological factors, great morphological diversity, and well-documented fossil record. Recent efforts from cold seep/hydrocarbon environments show that certain species of genera like *Uvigerina*, *Bolivina*, *Bulimina*, *Chilostomella*, *Epistominella*, *Globobulimina* and *Nonionella* can withstand low oxygen stressful environments[13,23–30]. On the other hand, some species are found to be paleoceanographically important, which can be used in identifying high organic carbon environments. Potential methane production rates are higher in organic-rich sediments with a rapid depletion of sulphate within the upper few centimetres, leading to ideal conditions for methanogenesis of organic matter[6].

Benthic foraminifera in methane seep and high organic environments

In recent years, there have been numerous attempts to determine methane fluxes from large methane reservoirs using different proxy records, including benthic foraminafera[4,12,16,17,26,27,29,31–33]. Highly depleted carbon isotope values of benthic and planktic foraminifera, organic carbon and different organic compounds (Diplopetrol, Archaeol) have also been used to detect methane fluxes in the Blake Nose area as well as Santa Barbara Basin[4,12,33,34].

One of the earliest attempts to understand a relation between methane release and benthic foraminifera was that of Wefer *et al.*[23], who used stable isotope composition of *Bolivina seminuda* and *Nonionella auris* to understand their response to methane fluxes. These authors suggested that *N. auris*, which feeds on methane-oxidizing bacteria, could be a reliable indicator of the presence of methane. A close association of *Bolivina ordinaria*, *B. albatrossi*, *Cassidulina neocarinata*, *Gavelinopsis translucens*, *Osangularia rugosa* and *Trifarina bradyi* with seeps was observed at hydrocarbon-seep bacterial mat and hydrocarbon vents in the Gulf of Mexico[24,35]. Some palaeoceanographically important cosmopolitan taxa (*Uvigerina*, *Bolivina*, *Chilostomella*, *Globobulimina* and *Nonionella*) were shown to be present in higher abundances in the seep zone at Eel River, northern California Margin[26]. In the Hydrate Ridge, Oregon, association of living benthic foraminifera near methane vents is higher than the non-venting sites, perhaps attracted by the rich bacterial food supply[13]. *Uvigerina peregrina* is an important species in seep zones at Hydrate Ridge and in several other areas, probably feeding on the rich bacterial source of the seep zone[13]. Hill *et al.*[27] suggested that higher abundance of *Bolivina tumida, Epistominella pacifica* and *U. peregrina*, near the active methane seep of the Santa Barbara Channel, may be a good indicator of high methane concentration. However, Barbieri and Panieri[29] did not observe any benthic foraminiferal evidence of cold-seep origin of the Miocene Terminal Formation of Italy though isotopic and megafaunal evidences do indicate cold-seep origin.

Taking into account the previous observations on benthic foraminifer response to methane fluxes, we surveyed numerous recent studies and found that some species of benthic foraminifera enrich seep-related marine settings, whereas some are closely related to high organic carbon content of marine sediments independent of the oxygen levels. The present study provides an opportunity to support or reject these arguments using relative abundances of dominant deep-sea benthic foraminifera at Hole 997A. While *Bolivina seminuda*, *N. auris*, *B. ordinaria*, *B. albatrossi*, *C. neocarinata* and *T. bradyi* may be related to methane-rich environments, numerous species of *Uvigerina* are closely tied to organic-rich environments independent of oxygen levels[36–41]. For example, high abundances of *U. peregrina* are related to sediments rich in organic carbon rather than to oxygen-depleted waters or methane settings[36,42–44]. Likewise, *Melonis barleeanum*, a significant species at Hole 997A, has been suggested to thrive in an environment rich in altered (oxidized) organic matter with intermediate to high organic flux[45]. Based on these observations and population trends of benthic foraminifera at Hole 997A, we classified *Bolivina paula, Cassidulina carinata, Chilostomella oolina, Fursekoina fusiformis, Globobulimina pacifica, N. auris* and *T. bradyi* as seep-related benthic foraminifera, and *Uvigerina hispido-costata, U. peregrina, U. proboscidea* and *M. barleeanum* as high organic carbon taxa independent of deep-sea oxygenation.

Location and lithology

ODP Hole 997A (Leg 164) is located on the crest of the Blake Outer Ridge (BOR; 31°50.588′N; 75°28.118′W; present-day water depth 2770.1 m; Figure 1), about 200 km off the east coast of the United States, northwest Atlantic[46]. The hole underlies a periphery of the subtropical central gyre and is under the profound influence of the warm saline Gulf Stream as well as the Western Boundary Undercurrent (WBUC), which transported clastic material to the Blake Ridge as sediment drift[47]. The shipboard study suggests a downward increase in sedimentation rate of drift sediments[46]. At present, the hole lies in a tectonically inactive setting close to the passive margin and has not been affected by the late Cenozoic tectonic activity or by the fluid flow along the major faults in the sediments[48]. The WBUC varies on glacial–interglacial timescales, carrying North Atlantic Deep Water (NADW) to the south along the eastern continental margin of North America[49,50]. The NADW production decreases or stops during glacial/cold periods[51]. The enhanced NADW production during warm intervals may have led to a stronger WBUC, bringing more drift sediments to the BOR[50].

The presence of strong Bottom Simulating Reflector (BSR) at Hole 997A indicates an interface between hydrate-bearing sediments above this reflector and sediments with free gas below it. Using different proxies (temperature, interstitial water chloride content, velocity and electrical resistivity data), it is proved that disseminated gas hydrate occurs throughout the sedimentary section between ~190 and 450 m below the seafloor (mbsf), ranging in age from 5.4 to 2.9 Ma[46]. Hole 997A has a penetration of 434.3 mbsf with a core recovery of 343.08 m, and comprises a thick Neogene sediment drift sequence composed of fine-grained, nannofossil-bearing hemipelagic sediments, which accumulated at unusually high rates[46] (Figure 2). The sedimentation rate was 42 m/Ma in the Pleistocene, 49 m/Ma in the late Pliocene, 172 m/Ma in the middle Pliocene and 79 m/Ma in the early Pliocene.

Materials and methods

We analysed 246 samples (at an average interval of 22.22 kyr per sample), each of 10 cubic cm volume from the Plio-Pleistocene interval (last 5.4 Ma). Samples were soaked in water with few drops of diluted H_2O_2 (2%) for 8–12 h and washed over a 63 μm size sieve. The oven-dried (at ~50°C) samples were sieved over a 125 μm sieve and split into suitable aliquots for faunal counts. Approximately 250 to 300 specimens of benthic foraminifera were picked from a suitable aliquot and their relative abundances were calculated. However, in a few cases we could obtain only ten individuals in the entire dried sample. Samples from the gas hydrate zone (5.4–2.9 Ma) have very low benthic population with individuals ranging from 10 to 129. The benthic individuals suddenly increase (reaching up to 1200 individuals) in the younger sediments since ~2.6 Ma onwards. As discussed earlier, we have grouped benthic foraminifera into two groups: seep-related and high-organic carbon-related (Table 1, Figure 3) based on the

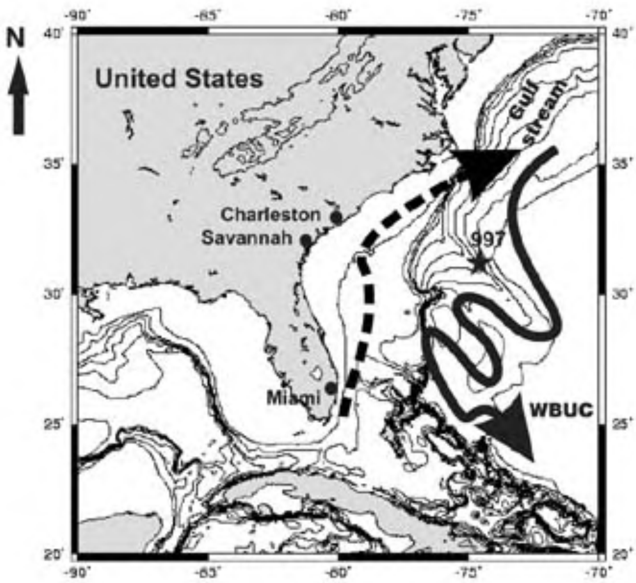

Figure 1. Location of Hole 997A on the Blake Outer Ridge, ~200 km off the east coast of the United States, northwest Atlantic[46]. The hole underlies a periphery of the subtropical central gyre and is under the influence of the warm saline Gulf Stream as well as the Western Boundary Undercurrent (WBUC).

Table 1. Seep (methane)-related and organic carbon-rich benthic foraminifera*

Seep (methane)-related benthic foraminifera
Bolivina paula
Cassidulina carinata
Chilostomella oolina
Fursenkoina fusiformis
Globobulimina pacifica
Nonionella auris
Trifarina bradyi
Organic carbon-rich benthic foraminifera
Uvigerina hispido-costata
Uvigerina peregrina
Uvigerina proboscidea
Melonis barleeanum

*The assignment of seep-related assemblages is based on refs 13, 23, 24, 26–28, 35, whereas that of organic carbon-rich benthic foraminifera is based on refs 36–41.

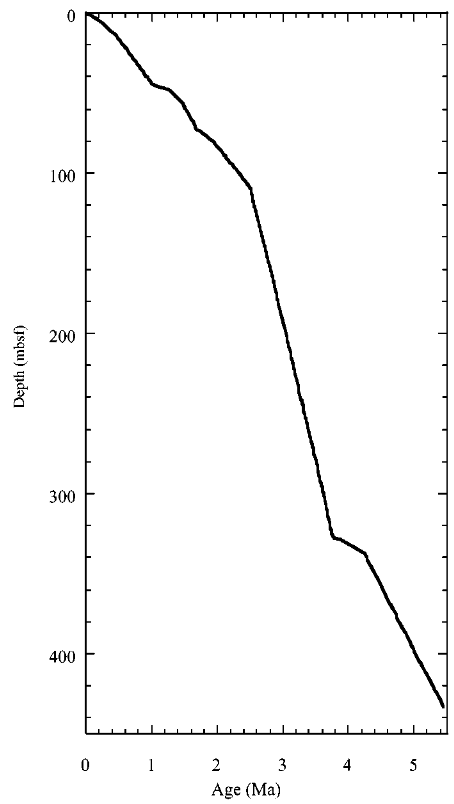

Figure 2. Depth vs age plot based on nannofossil datums[56] and updated to the timescale of Berggren *et al.*[57,58]. Sedimentation rates are higher below 100 m.

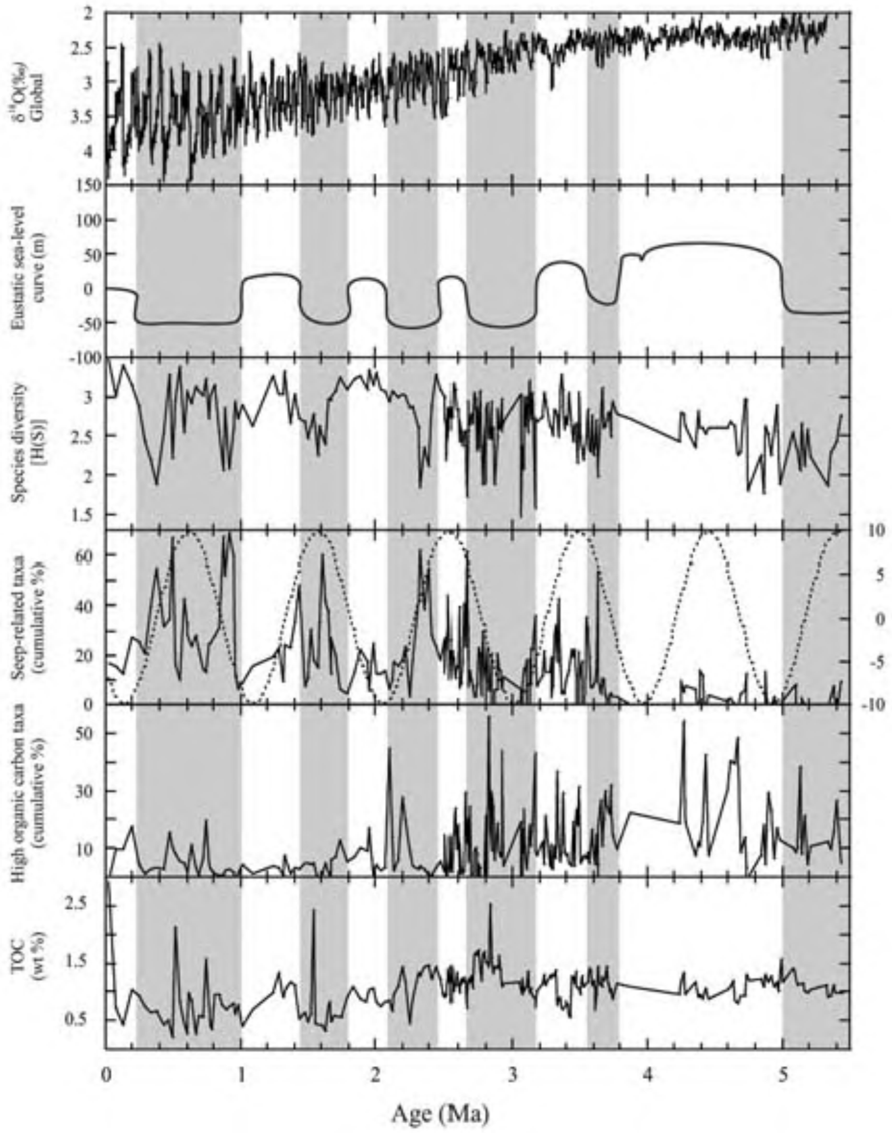

Figure 3. Cumulative abundances of seep-related and high organic carbon benthic foraminifera (Table 1) combined with species diversity [$H(S)$] and total organic carbon (wt %) at Hole 997A. Also plotted are $\delta^{18}O$ values[54] and global eustatic sea-level curve[55]. Shaded bars indicate proposed intervals of methane fluxes inferred from higher percentages of seep-related benthic foraminifera and lowered sea levels. Dotted line in the seep-related taxa panel represents fit of sine function [general model $f(x) = a1*\sin(b1*x + c1)$] using Matlab 7 software. Coefficients ($a1$, $b1$, $c1$) are with 95% confidence level.

studies from different gas seeps, hydrate regions and increased organic flux environments[13,23,24,26–28,35,36,39,42–45,52].

Species diversity [$H(S)$] was calculated using the Shannon–Wiener Diversity Index[53]. TOC was analysed using a TOC Analyzer (TOC-V_{CPH}; Shimadzu Corporation, Japan). The TOC analysis was run using 0.5 g of finely powdered and dried sample, soaked in 50 ml of millipore water with 20 drops of 1 N hydrochloric acid (HCl) and stirred with a magnetic stirrer for 2 h so that the inorganic carbonate is digested and the solution is homogenized. Then 2N HCl and 25% phosphoric acid were further added and sparged for 1.5 min to completely digest inorganic carbon and bring the pH of the solution to 2–3. The machine was standardized using potassium hydrogen phthalate. The calibration curve (best-fit line) was drawn through the scatter of readings of eight standard solutions with different concentrations (within the range 10–500 ppm). For the analysis of each sample we choose 2 to 5 injections, taking average of two readings (which show standard deviation less than 0.1 and coefficient of variation less than 2%) as the final value. The seep-related taxa, a few high organic carbon-related taxa, TOC values (wt %), $H(S)$, and global $\delta^{18}O$ values[54] are plotted along with the eustatic sea-level curve[55] to identify intervals of

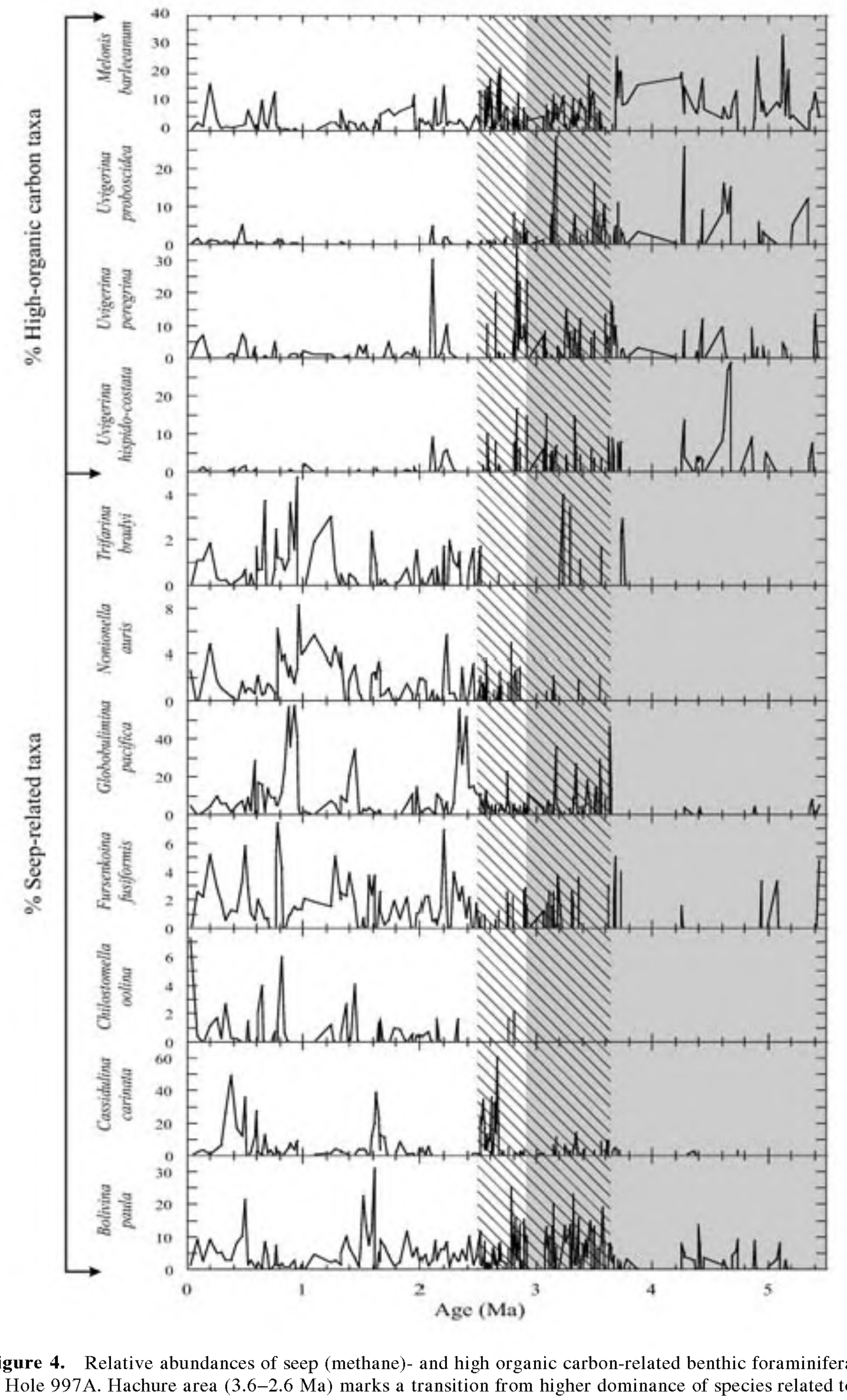

Figure 4. Relative abundances of seep (methane)- and high organic carbon-related benthic foraminifera at Hole 997A. Hachure area (3.6–2.6 Ma) marks a transition from higher dominance of species related to high organic carbon content to increased population of seep (methane)-related species. Shaded area is the present-day gas hydrate zone.

methane seepages and their impact on global climate (Figures 3 and 4). Interpolated ages are based on nannofossil datums[56] and updated to the timescale of Berggren *et al.*[57,58].

Results and discussion

Benthic foraminifera show significant changes corresponding to a change in sediment accumulation rate at

Hole 997A during the past 5.4 Ma. A major transition in benthic foraminifer assemblage occurs across 3.6–2.6 Ma (Figures 3 and 4), coinciding with a major increase in the northern hemisphere glaciation and beginning of the 41-kyr orbital forcing[59–61]. During the early Pliocene at Hole 997A (5.4–3.6 Ma), benthic foraminifera are dominated by high organic carbon taxa suggesting high organic content of the sediment, which could be linked to increased transport of drift sediments by WBUC to BOR during increased production of NADW and its southward flow (Figures 3 and 4). The early Pliocene climate was an interval of climate warmth, higher sea level and increased production of NADW[51,55]. The high organic carbon and increased sedimentation during the early Pliocene to mid Pliocene may have facilitated the formation of gas hydrate at the Blake Ridge[46]. The 3.6 to 2.6 Ma transition is marked by significantly higher sedimentation rate and a change from the dominance of high organic carbon taxa to the dominance of seep-related taxa in the younger interval (Figures 3 and 4). It is important to note that significant abundance of seep-related taxa as well as high organic carbon taxa during this interval indicates enhanced methane seepage, causing extremely stressful environment from a higher organic carbon environment.

The periods of higher percentages of seep-related benthic foraminifera and low species diversity [$H(S)$] at Hole 997A coincide with intervals of lowered sea level (grey bars in Figure 3). The TOC values show a general decrease since 3.6 Ma (Figure 3), during which time surface productivity fluctuated at Hole 997A[47]. Observations at Hole 997A do not support the argument that some of these seep-related species are related to organic-rich sediments. Rather, higher abundances of these species are seen in the interval with low organic carbon sediments (present study) and fluctuating surface productivity[47]. We thus suggest that the intervals of lowered sea level and high percentages of methane seep-related taxa at Hole 997A represent probable zones of methane fluxes that resulted from dissociation of gas hydrates owing to reduction of water column as well as water pressure. The intensity of methane fluxes increased since 3.6 Ma, coinciding with the higher populations of methane-related taxa (Figure 3). Overall, low species diversity in the methane rich zones indicates a stressful marine environment for benthic foraminifera[62].

It has been argued that lowered sea level causes reduction of hydrostatic pressure and destabilizes solid gas hydrate into the gaseous phase[5,63,64], although Kennett *et al.*[12] and Hill *et al.*[27] have linked dissociation of gas hydrate in the Quaternary interstadials to increase in bottom-water temperatures. Likewise, it has been suggested that a large amount of methane was released from the Blake Nose area to the atmosphere during the Late Paleocene Thermal Maximum[4]. Holbrook *et al.*[17], on the other hand, related destabilization of gas hydrate to the creation of gas migration pathways by the formation of sediment waves and related erosion at BOR after 2.5 Ma. Our results support the arguments of Haq[5], Maslin *et al.*[63] and Rothwell *et al.*[64] that the dissociation of gas hydrates at the BOR is related to reduced hydrostatic pressure during intervals of low sea level. The higher frequency of methane fluxes since ~3.6 Ma (increased abundances of seep-related benthic foraminifera) at Hole 997A, suggests a close relationship between methane release and changes in the northern hemisphere glaciation during the late Pliocene–Pleistocene[59,65,66].

Conclusion

The distribution of deep-sea benthic foraminifera combined with other climate proxies at ODP Hole 997A indicates five major episodes of methane releases linked to intense glacial events and lowered sea levels since the last 3.6 Ma, coinciding with increased northern hemisphere glaciation. Species diversity decreased during these intervals of methane fluxes. The transition 3.6–2.6 Ma is marked by more abrupt changes in benthic foraminifera when large continental ice sheets began to grow in the northern hemisphere. Intensification of the northern hemisphere glaciation caused lowering of sea level and allowed solid gas hydrate to dissociate into a gaseous phase by a drop in hydrostatic pressure. This transition is also marked by a change in the benthic assemblage at Hole 997A from organic carbon-rich fauna to methane-related benthic foraminifera.

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