Pteropods are among the first responders to ocean acidification and warming, but have not yet been widely explored as carriers of marine paleoenvironmental signals. In order to characterize the stable isotopic composition of aragonitic pteropod shells and their variation in response to climate change parameters, such as seawater temperature, pteropod shells (*Heliconoides inflatus*) were collected along a latitudinal transect in the Atlantic Ocean (31° N to 38° S). Comparison of shell oxygen isotopic composition to depth changes in the calculated aragonite equilibrium oxygen isotope values implies shallow calcification depths for *H. inflatus* (75 m). This species is therefore a good potential proxy carrier for past variations in surface ocean properties. Furthermore, we identified pteropod shells to be excellent recorders of climate change, as carbonate ion concentration and temperature in the upper water column have dominant influences on pteropod shell carbon and oxygen isotopic composition. These results, in combination with a broad distribution and high abundance, make the pteropod species studied here, *H. inflatus*, a promising new proxy carrier in paleoceanography.

Assessing the future impact of ocean acidification, the decline in oceanic pH due to anthropogenic CO₂ emissions, on marine ecosystems is difficult, as the complexity of ecosystems cannot be easily replicated in laboratory experiments. However, long-term evidence for ocean acidification and the associated responses of marine calcifiers can be found in the geological record, as past ocean temperature and chemistry can be derived from fossil calcium carbonate shells, as well as ecosystem responses such as species richness and net calcification. One prominent and straightforward candidate for such an approximation (proxy) of past conditions is the oxygen isotopic composition (δ¹⁸O), which reflects ocean temperature. It has been demonstrated in inorganic precipitation studies as well as in direct measurements of biogenically produced calcium carbonate, that the calcium carbonate-water oxygen isotopic equilibrium is determined by temperature and (sea)water δ¹⁸O (δ¹⁸OSW; see Table 1 for notations). This observation applies to both calcite and aragonite, with an aragonite-calcite fractionation on the order of 0.7 to 0.9‰. Most studies have analyzed the calcite produced by foraminifera in this context. Similar to foraminifera, pteropods secrete aragonite close to the calcium carbonate-seawater δ¹⁸O equilibrium, making them ideal candidates to study their oxygen isotopic composition. Pteropods are marine holoplanktonic gastropods inhabiting epipelagic and mesopelagic waters, down to >1000 m depths. Although pteropods are rarely explored as possible oceanographic proxy carriers compared to foraminifera, several aspects of their biology make them interesting targets. The approximately one year life cycle and diel vertical migrations of pteropods may yield a more integrative proxy record across epipelagic and mesopelagic water masses and across seasons in comparison to foraminifera, which are characterized by a shorter life span and potentially shallower calcification depths. Calcification depths of recent pteropods can be estimated by comparing the oxygen isotopic composition to the theoretical δ¹⁸O of aragonite precipitated in equilibrium with the seawater (δ¹⁸Oₘₛ), which is calculated from temperature profiles and δ¹⁸OSW. Temperature at the time of calcification can be reconstructed directly from δ¹⁸OPTERO, the oxygen isotopic composition of pteropod shells (recent or fossil).

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1 Institute of Geosciences, Christian-Albrechts-Universität zu Kiel, Ludewig-Meyn-Str. 10, 24118, Kiel, Germany.
2 Naturalis Biodiversity Center, P.O. Box 9517, 2300 RA, Leiden, The Netherlands.
3 Institute for Biodiversity and Ecosystem Dynamics (IBED), University of Amsterdam, P.O. Box 94248, 1090 GE, Amsterdam, The Netherlands.
4 Leibniz-Labor für Altersbestimmung und Isotopenforschung, Christian-Albrechts-Universität zu Kiel, Max-Eyth-Str. 11–13, 24118, Kiel, Germany.
5 Plymouth Marine Laboratory, Plymouth, PL1 3DH, United Kingdom.
6 Department of Oceanography, University of Hawai‘i at Mānoa, 1000 Pope Road, Honolulu, HI, 96822, USA. Correspondence and requests for materials should be addressed to N.K. (email: nina.keul@gmail.com)
δ¹³C in calcium carbonate shells is assumed to be in equilibrium with, or offset by a constant amount from δ¹³CDIC, the carbon isotopic composition of dissolved inorganic carbon (DIC, the sum of CO₂(aq), H₂CO₃, HCO₃⁻, and CO₃²⁻). However, several studies found discrepancies for other groups of calcifiers, e.g. foraminifera²⁰, as physiological processes, such as the respiration of symbionts in foraminifera, influence shell δ¹³C²¹. Juranek and colleagues²² found a correlation between pteropod δ¹³Cptero and carbonate ion concentration and hypothesized that this was caused either by a carbonate ion dependence, also demonstrated for foraminifera²³, or by the influence of temperature on metabolic CO₂ incorporation.

The life cycle of the pteropod species Helicocnoides inflatus (d’Orbigny, 1834¹⁴), formerly and more commonly known as Limacina inflata, has been estimated to be approximately 7–9 months, with a maximum life span of about one year²⁴,²⁵. Reproduction occurs continuously throughout the year, with females retaining developing embryos in the mantle cavity up to a size of about 70 μm until release of the veliger larvae²⁶. Sediment trap studies revealed high interannual variability in the abundance of H. inflatus as well as a pronounced seasonal cycle in abundance, e.g. in the Sargasso Sea²⁶. Here, the highest flux of shells was found during summer, whereas the flux throughout the rest of the year was very low²⁶,²⁷. While H. inflatus has been found as deep as 1000 m, this species primarily occurs in the upper water column²⁴. Seasonal shifts in pteropod depth habitat were also reported for the Sargasso Sea, with H. inflatus preferring shallower waters in the fall (100 to 250 m) than in the spring/early summer (200–400 m)²⁸. Diel vertical migration, common in pteropods, has been described for H. inflatus, with most of the population below 200 m during the day and highest night-time abundance in the upper 75 m²⁹. The species also undergoes ontogenetic migration: juveniles tend to stay in surface waters, whereas adults are mainly found in deeper waters³⁰.

Here we present δ¹⁸Optero and δ¹³Cptero measurements of Helicocnoides inflatus shells to assess the potential of pteropods to serve as proxies of seawater temperature (via δ¹⁸O) and carbonate ion concentration (via δ¹³C). We particularly focus on the calcification depth of the species and characterize environmental controls on shell composition. Our material was collected along a meridional transect in the Atlantic Ocean, ranging from 31° N to 38° S. This provides an unique opportunity to assess calcification depths across the entire Atlantic basin, in contrast to previous sediment trap studies that were conducted within a single oceanographic context¹²,¹⁶,¹⁸,¹⁹. Furthermore, the broad spatial scope of the study allows us to assess different environmental parameters as controls on the stable isotopic composition in pteropod shells, as oceanographic conditions change significantly over such a large latitudinal range. The calibrations established here will be of use to the ocean acidification and paleo-oceanographic community, as the studied species, H. inflatus, occurs in high abundance in sediments worldwide³⁰, for instance, in the Central and South Atlantic³¹ or the Caribbean Sea³².

**Results**

**Surface distribution of oceanographic parameters.** The warmest surface temperatures in the study area of the Atlantic Ocean, up to ~30 °C, usually occurred in October/November just north of the equator (around 10° N; Fig. 1a). Temperature decreased gradually both north and south of this maximum to approximately 12 °C at 40° S, where our southernmost station (station 66) was located. The surface salinity distribution mimicked this general latitudinal zonation (Fig. 1b), however, at the latitude of the temperature maximum, low salinities of 35 to 36 prevailed. Highest surface salinities (37–37.5) occurred in two locations west of 30° W: one in the North Atlantic at around 25° N and one in the South Atlantic at around 20° S. The three southernmost stations (stations 60, 62, 66) lie in an area characterized by mean salinities between 35 and 36. Theoretical values for δ¹⁸O of aragonite (δ¹⁸Oara), taking into account δ¹⁸O of DIC in seawater and ambient temperatures, as well as values for δ¹³CDIC were calculated for surface waters (Fig. 1c, d; see Methods and Table 1 for notations). The strong influence of temperature on the surface distribution of δ¹⁸Oara is evident from the latitudinal zonation of this parameter (Fig. 1c). Lowest surface δ¹⁸Oara values (~−1.5‰) occurred just north of the equator at 10° N, in the same area where temperature was highest and salinity lowest. From this minimum, surface δ¹⁸Oara increased continuously to the north and to the south until highest values were reached in the southernmost part of the transect (stations 62 and 66: ~+2‰). The distribution of δ¹³CDIC in surface waters also exhibited latitudinal zonation, with highest values (~+2‰) occurring at the equator around 0° W and in the Western Atlantic south of 40° S (Fig. 1d). From the equator, surface values gradually decreased towards both the north and south with lowest surface δ¹³CDIC values in the North Eastern Atlantic.

**δ¹⁸O in the water column and in pteropod shells.** Latitudinal variations in calculated δ¹⁸Oara for equilibrium aragonite values at different depths in the water column reflect strong thermal stratification between 20° N and 20° S, with a maximum in stratification around the equator (4° N), where values ranged from about −1.1‰ at the surface to +2.6‰ at 300 m depth (Fig. 2a). In comparison, δ¹⁸Oara in the southern temperate region, e.g. at 34° S (station 62), was less variable across the water column (from +2.1‰ at the surface to +1.5‰ at 300 m depth).
Individual pteropod $\delta^{18}O_{\text{ptero}}$ values varied with latitude (see Table 2, S2 and Fig. 2a): the lowest values (minimum: $-0.44\%o$) occurred at the equator and increased towards higher latitudes (maximum: $+2.11\%o$). Variability between specimens at each station was close to instrument precision, with an average standard deviation of 0.14 (range 0.05–0.28‰). The pteropod oxygen isotopic composition measured here was comparable to the study of Juranek and coworkers$^{12}$ on the same species ($+0.15$ to $+2.04\%o$), in which specimens from a one year-sediment trap in the Sargasso Sea were analyzed. Another study, however, reported more depleted oxygen isotopic values (approximately by 0.8‰) for the same species from the Sargasso Sea, which likely derived from low temperature ashing of the samples before isotopic measurement$^{16}$.

Average pteropod $\delta^{18}O_{\text{ptero}}$ values were similar to those calculated near the 75m-depth isopleth of $\delta^{18}O_{\text{ara}}$ (Fig. 2a), with individual measurements corresponding to values typically found between the surface and a maximum depth.
of 150 m. This result was also illustrated by the strong positive relationship between pteropod δ¹⁸Oₚtero and δ¹⁸Oara in the upper 75 m of the water column (p < 0.05; R² = 0.89 to 0.91; Table 3, top). Likewise, we found a strong linear relationship between temperature and pteropod δ¹⁸Oₚtero (p < 0.05; R² = 0.87 to 0.86) in the upper 75 m of the water column (Fig. 3, Table 3, top).

δ¹⁸O in the water column and in pteropod shells. Pteropod shell δ¹³Cₚtero showed little variation in samples that were collected between 32° N and 26° S (+0.77‰ ± 0.23‰), however, δ¹³Cₚtero increased sharply with mean values of +1.28, +1.60, and +1.97‰ at the southernmost stations 60, 62, and 66, respectively (Fig. 2b, Table 2 and S2). Calculated variation of δ¹³CₐDIC in the water column was highest at low latitudes, where the variability of δ¹³Cₚtero in pteropod shells was very low (Fig. 2b). Pteropod shell δ¹³Cₚtero at most stations was lower than calculated δ¹³CₐDIC in the water column. This result was previously observed in pteropod shells that calcified in shallow waters12, and was attributed to the carbonate ion effect, where higher carbonate ion concentrations caused lower δ¹³Cₚtero values than in equilibrium with δ¹³CₐDIC (see Discussion). We find the strongest linear relationship between pteropod δ¹³Cₚtero and carbonate ion concentration in the upper water column (Fig. 4a), with a negative regression of δ¹³Cₚtero = −0.02 * Carbonate (50 m) + 5.31 (p < 0.05, R² = 0.92; Table 3, middle). Pteropod shells from the southernmost stations have high δ¹³Cₚtero values, where phytoplankton standing stock was high in surface waters (see Discussion below and Fig. 4b). We observe a significant positive relationship between δ¹³Cₚtero and δ¹⁸Oₚtero in pteropod shells (p < 0.05, R² = 0.68; Fig. 5). Interestingly, this relationship is mostly influenced by measurements performed on shells from the three southernmost stations (60, 62 & 66; indicated by open circles in Fig. 5): omitting these stations renders the relationship statistically non-significant (p > 0.05). Comparison of our results to previous findings on the same species16,18,19 revealed that the slope was similar to prior work (Fig. 5). However, the δ¹³Cₚtero values presented here cover a broader range of δ¹³Cₚtero in shells of Heliconoides inflatus pteropods. (a) δ¹⁸Oₚtero (inversed y-scale) and (b) δ¹³Cₚtero along a meridional transect in the Atlantic Ocean. Measurements on individual shells are shown (open circles) and closed circles represent averaged values. δ¹⁸Oara and δ¹³CₐDIC isopleths for different depths in the water column are shown by colored lines as indicated by the legend.

Table 2. Average δ¹⁸Oₚtero and δ¹³Cₚtero (in ‰) based on measurements of individual pteropod shells (N = number of shells measured; SD = Standard Deviation, n.a. = not available, as n=2).

| CTD Station | Latitude | Longitude | N | δ¹⁸Oₚtero | SD | δ¹³Cₚtero | SD |
|-------------|----------|-----------|---|-----------|----|-----------|----|
| 15          | 32.02    | −30.74    | 3 | 1.06      | 0.05 | 0.82      | 0.08 |
| 23          | 23.16    | −40.60    | 3 | 0.43      | 0.20 | 0.45      | 0.16 |
| 25          | 20.57    | −38.59    | 3 | 0.18      | 0.09 | 0.68      | 0.10 |
| 37          | 4.03     | −26.47    | 3 | −0.13     | 0.28 | 0.66      | 0.20 |
| 42          | −4.62    | −25.00    | 3 | 0.04      | 0.11 | 0.77      | 0.04 |
| 48          | −15.29   | −25.05    | 3 | 0.41      | 0.14 | 0.56      | 0.08 |
| 50          | −18.52   | −25.10    | 2 | 0.77      | n.a. | 0.84      | n.a. |
| 56          | −25.75   | −24.99    | 3 | 1.08      | 0.17 | 0.83      | 0.10 |
| 60          | −30.20   | −27.92    | 3 | 0.95      | 0.25 | 1.28      | 0.12 |
| 62          | −34.15   | −33.49    | 3 | 1.40      | 0.07 | 1.60      | 0.17 |
| 66          | −38.11   | −39.33    | 3 | 2.06      | 0.07 | 1.97      | 0.21 |
Table 3. Linear relationships between average pteropod shell $\delta^{18}O_{\text{ptero}}$ (top), $\delta^{13}C_{\text{ptero}}$ (middle and bottom) and water parameters: carbonate ion concentration ($\mu$mol/kg), temperature (°Celsius), salinity, Chlorophyll $a$ (Chl $a$), $\delta^{18}O_{\text{sal}}$ (‰) and $\delta^{13}C_{\text{DIC}}$ (‰). Regressions were performed against parameters at specific depths; the adjusted $R^2$ is reported when $p < 0.05$. n.s. indicates non-significant linear regressions ($p > 0.05$). Linear regressions for correlations with water parameters at 50 m depths are listed above the tables, values in parentheses are the respective standard errors.

than observed in previous studies that were located in environments with less oceanographic variation (e.g., sediment trap studies, Sargasso Sea).

Discussion

Deriving past ocean temperature and chemistry from fossil pteropod shells provides a wealth of information about past climate change events. The present study shows that the species $H$. inflatus is well suited for paleo-reconstructions, as the stable isotopic composition of their shells can be used to track two climate change indicators: $\delta^{18}O_{\text{ptero}}$ records temperature (Fig. 3) and carbonate ion concentration is traced by $\delta^{13}C_{\text{ptero}}$ (Fig. 4a). Both proxies can be measured simultaneously on a single pteropod shell, making pteropods particularly promising new proxy carriers. Furthermore, we demonstrate that $H$. inflatus records latitudinal ranges of surface water parameters, as suggested previously from sediment trap studies at a local scale$^{21,16}$. We confirm shallow water calcification of this species for a large area of the Atlantic Ocean (31° N to 38° S), which, in combination with its basin-wide distribution, renders $H$. inflatus an ideal candidate for proxy reconstructions.
Based on a comparison between the oxygen isotopic composition of pteropod shells and that of seawater, we observed no systematic latitudinal variation in calcification depth (Fig. 2a). Oxygen isotopic values for *H. inflatus* strongly correlated with $\delta^{18}O_{\text{ara}}$ equilibrium values in the upper 75 m of the water column (Fig. 2a, Table 3, top). These findings corroborate observations from other studies on the same species in the Sargasso Sea, suggesting calcification from 50–250 m depths\(^{12,16}\). Such shallow calcification depths are in contrast to reported preferential occurrences of *H. inflatus* in deeper waters (to 600 m) in the Sargasso Sea\(^ {17}\). One possible explanation is that pteropods preferentially calcify near the surface, where calcification is energetically favored due to warmer temperatures, affecting calcium carbonate saturation. Furthermore, most chlorophyll $a$, and thus potential food resources, occurs in the upper water column, which may be another reason why *H. inflatus* calcifies in shallow waters.

Pteropod shells are produced over several months, therefore reflecting the sum of environmental conditions experienced throughout the animal's life. Thus, single-shell measurements, as presented here, are an average of these conditions, with a bias toward the more recently calcified material, as this makes up the largest part of the shell\(^ {16}\). Pteropods alter their depth habitat daily, seasonally, and ontogenetically\(^ {9,17}\). Consequently, the aragonite of a single pteropod shell could have been produced across a range of depths. Accordingly, the estimated calcification depth of about 75 m may be the average of varying isotopic signatures from different calcification depths.

Figure 3. $\delta^{18}O_{\text{ptero}}$ versus temperature at 50 m depth. We observed a significant negative relationship between $\delta^{18}O_{\text{ptero}}$ and temperature ($p < 0.05$; $R^2 = 0.86$): $\delta^{18}O_{\text{ptero}} = -0.140 (±0.018) \times \text{Temperature} + 3.919 (±0.404)$. Values in brackets indicate the standard error. Average standard deviation is depicted by the error bar in the lower left corner.

Figure 4. Relationship between $\delta^{13}C_{\text{ptero}}$ and water parameters. (a) $\delta^{13}C_{\text{ptero}}$ versus average carbonate ion concentration in the upper 80 m. The carbonate ion effect on $\delta^{13}C_{\text{ptero}}$ is seen in the negative relationship between carbonate ion concentration and $\delta^{13}C_{\text{ptero}}$ (solid line). Due to high phytoplankton productivity related $^{13}C$ enrichment, stations 60, 62 & 66 were removed; the horizontal line indicates the application limit of the calibration (see text). (b) $\delta^{13}C_{\text{ptero}}$ versus chlorophyll $a$ (Chl. $a$) fluorescence. Stations 60, 62 & 66 marked in red and station 60 is marked by an asterisk (see Discussion). Error bars in the lower left corner indicate average standard deviation.
the effects of 13C enrichment via photosynthesis and the carbonate ion effect on pteropod water column) in the south subtropical convergence province (stations 62 and 66, see ref. 23). Photosynthesis Phytoplankton standing stock (chlorophyll concentration) was very high in surface waters (upper 75 m of the water column, rather than in deeper waters12,16. The material of a single pteropod shell is probably also the product of several seasons, as the average life span of several pteropod species is on the order of one year15. Sediment H sedimentation or (sub)species (referred to as waters (south of 34°S) along a similar transect (AMT24) were reported to be morphologically distinct, having element compositional differences can still be caused by depth habitat preferences of different sub-species 26. All fluorescence and δacross most of the transect (average δlate strongly with seasonal temperature variations in the water column. The absence of an offset in time between δptero and δH as a transitional station: it is located in the oligotrophic south Atlantic gyral province with low chlorophyll a concentrations and H. inflatus specimens from southern temperate waters (south of 34°S) along a similar transect (AMT24) were reported to be morphologically distinct, having coarser and thicker shells than specimens from the rest of the transect, and thus may represent a distinct population or (sub)species (referred to as H. inflatus S)24. In these respects, station 60 (30.20 °S) should be regarded as a transitional station: it is located in the oligotrophic south Atlantic gyral province with low chlorophyll a concentrations and H. inflatus specimens that were morphologically similar to specimens from tropical and subtropical waters, but average δC in their shells was relatively high (Fig. 4b, station marked by asterisk). While the (calcium) isotopic signature of foraminiferal sub-species has been demonstrated to be the same25, small trace element compositional differences can still be caused by depth habitat preferences of different sub-species26. All pteropods analyzed here calcify in the same water depth (upper 75 m, Fig. 2a), allowing us to assume that the potentially different (sub) species have similar isotopic signatures under the same environmental conditions. Pteropod δC did not show much variation between 32° N and 26° S (Fig. 2b), where chlorophyll a concentration was much lower (Fig. 4b). Apparently, photosynthesis by these low phytoplankton concentrations did not cause a 13C enrichment of the DIC pool, explaining the relatively low δC values of the pteropods across most of the transect (average +0.70‰). However, as we observe no correlation between chlorophyll a fluorescence and δC, in the majority of the pteropod shells analyzed (Fig. 4b), other influences on δC should be explored, such as the carbonate ion effect. This effect was reported in foraminiferal13,27 and in pteropods15, and describes the inverse relationship between carbonate ion concentration and δC which is also apparent in our results (Fig. 4a, R2 = 0.94, p < 0.05). For the southernmost stations, it is impossible to disentangle the effects of 13C enrichment via photosynthesis and the carbonate ion effect on pteropod δC (Table 3). We therefore performed linear regressions on δC and water column parameters while excluding stations 60, 62, and 66 (Table 3, bottom). Excluding these three stations clearly demonstrates the carbonate ion effect on δC (Table 3, bottom, R2 = 0.84, p < 0.05) in the upper water column of an area where no 13C enrichment is occurring (no effect of chlorophyll a on δC Table 3, bottom, p > 0.05).
Our study shows that the pteropod species *H. inflatus* calcifies across a number of oceanographic provinces in the Atlantic at the same, shallow depth (upper 75 m of the water column, Fig. 2a, Table 3), making these pteropod shells good recorders of surface water masses. Correlations between stable isotopic composition of shells and parameters of the water column indicate that *H. inflatus* shells are good proxy carriers for temperature and carbonate ion reconstructions with the following regressions (for values at 50 m depth):

\[
\text{temperature} = \frac{\delta^{18}O_{\text{ptero}}}{} - 3.919 (\pm 0.404) - 0.140 (\pm 0.140)
\]

with \( p < 0.05, R^2 = 0.86 \) (Table 3, top), and

\[
\text{carbonate ion concentration} = \frac{\delta^{13}C_{\text{ptero}}}{} - 3.067 (\pm 0.386) - 0.010 (\pm 0.002)
\]

with \( p < 0.05, R^2 = 0.92 \) (Table 3, bottom), only valid for \( \delta^{13}C_{\text{ptero}} < 1\% \) (see Discussion below).

The uncertainty in the estimations for temperature (equation 1) and carbonate ion concentration (equation 2) based on an error propagation calculation sums to an error of 17% and 25%, respectively, assuming a measurement precision of 0.09% and 0.05% for the measurements of \( \delta^{18}O_{\text{ptero}} \) and \( \delta^{13}C_{\text{ptero}} \), respectively. Please note that the regressions reported above (equations 1 and 2) have been derived from different datasets. While the \( \delta^{18}O_{\text{ptero}} \)-temperature regression (equation 1) includes all stations, the \( \delta^{13}C_{\text{ptero}} \)-carbonate ion regression (equation 2) is not valid for high productivity waters (here 31° S to 38° S), as the \( \delta^{13}C_{\text{ptero}} \) in these regions may be influenced by \( ^{13}C \) enrichment (see Discussion above). Consequently, the calibration (equation 2) should only be used on \( \delta^{13}C_{\text{ptero}} \) values < 1% limiting the resolvable carbonate ion concentration to values of 200 \( \mu \)mol/kg-sw or higher.

*Heliconoides inflatus* is a pteropod species that not only occurs in the Atlantic, but has a circumglobal distribution in tropical and subtropical waters (including the Caribbean, Mediterranean and Indo-Pacific). Therefore, it is a good proxy carrier to assess surface water variations over paleo-timescales worldwide. *Heliconoides* is the oldest known pteropod genus in the fossil record (72–79 million years ago (mya)\(^28\)), and the species *H. inflatus* has been described to occur at least since the early Miocene (Aquitanian) from the Aquitaine and North Sea basins (23.03–20.44 mya\(^28\) and pers. comm. Janssen 2017). One limitation on the application of this new proxy is the occurrence of well-preserved pteropod shells in sediments, confined to waters above the lysocline of aragonite. However, there are a number of sediment cores available in which *H. inflatus* is abundant and where the calibrations reported here can be applied. The CAR-MON2 core\(^29\) would be an ideal candidate from the Caribbean Sea, as it contains *H. inflatus* in great abundance. The core spans the last 250,000 years, and the associated changes in the ocean’s temperature and carbonate ion concentration during glacial/ interglacial cycles are well resolvable by the proxy calibrations reported here. This holds true even under the restriction of the \( \delta^{13}C_{\text{ptero}} \) calibration, as surface carbonate ion concentration in the Caribbean Sea has been >250 \( \mu \)mol/kg-sw for the last 100,000 years\(^30\).

**Methods**

**Pteropod collection.** Bulk zooplankton was collected on the Atlantic Meridional Transect Cruise 22 (AMT22) between 10/19/2012 and 11/16/2012. Oblique tows were conducted with bongo nets (200 µm, 333 µm), towed between on average 361 m depth and the sea surface. Pteropods were collected from a total of 11 stations, between 31° N to 38° S latitude, in the pre-dawn hours (Table S1). After collection, pteropods were immediately fixed in pure ethanol (96–99%), which was renewed within 12–24 hours of collection. Specimens were stored at −20°C until analysis.

**Measurements of stable isotopes (\( \delta^{18}O \) and \( \delta^{13}C \)) of pteropod shells.** Pteropods (*H. inflatus*) within a narrow size range (800–1200 µm shell width) were removed from ethanol and dried at room temperature. All individual shells were weighed on a microbalance to ensure sufficient material for isotopic analysis (sample mass 120 ± 60 (1 SD) µg on average). Shells were broken to allow removal of the soft-tissue. All shell pieces were collected, triple rinsed with ultrapure water, dried at room temperature and weighed. The isotopic composition was analyzed at the Leibniz Laboratory for Radiometric Dating and Stable Isotope Research (Kiel University, Germany) using a Kiel IV carbonate preparation device connected to a ThermoScientific MAT 253 mass spectrometer. The aragonitic shells were reacted with 100% phosphoric acid (H₃PO₄) under vacuum at 75°C, and the evolved carbon dioxide gas was analyzed eight times for each individual sample. All values are reported in the Vienna Pee Dee Belemnite notation (VPDB) relative to NBS19. Precision of all different laboratory internal and international standards (NBS19) is \( \leq \pm 0.05\% \) for \( \delta^{13}C \) and \( \leq \pm 0.09\% \) for \( \delta^{18}O \) values. For notations related to shell chemistry, see Table 1.

Isotope values are reported in standard \( \delta \) notation where:

\[
\delta^{18}O = \left[ \frac{^{18}O_{\text{sample}}}{^{18}O_{\text{standard}}} - 1 \right] \times 1000
\]

(3)

\[
\delta^{13}C = \left[ \frac{^{13}C_{\text{sample}}}{^{13}C_{\text{standard}}} - 1 \right] \times 1000
\]

(4)
Seawater parameters: temperature, salinity, carbonate chemistry and chlorophyll a. Seawater temperature, salinity, and chlorophyll a concentrations in the upper 500 m of the water column were obtained by conductivity-temperature-depth (CTD) casts (Sea-Bird Electronics, models: ocean logger, SBE45, 9plus) and Chelsea MKIII Aquatracka Fluorometer, respectively. Sensors were calibrated and data archived by the British Oceanographic Data Centre (BODC). Discrete seawater samples taken from Niskin bottles were used to measure pH, TA (total alkalinity), DIC, and DIC. In order to calibrate the CTD chlorophyll fluorometer, discrete Chlorophyll a samples were analyzed fluorometrically following standard acetone extraction. Briefly, discrete chlorophyll a samples were filtered through GF/F filters (0.7 μm) and placed in acetone for 18–36 hours before fluorescence was measured on a Turner Designs AU10 fluorometer.

pH was measured (~11 samples per station) spectrophotometrically according to Clayton and Byrne. TA was measured at selected depths, including ~3 samples per station (e.g. at approximately 300, 100 and 2 m depth for station 15), and analyzed by open-cell-irritation. TA measurements were related to salinity and temperature according to the polynomial described by Lee and colleagues, and were subsequently used to estimate TA at all depths. TA and pH were used to calculate the complete C-system (DIC, bicarbonate, carbonate, Omega and Residue Factor at all depths) using the CO2SYS software. These calculations were consistent with measured DIC (at selected depths, ~3 depths per station, same depths as TA measurements) and surface pCO₂ (CO₂ partial pressure) measured continuously every 20 minutes.

Seawater composition in the sampling area. In order to characterize environmental controls on pteropod stable isotopic composition (δ18O_ptero and δ13C_ptero), seawater δ18O_sw and δ13C_in situ isoleths were calculated (surface to 300 m). The salinity (δ18O_sw, seawater: sw) calibrations from Le Grande and Schmidt for Atlantic provinces were used, with CTD-derived salinity (S) as an input parameter: North Atlantic (δ18O_sw = 0.55*S − 18.98), Tropical Atlantic (δ18O_sw = 0.15*S − 4.61) and Southern Atlantic (δ18O_sw = 0.51*S − 17.40). Thereafter, δ18O_sw was calculated from temperature and δ18O_sw according to: δ18O_sw = (T − 20)/(-4.42) + δ18O_sw, using temperature (T) from CTD measurements. There was no suitable calibration for the correlation of δ13C_in situ and DIC, therefore we used the GLODAP data set (http://cdiac.ornl.gov/oceans/GLODAPv2) to calculate linear regressions between δ13C_in situ and DIC. We defined six oceanic provinces according to latitude (45°N–30°N, 30°N–15°N, 15°N–0°N, 0°S–23°S, 23°S–30°S, 30°S–45°S) and used all available data in the upper 105 m between 10°E and 60°E, yielding six regressions for the relationship between δ13C_in situ and DIC (Table S3). The uncertainty in these calculations based on an error propagation calculation assuming a DIC concentration of 2200 ± 10 μmol/kg sums to an average error of 22% for the δ13C_in situ estimation and to an error of 5% in the δ18O_sw estimation, when assuming a salinity of 35 ± 0.1 and temperature of 24 ± 0.1 °C. These values have been calculated using the standard errors listed in Table S3 and the respective publications. All data from the World Ocean Database (WOD) and GLODAP were used to generate surface distribution maps of the Atlantic for temperature, salinity and seawater δ18O.sw and δ13C_in situ. Plots present average values from October through November in order to obtain a representation of the typical surface distribution of these parameters during the period of the cruise (10/13/2012 to 11/19/2012). The WOD data collection contained all surface data available from 1986 to 2011, and the GLODAP data collection contained all data from 1972 to 2011.

Statistical analyses. To test the effect of temperature, salinity, carbonate ion concentration, chlorophyll a concentration, δ18O in situ and δ13C_in situ on pteropod shell isotopic composition (δ18O_ptero and δ13C_ptero), linear regressions were calculated for specific depths (2, 25, 50, 75, 100, 200, 250, 300 m). Temperature, salinity and chlorophyll a were taken from the CTD casts. Carbonate ion concentration was interpolated to these depths, while δ18O_in situ and δ13C_in situ were calculated at these depths (see above).

Data availability. All data generated or analyzed during this study are included in this published article (and its Supplementary Information files).
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