Palaeogeography, Palaeoclimatology, Palaeoecology 302 (2011) 31–42

Contents lists available at ScienceDirect

Palaeogeography, Palaeoclimatology, Palaeoecology

journal homepage: www.elsevier.com/locate/palaeo

# Annually resolved $\delta^{13}C_{shell}$ chronologies of long-lived bivalve mollusks (*Arctica islandica*) reveal oceanic carbon dynamics in the temperate North Atlantic during recent centuries

Bernd R. Schöne [a,*], Alan D. Wanamaker Jr. [b], Jens Fiebig [c], Julien Thébault [d], Karl Kreutz [e]

[a] *Department of Applied and Analytical Paleontology (INCREMENTS), Earth System Science Research Center, Institute of Geosciences, University of Mainz, Johann-Joachim-Becherweg 21, 55128 Mainz, Germany*
[b] *Department of Geological and Atmospheric Sciences, Iowa State University, Science I, Ames, IA 50011-3212, USA*
[c] *Institute of Geociences, Goethe-University of Frankfurt, Altenhöferallee 1, 60438 Frankfurt a.M., Germany*
[d] *Université de Bretagne Occidentale, Institut Universitaire Européen de la Mer, Laboratoire des Sciences de l'Environnement Marin (UMR6539 UBO/IRD/CNRS), Technopôle Brest-Iroise, Place Nicolas Copernic, 29280 Plouzané, France*
[e] *Climate Change Institute, University of Maine, Orono, ME 04469-5790, USA*

ARTICLE INFO

*Article history:*
Received 4 December 2009
Received in revised form 26 January 2010
Accepted 7 February 2010
Available online 13 February 2010

*Keywords:*
Sclerochronology
Stable carbon isotope ratio
Carbon dioxide
Dissolved inorganic carbon
Oceanic Suess effect

ABSTRACT

The ability of the ocean to absorb carbon dioxide is likely to be adversely affected by recent climate change. However, relatively little is known about the spatiotemporal variability in the oceanic carbon cycle due to the lack of long-term, high-resolution dissolved inorganic carbon isotope ($\delta^{13}C_{DIC}$) data, especially for the temperate North Atlantic, which is the major oceanic sink for anthropogenic $CO_2$. Here, we report shell carbon isotope values ($\delta^{13}C_{shell}$), a potential proxy for $\delta^{13}C_{DIC}$, of old-grown specimens of the long-lived bivalve mollusk, *Arctica islandica*. This paper presents the first absolutely dated, annually resolved $\delta^{13}C_{shell}$ record from surface waters of the North Atlantic (Iceland, Gulf of Maine) covering the time interval between 1753 and 2003. According to our results, the $\delta^{13}C_{shell}$ data were unaffected by trends related to ontogenetic age. However, the shell carbonate was precipitated with a constant offset from expected equilibrium by $-1.54$ to $-2.7 \pm 0.2$‰ corresponding to a 6.2 to $10.8 \pm 0.8$% contribution of respiratory $CO_2$ ($-25$‰). The offset did not appear to vary through the lifetime of individual specimens and among specimens. Therefore, the $\delta^{13}C_{shell}$ data of this species can very likely be used as a measure of $\delta^{13}C_{DIC}$.

Furthermore, shell stable carbon isotope chronologies exhibited habitat-specific differences and a significant inter-annual and decadal variability related to the natural carbon cycle. In addition, a distinct negative $\delta^{13}C_{shell}$ shift was found reflecting the oceanic Suess effect, i.e. the admixture of anthropogenic $CO_2$. However, this shift only occurred after the early 1920s when a major climate regime shift led to a northward movement of the oceanic Polar Front in the Nordic Seas and a large-scale reorganization of atmospheric and oceanic currents in the North Atlantic. This likely resulted in a reduced admixture of cold Polar water onto the North Icelandic shelf (through the East Iceland Current) and the Gulf of Maine (through the Labrador Current) with an increased volume of warmer, isotopically well-equilibrated Atlantic waters. Our shell-based $\delta^{13}C_{DIC}$ proxy record provides the basis to quantitatively assess natural and anthropogenically induced patterns of carbon uptake in the North Atlantic.

## 1. Introduction

Approximately $118 \pm 19$ Pg C (1 Pg C $= 10^{15}$ g C $=$ 1 Gt carbon), i.e. ca. 30% of the total carbon dioxide emitted to the atmosphere by human activity between 1800 and 1994 has been absorbed by the oceans (Sabine et al., 2004) in the form of dissolved inorganic carbon (DIC). Carbon dioxide ($CO_2$) resulting from the combustion of fossil energy sources, deforestation and changes in land use is depleted in $^{13}C$ (Tans, 1981). During the last 200 years the stable carbon isotope ratio of atmospheric $CO_2$ ($\delta^{13}C_{atm}$) has thus changed from values of ca. $-6.3$ to $-8.1$‰ (Friedli et al., 1986; Francey et al., 1999; Keeling et al., 2005). Furthermore, the admixture of anthropogenic $CO_2$ has led to a significant negative shift of the stable carbon isotope ratio of the oceanic DIC ($\delta^{13}C_{DIC}$) (Nozaki et al., 1978; Druffel and Benavides, 1986; Böhm et al., 1996; Swart et al., 1998; Lazareth et al., 2000; Moore et al., 2000; Böhm et al., 2002), also known as the oceanic Suess effect (Gruber et al., 1999). The rate of change of the $\delta^{13}C_{DIC}$ signature can be used to estimate the oceanic uptake rate of atmospheric $CO_2$ (Gruber et al., 2002; Quay et al., 2003, 2007). This uptake rate, however, underlies significant spatiotemporal variations which are poorly

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[*] Corresponding author. Tel.: +49 6131 39 24757; fax: +49 6131 39 24768.
*E-mail address:* schoeneb@uni-mainz.de (B.R. Schöne).

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doi:10.1016/j.palaeo.2010.02.002

understood (Brix et al., 2004). Quantification of such fluctuations is fundamentally important for climate modeling, because changes in the strength of the oceanic carbon sink are inversely correlated to the $CO_2$ concentration in the atmosphere.

Existing studies on the inter-annual variability of the oceanic carbon inventory rely upon relatively short-term field observations of DIC and $\delta^{13}C_{DIC}$ starting in the late 1970s (Gruber et al., 2002). Such data are insufficient and far too brief to fully understand the nature of the oceanic carbon cycle either in terms of identifying the underlying processes impacting the preindustrial variability or in comprehending and predicting future changes under different climate forcings. While the stable carbon isotope data of corals and sclerosponges in particular have been employed to extend the $\delta^{13}C_{DIC}$ record in tropical settings beyond the instrumental era (Nozaki et al., 1978; Druffel and Benavides, 1986; Böhm et al., 1996; Swart et al., 1998; Lazareth et al., 2000; Moore et al., 2000; Böhm et al., 2002), very little is known from extratropical regions prior to direct measurements.

Stable carbon isotope data from shells ($\delta^{13}C_{shell}$) of the bivalve mollusk, *Arctica islandica*, can likely provide the missing information. This species is particularly suitable to reconstruct the oceanic carbon history for the following reasons.

1. The ocean quahog is the longest-lived (non-colonial) animal reaching live-spans of up to 374 or 407 years (Schöne et al., 2005a; Wanamaker et al., 2008a).
2. Annual growth interruptions result in the formation of distinct growth lines which provide an ideal means of precise calendar dating (Jones, 1980; Marchitto et al., 2000; Schöne et al., 2005b). In surface waters, growth line formation (= retardation of shell growth) in this species occurs during a time interval of about two months (September/October). It starts about a month after the seasonal temperature maximum has been attained. In deeper waters, maximum temperature only occurs in winter when the thermocline disrupts, and warm surface waters are mixed downward. Accordingly, annual growth lines of specimens living below the thermocline form between December and February (Schöne, 2008). It is currently assumed that growth line formation in *A. islandica* is triggered by spawning (Jones, 1980). Pre-mature specimens (i.e. before an age of 10 to 13; Thompson et al., 1980a; Rowell et al., 1990), however, also form annual growth lines at the exact same season of the year. According to Thompson et al. (1980b), juvenile *A. islandica* specimens mimic reproduction during pre-maturity.
3. Unlike many short-lived bivalves, *A. islandica* does not seem to exhibit age-related trends in $\delta^{13}C_{shell}$, but probably forms its shell with a constant offset from expected equilibrium with the DIC of the ambient water (Erlenkeuser, 1976; Schöne et al., 2005a).
4. Furthermore, *A. islandica* inhabits the entire northern North Atlantic (Nicol, 1951), i.e. the largest oceanic sink — relative to the size of this ocean — for atmospheric $CO_2$ (Gruber et al., 2002).

In the present study, we investigated the potential of $\delta^{13}C_{shell}$ chronologies of *A. islandica* as recorders of the $\delta^{13}C_{DIC}$ history of the temperate North Atlantic. Particular questions included: Are the $\delta^{13}C_{shell}$ time-series affected by vital effects? Are variations of the natural carbon cycle recorded in the shells? Is the long-term oceanic Suess effect preserved in the shell records of *A. islandica* as recently suggested by Butler et al. (2009), and does this trend differ from the $\delta^{13}C$ trend in tropical surface waters and the atmosphere in regard to timing and strength? Our results were contrasted to sclerosponge records (Böhm et al., 1996, 2002) from tropical settings, to atmospheric data and to sparsely available direct $\delta^{13}C_{DIC}$ measurements. Results of this study can provide an important building block toward a better understanding of spatiotemporal differences of the ocean's capacity for absorbing anthropogenic $CO_2$ and to evaluating the potential consequences for future climate change and ecosystems.

## 2. Material and methods

Four shells of the ocean quahog, *A. islandica*, were collected alive by dredging from surface waters (ca. 30 m water depth) in the North Atlantic Ocean. Three specimens came from Icelandic waters, while one came from the Gulf of Maine (see Wanamaker et al., 2008b for complete details) (Fig. 1). Two of the Iceland shells ('Langanes 5' and 'Langanes 9'; live-collected in November 2003) lived offshore of Langanes (N66°16′, W14°55.20′), northeastern Iceland, and were largely influenced by the cold East Iceland Current (EIC; Fig. 1). The third Iceland shell, however, grew in a biologically more productive nearshore environment near Flatey Island (ca. N66°11′, W77°51′; live-collected in 1986), north Iceland, which is primarily controlled by the Irminger Current (IC), a warmer branch of the North Atlantic Current (NAC; Fig. 1). The Western Gulf of Maine shell (N43°39′ 22.14″, W69°48′6.01″) was exposed to a mixture of the cold Labrador Current (LC) and relatively warm water masses derived from the Gulf Stream (Gatien, 1976; Wanamaker et al., 2008b). Selection of different localities and environmental settings enabled the identification of potential spatiotemporal incoherencies of the oceanic carbon sink and/or differences in the natural carbon cycle.

### 2.1. Shell preparation

In preparation for annual growth pattern and isotope analyses, one valve of each specimen was mounted on a plexiglass block and a quick-drying epoxy resin (JB KWIK-Weld) applied to the valve surface. Two immediately adjacent, three-millimeter-thick sections were cut from the valves along the height axis and perpendicular to the annual growth lines, with a Buehler Isomet 1000 low-speed precision saw using a 0.4 mm thick low concentration-diamond wafering blade (LC15). The shell slabs were subsequently mounted on

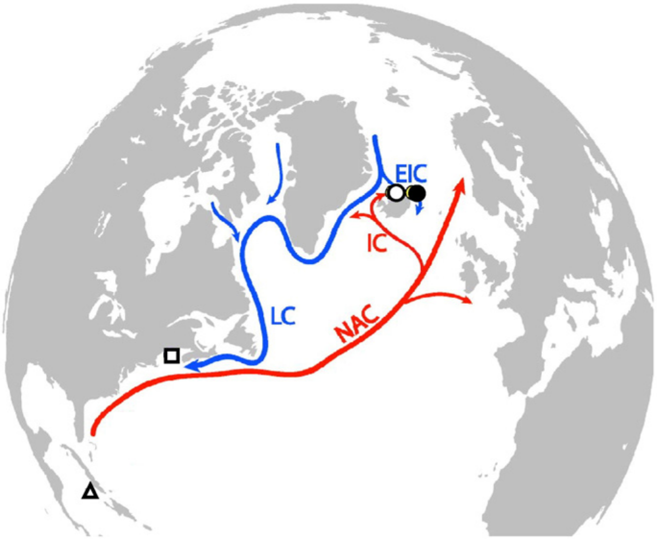

**Fig. 1.** Map showing major warm (NAC = North Atlantic Current; IC = Irminger Current) and cold (LC = Labrador Current; EIC = East Iceland Current) currents in the North Atlantic and sampling localities of the biogenic hard parts (bivalve shells and sclerosponges) that were used for analyses of stable carbon isotopes. The shell from Flatey (open circle) was exposed to stronger influence of the warm IC, a branch of the NAC, whereas the two specimens from Langanes (filled circle) and the shell from the Gulf of Maine (open square) received relatively larger amounts of cold water from the Polar region (LC, EIC). Tropical sclerosponge (Jamaica; open triangle) carbon isotope data (Böhm et al., 1996, 2002) were taken from the literature for comparison with the new bivalve record.