A reversed carbon isotope gradient and Pacific deep water formation during the mid-Pliocene warm period

INTRODUCTORY PARAGRAPH

Geologic intervals of sustained warmth such as the mid-Pliocene warm period can inform our understanding of future climate change, including the long-term consequences of oceanic uptake of anthropogenic carbon. Here we generate new carbon isotope records, and synthesize existing records, to reconstruct Pliocene deep ocean circulation in the Pacific Ocean. We show that the mid-depth carbon isotope gradient in the North Pacific was reversed during the mid-Pliocene in comparison to today, which implies deep-water formation in the subarctic North Pacific Deep Water. An isotopically enabled climate model that simulates this North Pacific Deep Water reproduces a similar carbon isotope pattern. The modelled North Pacific deep ocean dissolved inorganic carbon content decreases slightly, which is counterbalanced by an increase at the surface. Globally the carbon budget increased by ~1.6%. Although the modelled Pliocene ocean maintains a carbon budget similar to the present, the change in deep ocean circulation configuration causes pronounced downstream changes in biogeochemistry.

INTRODUCTION

The ocean is the largest reservoir of exchangeable carbon in the climate system (~38,000 Petagrams Carbon, PgC) and has absorbed approximately a third of anthropogenic carbon emissions [Pachauri et al., 2014]. So far most of this anthropogenic carbon has been absorbed in the upper 700 m of the water column, but there are broad implications for the role of deep ocean circulation in future long-term carbon storage. The deep arm of the meridional overturning circulation moves water masses along the ocean depths, and as these waters age they accumulate dissolved inorganic carbon (DIC) [Rahmstorf, 2002]. The North Pacific acts as an area of carbon storage because it is at the end of the “global conveyor belt” and contains a large amount of DIC. Currently no deep-water formation occurs in the North Pacific because of a strong halocline that prevents deep convection [Warren, 1983]. However, proxy and modelling results suggest that North Pacific deep-water formation may have occurred during past warm periods, potentially impacting the ocean’s ability to store carbon [Kwiek and Ravelo, 1999; Thomas, 2004; Rae et al., 2014; Burls et al., 2017; McKinley et al., 2019].

The mid-Piacenzian stage of the Pliocene (3.264 - 3.025 Ma), often referred to as the mid-Pliocene warm period, is an age of sustained warmth with the global mean surface temperature estimated at 2-3°C above pre-industrial and atmospheric CO₂ (350-400 ppmv) similar to current anthropogenic levels [Haywood et al., 2020; la Vega et al., 2020;McClymont et al., 2020]. As it is a potential analogue for future climate change, there are intense efforts to characterize the mid-Piacenzian through proxy reconstructions and modelling [McClymont et al., 2020]. Many modelling efforts fail to simulate the magnitude of polar amplification and the reduced meridional temperature gradient implied by the proxy reconstructions [Brierley et al., 2015]. As the meridional temperature gradient is tightly coupled to atmospheric heat and water vapor transport, this limitation affects our understanding of climate dynamics necessary to sustain global warmth [Burls and Fedorov, 2014b; Fedorov et al., 2015; Burls and Fedorov, 2017].
In a modelling experiment that alters cloud albedo to simulate a reduced meridional temperature gradient comparable to Pliocene reconstructions, a reduction in northward atmospheric water vapor transport eroded the North Pacific halocline [Burls et al., 2017]. Subsequently, the weakened upper ocean stratification in the subarctic North Pacific enabled active ocean ventilation and deep-water formation. Importantly, the model ran for ~3000 simulation years to allow for the deep ocean to adjust to the Pliocene forcing and the Pacific Meridional Overturning Circulation (PMOC) fully appeared only ~1500 years into the simulation; many climate models are run for ~500-1000 years which is not enough time for the deep ocean to fully equilibrate.

For this study we present new stable isotope data and compile existing records from the Pacific Ocean to reconstruct deep ocean water masses and investigate the spatial extent of a Pliocene PMOC. We compare this $\delta^{13}$C synthesis to an isotopically-enabled climate model (Community Earth System Model, CESM – see Methods) and show good proxy and model agreement in support of the formation of North Pacific Deep Water (NPDW) during the mid-Pliocene warm period. Because of NPDW formation, the DIC concentration decreases in the deep Pacific and increases in the surface ocean, with possible biogeochemically cascading effects such as increased productivity and altered biogeochemical cycles in the Eastern Equatorial Pacific [Lyle and Baldauf, 2015; Shankle et al., 2021]. Overall, there is little modelled change in global carbon storage during the mid-Pliocene warm period.

MAPPING NORTH PACIFIC CIRCULATION WITH CARBON ISOTOPES

Carbon isotopes of DIC ($\delta^{13}$C$_{\text{DIC}}$) trace water mass circulation in the modern ocean. The $\delta^{13}$C of benthic foraminifera shells, which is a proxy for $\delta^{13}$C$_{\text{DIC}}$, can be used to reconstruct water mass circulation in the past [Kroopnick, 1985; Curry and Oppo, 2005; Schmittner et al., 2017]. The ability to trace water masses using $\delta^{13}$C reflects the fact that biological and physical formation processes create different $\delta^{13}$C “end members” where water masses form today. That is, the $\delta^{13}$C values in the surface source areas for deep water formation in the North Atlantic and Southern Ocean are different enough from one another to monitor a water mass sinking and moving through the ocean’s basins.

In the modern ocean, North Atlantic Deep Water (NADW) has a positive $\delta^{13}$C value ($\sim$$+0.6$) and Antarctic Bottom Water (AABW) has a negative $\delta^{13}$C value ($\sim$$-0.2$)[Gu et al., 2020]. As these water masses traverse the ocean depths, they accumulate respired carbon and become more negative in their $\delta^{13}$C values. The most negative $\delta^{13}$C water mass is Pacific Deep Water (PDW, $\sim$$-0.3$, [Kroopnick, 1985]), originally sourced from the South Pacific as a mixture of NADW, Antarctic Intermediate Water (AAIW) and AABW, which reflects the long circulation pathway and respiration of organic carbon.

Shatsky Rise is ideally positioned to monitor changes in North Pacific deep ocean circulation. In the northwest Pacific Ocean, Shatsky Rise is proximate to the western intensification of surface ocean currents [Lozier, 2010] and multiple Ocean Drilling Program (ODP) sites on this bathymetric high allow for a near vertical depth transect of deep Pacific waters. Correlating our new $\delta^{18}$O records (Supplemental Figure 1) to reference curves (e.g., global benthic $\delta^{18}$O stack Probstack) demonstrate good stratigraphic coverage of the middle Pliocene and permit identification of the mid-Pliocene warm period at ODP Sites 1209 and 1210 (2387 and 2574 m water depth, respectively).
Our new δ¹³C data (Supplemental Figure 1), when combined with previously published records [Whitman and Berger, 1993; Mix et al., 1995; Kwiek and Ravelo, 1999; Tian et al., 2002; Jian et al., 2003; Shackleton et al., 2006; Tiedemann et al., 2007; Karas et al., 2009; Venti and Billups, 2012; McClymont et al., 2016; Caballero-Gill et al., 2019] (Table 1), permit us to reconstruct intermediate and deep ocean circulation in the Pacific during the mid-Pliocene warm period (Figure 2 and 3). Undoubtedly there is δ¹³C variability within the mid-Pliocene warm period owing to glacial-interglacial cycles and other sources, but the resolution of the data does not allow for use to characterize this at this time. Although some of the sites included are from continental margins (e.g. California Margin) and marginal seas (e.g. South China Sea) that may be influenced by boundary effects (e.g. Schmittner et al., 2017; Figure 2), we include all the available data here for completeness. Symbols for open ocean sites are outlined in black, while those possibly influenced by boundary effects are outlined in grey. Shatsky Rise is in the open ocean and productivity during the mid-Pliocene warm period was low (Abell et al., 2020; Lam et al., 2021; Venti et al., 2017), as it is today, so the lowering of δ¹³C of epibenthic foraminifera resulting from a phytodetritus layer should not be an issue (i.e. Mackensen et al., 1993).

We calculated an anomaly from pre-industrial by subtracting nearby Ocean Circulation and Carbon Cycling (OC3) late Holocene sediment and modern water column δ¹³C values [Schmittner et al., 2017] from the mid-Pliocene values (Table 1). These anomaly maps (Figure 2 and 3, sediment in diamonds and modern δ¹³C ocean water in squares) show a pattern with positive δ¹³C anomaly values in the North Pacific and a negative δ¹³C anomaly values in the South Pacific. These δ¹³C anomaly patterns suggest changes in PMOC related to either changes in water mass formation distribution (where a water mass forms), character (how biological processes influence δ¹³C) and/or ventilation (how quickly the ocean overturns).

The positive δ¹³C anomaly in the North Pacific between the Pliocene and Late Holocene time slices suggests mid-Pliocene NPDW existed as a mid-depth water mass (core at ~1500 m), similar to modern North Atlantic Deep Water. High CaCO₃ and opal accumulation and preservation in the subarctic North Pacific (Figure 1) also supports younger ventilation ages in the deep ocean [Burls et al., 2017]. In the modern ocean, the North Pacific has little to no CaCO₃ preservation owing to old, acidic water, while opal is undersaturated, accumulating only in areas undergoing deep convection (coastal upwelling, Southern Ocean) where the rain rate is high. Our Pliocene reconstructions (Figure 2 and 3) indicate that NPDW extended to the equator where it met AAIW. AAIW has a negative δ¹³C anomaly that likely reflects changes in the δ¹³C of this Southern Ocean end member, which may be better constrained in the future by new records from recently completed IODP cruises.

### SIMULATING THE IMPACT OF NORTH PACIFIC DEEP WATER FORMATION ON CARBON ISOTOPES

In two fully-coupled climate simulations forced with changes in cloud radiative properties that give rise to large-scale warming patterns resembling Pliocene reconstructions, NPDW formation and a PMOC appear once the deep ocean starts to equilibrate, between ~500 to 1500 year into the simulation. The imposed modifications in cloud radiative properties support reduced meridional and zonal sea surface temperature (SST) patterns resembling that of the Early and mid-Pliocene (mid-Piacenzian) SST reconstructions respectively, which are challenging to simulate.
using standard modelling methods (i.e. Haywood et al., 2013; Haywood et al., 2020), particularly with respect to the reduced gradients of the early Pliocene (Supplemental Figure 2). Both simulations are performed with the Community Earth System Model (CESM) with active biogeochemistry [Moore et al., 2004] and carbon isotopes [Jahn et al., 2015].

These simulations produce a strong and weak PMOC, respectively, and illustrate the impact of changing Pliocene SST gradients on the hydrological cycle and North Pacific Deep Water formation. The Early Pliocene-like simulation is configured similarly to Burls et al. (2017) who described how changes in the hydrological cycle in response to Pliocene SST patterns lead to the erosion of the North Pacific halocline, forming NPDW and a PMOC. The mid-Pliocene-like simulation is configured to align more with mid-Pliocene SST reconstructions (see Methods, Supplemental Figure 2). Although these simulations are similar to Burls et al., 2017, both simulations are now geochemically-enabled and include carbon isotopes to facilitate a pair-wise comparison with benthic foraminifera $\delta^{13}C$ values. The large-scale warming patterns and the associated hydrological cycle responses give rise to surface ocean buoyancy changes that drive weak (strong) PMOC cells within the mid-(Early) Pliocene-like experiments. The impact of a PMOC on $\delta^{13}C_{DIC}$ in both simulations shows spatial similarity with the reconstructed Pacific $\delta^{13}C$ benthic foraminifera values (Figure 2 and 3, Supplemental Figure 3), but the weaker PMOC that develops in the mid-Pliocene-like simulation aligns more closely with the reconstructed mid-Piacenzian $\delta^{13}C$ values. We present both experiments because $CaCO_3$ preservation records from the subarctic North Pacific suggest that the depth of North Pacific Deep Water formation (and therefore likely PMOC strength) fluctuated throughout the Pliocene [Burls et al., 2017], likely in response to variations in SST and density gradients.

Horizontal basin-wide cross sections at 1000-1500, 2200-2800, and 3200-4000 m reveal an east-west structure in $\delta^{13}C$ due to the development of a PMOC (Figure 2, Supplemental Figure 3). Similar to the modern-day Atlantic, the southward flow of the PMOC is concentrated within a western boundary current (i.e. western intensification) where there are high $\delta^{13}C$ values and the eastern return flow has low $\delta^{13}C$ values. Sites 1209 and 1210 were targeted because they are situated near the western boundary, but owing to their depth they are not within the core of the modelled PMOC in the simulations. Nonetheless, when isolating the 2200-2800 m depth range the $\delta^{13}C$ pattern between the data (symbols) and model (contours) largely agrees. That is, Sites 1209 and 1210 have higher $\delta^{13}C$ values because they are near the western boundary and Site 1018 has low $\delta^{13}C$ values owing to the accumulation of respired carbon in the eastern return flow. The $\delta^{13}C$ data largely correspond with the simulated changes (contours) in the age of each water mass, with the PMOC leading to a tongue of younger water centered at about 1500 m, penetrating to about 3500 m depth, and extending southward from a maximum $\delta^{13}C$ value near 50$^\circ$N [Burls et al., 2017]. For the abyssal North Pacific, on the other hand, ages increase relative to pre-industrial control values.

CHANGES IN OCEAN CARBON CYCLE

A modelled increase in ventilation of the deep North Pacific during the mid-Pliocene altered the distribution of ocean carbon globally in comparison to pre-industrial (Supplemental Figure 4 and 5). The North Pacific DIC reservoir decreased in comparison to the pre-industrial
control (Supplemental Figure 6-8) – the deep ocean (>500 m water depth) reservoir decreases by 0.6%, or ~82 Pg C and, interestingly, this is counterbalanced by an increase in the surface ocean (<500 m water depth) of 1% or ~19 Pg C. The overall ocean carbon content increased by 1.6% with an increase of ~533 Pg C (1.6%) and ~57 Pg C (1.2%) in the deep and surface ocean, respectively. (For reference, the modern ocean absorbed ~34 Pg of anthropogenic carbon between 1994 and 2007 [Gruber et al., 2019].)

The reconfiguration of North Pacific deep ocean circulation likely changed the spatial distribution of nutrient availability throughout the global ocean as evidenced by biogenic opal and alkenone mass accumulation rates [Lawrence et al., 2013]. The high latitudes were productive and there was enhanced export production/preservation, which is particularly evident in the North Pacific where reduced stratification brought nutrients to the surface and supported opal production/preservation [Figure 2, Haug et al., 1999; Burls et al., 2017]. Incomplete nutrient utilization [Haug et al., 1999] in the source water regions of NPDW may have exported excess nutrients to other regions. For instance, this Pliocene deep ocean circulation configuration and carbon distribution had knock-on effects and altered biogeochemical cycles in the eastern equatorial Pacific (Shankle et al., 2021) and increased productivity [Lyle and Baldauf, 2015]. With the onset of Northern Hemisphere glaciation, the PMOC shut down (Burls et al., 2017) and North Pacific deep ventilation ceased, as reflected in the CaCO$_3$ (Figure 1b) and opal accumulation (Figure 1c) at Site 882, Haug et al., 1999). Productivity shifted from high latitudes to lower latitudes during glaciations [Lawrence et al., 2013] and deep ocean transport increased between the Pacific and Atlantic Basins (Woodard et al., 2014).

An increase in surface ocean DIC may have contributed to the higher than pre-industrial CO$_2$ levels during the mid-Pliocene. High resolution CO$_2$ estimates show the average atmospheric CO$_2$ level was ~360 ppm during the mid-Piacenzian [la Vega et al., 2020]; higher DIC relative to pre-industrial may have caused a greater CO$_2$ flux from the ocean to the atmosphere and likely contributed to mid-Pliocene high CO$_2$.

In summary, the reconstructed carbon isotope gradients, along with isotope-enabled modelling, provide strong support for a major reorganization of the global ocean conveyor belt during the warm Pliocene, with an active PMOC cell having large impacts on Pacific Ocean ventilation and global carbon distribution.

MATERIALS AND METHODS

Site Locations

Ocean Drilling Program Sites 1209 (32°39.1001’N, 158°30.3560’E, 2698 m water depth) and 1210 (32°13.4123’N, 158°15.5618’E, 2585 m water depth) are located on the Shatsky Rise in the Northwest Pacific Ocean. Few sites in the Northwest Pacific Ocean have been drilled by the International Ocean Discovery Program; these sites on the Shatsky Rise are the only sites proximal to the modeled North Pacific Deep Water with the foraminifera preservation necessary for stable isotope reconstructions during the Pliocene.

Sample Preparation and Stable Isotope Analyses
Sediment samples were washed and picked for the benthic foraminifera *Cibicidoides wuellerstorfi* (>150 μm size fraction). Approximately 3-5 foraminifera were analyzed for oxygen and carbon stable isotope analyses on the REDACTED. Long-term analytical reproducibility for NBS-19 is ±0.06 and 0.05 (1-standard deviation) for carbon and oxygen isotopes, respectively.

Age model

Orbitally tuned age models for Sites 1209 and 1210 were constructed using HMM-Stack [Lin et al., 2014; Butcher et al., 2017] to align the benthic oxygen isotope records with Prob-stack [Ahn et al., 2017], which is an updated version of LR04 [Lisiecki and Raymo, 2005]. Sampling resolution is one sample every ~3.8 ka and ~5.2 ka for Sites 1209 and 1210, respectively.

Calculating Pliocene to Core Tops and Pre-industrial Water Column δ¹³C Anomalies

To calculate a δ¹³C anomaly from pre-industrial for the mid- and Early Pliocene we used the Ocean Circulation and Carbon Cycling (OC3) database [Schmittner et al., 2017]. The OC3 database includes pre-industrial δ¹³C natural ocean water column bottle data and Late Holocene δ¹³C from the benthic foraminifera genus *Cibicides* (without species-specific adjustments). We used the OC3’s horizontal great-circle distance (Δd) and the vertical distance (Δz) search options to identify pre-industrial samples closest to the drilled site locations. We started with OC3’s “conservative” option (Δd = 500 km, Δz = z/10) and expanded to OC3’s “liberal” option (Δd = 1000 km, Δz = z/5) as necessary. In some instances, it was necessary to widen the great-circle distance further (Table 1). The published Pliocene δ¹³C values were adjusted using species-specific corrections (i.e. *Uvigerina* spp. were adjusted by 0.90‰ after [Duplessy et al., 1984]). The anomaly was calculated by subtracting pre-industrial OC3 δ¹³C estimates from the mid- and Early Pliocene δ¹³C values (i.e. Pliocene – pre-industrial).

Modelling Methods

Three fully-coupled simulations, referred to as the Pre-industrial Control, mid-Pliocene-like (mid-Piacenzian) and Early-Pliocene-like experiments respectively, were performed using the Community Earth System Model (CESM) v1.2.2.2. All simulations were run with active biogeochemistry [Moore et al., 2004] and carbon isotopes [Jahn et al., 2015]. Given the multi-millennial timescale over which the deep ocean and biogeochemical cycles equilibrate, and the computational cost associated with running a fully-coupled climate model with numerous tracers in the ocean component, a CESM version with a relatively low horizontal resolution and CAM4 as the atmospheric component (also referred to as CCSM4) is used. This CESM configuration has atmosphere and land components on a T31 spectral grid (horizontal grid of 3.75 x 3.75 degrees), while the ocean and sea ice components employ a tri-polar grid with a resolution that ranges from 3 degrees near the poles to 1 degree near the equator (see Shields et al., [2012] for further details). Each simulation was initialized from modern ocean conditions and run for 3000 years.

The pre-industrial simulation was run using the default “B_1850_BGC-BDRD” component set in which atmospheric CO₂ is set at 284.7ppmv. The only exception to the default setup being the inclusion of the carbon isotope code in which atmospheric δ¹³C values are set to -6.379. Spatial warming patterns resembling reconstructed mid- and Early Pliocene warming patterns (Supplemental Figure 2) were obtained by changing the meridional cloud albedo gradient within these experiments. For the Early Pliocene-like experiment the liquid water path was reduced
polewards of 15°N&S by 60% while the ice and liquid water paths were increased equatorward of 15°N&S by 240%, but only in the shortwave radiation scheme. These changes act to increase cloud albedo in the tropics and reduce it in the extra tropics – the exact percent by which the LWP and IWPs are scaled correspond to experiment 16 in Burls and Fedorov [2014b]. A more detailed analysis of the applicability of this experimental design to Early Pliocene conditions is detailed in Burls and Fedorov [2014a], also see the method section of Burls et al. [2017] for an in-depth discussion of the framework and previous literature motivating this approach. This simply reproduces the changes in the meridional structure of net cloud radiative forcing required to reproduce Pliocene warming patterns, rather than the exact mechanism which could have occurred via shortwave, longwave, or both. These radiative forcing changes may have been realized by changes in a number of cloud properties in addition to liquid and ice water content and sustained by different Pliocene atmospheric aerosol concentrations or unresolved cloud feedbacks to elevated CO₂ levels during the Pliocene. The appeal of this approach is that the warming patterns are largely reproduced and their impact on the hydrological cycle and surface buoyancy gradient that affect the meridional overturning circulation simulated. Note that while in the Early Pliocene-like experiment CO₂ was set to 280ppm in the atmospheric component (the global warming is supported instead by the cloud albedo changes and the feedbacks that they invoke), atmospheric CO₂ was set to 400ppm for the ocean biogeochemistry and carbon isotope components. Using pre-industrial values of CO₂ in the atmospheric model is justified by the fact that the imposed modifications in cloud radiative properties dominate the simulated climatic changes. Likewise, modern paleogeography was used for all three experiments, as potential changes in, for example, ocean throughways would have very minor effects in comparison (e.g. Brierley and Fedorov [2016]). For the mid-Pliocene-like experiment, in order to achieve the more modest reduction of the large-scale SST gradients suggested by the proxy data, the liquid water path was reduced poleward of 15°N&S by 50% while the ice and liquid water paths were increased equatorward of 15°N&S by 240% while atmospheric CO₂ was set to 400ppm in an attempt to be more physically consistent (as mentioned above, in reality Pliocene warmth was likely maintained by elevated Pliocene CO₂ levels with the imposed cloud radiative forcing changes resulting from unresolved cloud feedbacks to the elevated CO₂ levels or different Pliocene atmospheric aerosol concentrations or some combination of both.

We used previously-published alkenone records (Supplemental) and the core tops/pre-industrial SST values [Tierney et al., 2019] to estimate SST anomalies for the mid-Pliocene and Early Pliocene. In general, there is good agreement between the estimated and modelled SSTs showing greater warming at high latitudes and upwelling regions during the mid-Pliocene and Early Pliocene.

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DATA AVAILABILITY STATEMENT: The stable isotope data generated here for Shatsky Rise and those compiled for the spatial analyses are available at Pangea doi:XXXX

FIGURE CAPTIONS

Figure 1: Benthic isotope records of Prob-stack (A), Site 882 CaCO$_3$ MAR (B), and Site 882 Opal MAR (C). The grey shading indicates the mid-Pliocene warm period.

Figure 2: Reconstructed benthic $\delta^{13}$C and modelled (shaded contours in ‰) anomalies for the mid-Pliocene warm period (3.264 - 3.025 Ma). The benthic $\delta^{13}$C anomaly (‰) is calculated from the OC3 database [Schmittner et al., 2017] with Late Holocene core top (diamonds) and modern $\delta^{13}$C ocean water (squares) values. Horizontal basin-wide $\delta^{13}$C anomalies for 1000-1500 m (A), 2200-2800 m (B), and 3200-4000 m (C) water depths. The largest anomaly is found in the west due to a deep western boundary current (e.g. Burls et al. [2017]) related to western intensification of ocean circulation.

Figure 3: Observed benthic $\delta^{13}$C and modelled zonal mean (shaded contours in ‰) anomalies for the mid-Pliocene warm period (3.264 - 3.025 Ma). The vertical cross section across the Pacific shows the core and spatial extent of the PMOC.

Table 1: Site information including site name, longitude, latitude and bottom water depth (m). OC3 data including pre-industrial $\delta^{13}$C natural ocean water column bottle data and Late Holocene $\delta^{13}$C from the benthic foraminifera genus Cibicides with the relevant search parameters for each site. Mid-Pliocene (mid-Piacenzian) warm period $\delta^{13}$C values, standard deviation and number of data points within the interval. Calculated anomaly (mid-Pliocene warm period minus pre-industrial/Late Holocene). References used in the compilation.

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