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The imprint of anthropogenic CO_2 in the Arctic Ocean: evidence from planktic $\delta^{13}C$ data from watercolumn and sediment surfaces

D. Bauch^{a,*}, J. Carstens^b, G. Wefer^b, J. Thiede^c

^aGEOMAR, Forschungszentrum für Marine Geowissenschaften der Christian-Albrechts-Universität, D-24148 Kiel, Germany

^bFachbereich Geowissenschaften, Universität Bremen, D-28334 Bremen, Germany ^cGEOMAR and Alfred-Wegener-Institut für Polar-und Meeresforschung, D-27568 Bremerhaven, Germany

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Abstract

 δ^{13} C values of *N. pachyderma* (sin.) from the water column and from core top sediments are compared in order to determine the ¹³C decrease caused by the addition of anthropogenic CO₂ to the atmosphere. This effect, which is referred to as the surface ocean Suess effect, is estimated to be about $-0.9\%_{00}$ ($\pm 0.2\%_{00}$) within the Arctic Ocean halocline waters and to about $-0.6\%_{00}$ ($\pm 0.1\%_{00}$) in the Atlantic-derived waters of the southern Nansen Basin. This means that the area where the Arctic Ocean halocline waters are formed, the Arctic shelf regions, are relatively well ventilated with respect to CO₂. Nevertheless, δ^{13} C of dissolved inorganic carbon ($\delta^{13}C_{DIC}$) in the Arctic Ocean halocline waters is far from isotopic equilibrium. Absolute values of δ^{13} C of *N. pachyderma* (sin.) covary with the surface ocean Suess effect, and we interprete changes in both parameters as a reflection of the degree of ventilation of the waters on the shelf sea. Measurements of δ^{13} C of *N. pachyderma* (sin.) in the Arctic Ocean from plankton tows reveal a "vital effect" of about $-2\%_{00}$, significantly different from other published values. A first-order estimate of the total anthropogenic carbon inventory shows, that despite of its permanent sea-ice cover, the Arctic Ocean, with 2% of the global ocean area, is responsible for about 4–6% of the global ocean's CO₂ uptake. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

In this study we present δ^{13} C data of the polar planktic foraminifer *Neogloboquadrina pachyderma* (sinistral) from the water column and from core top sediments taken

^{*} Corresponding author. Tel.: + 49-431-600-2252; fax: + 49-431-600-2941.

E-mail address: dbauch@geomar.de (D. Bauch).

in the Nansen Basin of the Arctic Ocean. From this, interpretation of δ^{13} C data in sediment records is possible. Additionally, the comparison of δ^{13} C data of *N. pachy-derma* (sin.) from the water column and from core top sediments gives information about the invasion of anthropogenic CO₂ into the surface ocean water.

Planktic carbon isotope records and their interpretations are much debated, because several factors can complicate the interpretation. These may be species-dependent 'vital effects', depth-habitat, or global sea water δ^{13} C variations of total CO₂ due to gas exchange with the atmosphere in combination with local productivity and community respiration (e.g. Berger et al., 1981; Labeyrie and Duplessy, 1985). Nevertheless, the understanding of δ^{13} C values of planktic foraminifers from marine sediments is an important issue because these records can be linked to past changes in atmospheric CO₂ concentrations (e.g. Keir, 1995).

At high latitudes N. pachyderma (sin.) is the dominant species and the only planktic foraminifer providing long and continuous downcore isotope records. During the recent past, the δ^{13} C composition of the atmosphere has changed by about 1.4%, due to the anthropogenic CO_2 increase, from a pre-industrial value of about -6.4%(Friedli et al., 1986, Francey et al., 1999) in 1850 to about -7.8% (Keeling et al., 1989) in 1987. This decrease is imprinted on the δ^{13} C of the DIC (δ^{13} C_{DIC}) in the ocean, but due to the slow rate of isotopic equilibration the ocean surface water is generally not in isotopic equilibrium with the atmosphere (e.g. Lynch-Stieglitz et al., 1995), and therefore the amount of decrease in $\delta^{13}C_{DIC}$ of ocean surface water will depend on the degree of isotopic equilibration with the atmosphere at that specific location and the extent of dilution with deeper waters, which do not show any or a small anthropogenic imprint due to their age. The interior of the Arctic Ocean is covered by sea-ice throughout the year, therefore air-sea exchange in this area is restricted. But an intense flux of carbon from the atmosphere to the ocean occurs over the broad and shallow shelf areas due to extensive biological production during the summer (Anderson et al., 1990; Walsh et al., 1989; Sambrotto et al., 1984). Additionally, the uptake of atmospheric CO₂ can be caused by cooling of surface waters and a concomitant increase in solubility (Simonsen and Haugan, 1996). Carbon budgets for the Arctic Ocean have been calculated based on volume flows, and the perturbation due to the addition of anthropogenic CO₂ has been estimated (Anderson et al., 1998; Lundberg and Haugan, 1996). Because the contribution of anthropogenic CO_2 is small compared to the overall fluxes, the uncertainty of this approaches is high with respect to the uptake of anthropogenic CO_2 . In this study we present independent evidence of the amount of anthropogenic CO_2 increase in the Arctic Ocean.

2. Material and methods

During the ARK IV/3 expedition in 1987 (Polarstern Scientific Party, PSSP 1988) plankton tows and sediment samples were collected (for station locations see Fig. 1). Plankton-tow samples were taken using vertical tows in the upper 500 m of the water column with a mesh size of 63 μ m using a multinet device (Fa. Hydrobios) and stored in a buffered 2% formalin–seawater solution. Plankton-tow sample collection and

handling procedure are described in detail by Carstens and Wefer (1992), and results with respect to habitat, calcification depth and oxygen isotopes have been published elsewhere (Carstens and Wefer, 1992; Bauch et al., 1997). For each isotope measurement of *N. pachyderma* (sin.), conducted at the laboratory of Bremen University, about 10–20 and 20–30 individuals showing a secondary calcification were picked from the size fractions > 250 and 160–250 µm, respectively. Measurements of δ^{13} C of sediment surface samples were conducted on 20–30 calcified individuals of *N. pachyderma* (sin.) taken from the size fraction 125–250 µm (Köhler, 1992).

Radiocarbon AMS dating for some of the core tops reveal ages of about 3–3.5 kyr. This relatively old ages are due to low sedimentation rates and bioturbation (Köhler, 1992; Spielhagen and Erlenkeuser, 1994). The δ^{13} C data precisions is 0.07‰ based on replicate analyses of the working standard. Isotope measurements are presented using the conventional δ^{13} C notation relative to PDB.

In this study we compare core top with water-column data of *N. pachyderma* (sin.). For isotope measurements size fractions of 125–250 and 160–250 μ m were taken for the sediment and water-column samples, respectively. We consider the size-effect due to different size fractions to be insignificant, because the average size of individuals picked from the 125–250 μ m fraction is biased towards the upper end of the size spectrum due to the great number of large individuals in each sample, while for the size fraction 160–250 μ m in most cases all individuals were taken for isotopic analysis and the average size is centered around the mean of the interval.

3. Hydrographic setting and N. pachyderma (sin.) results from previous studies within the Arctic Ocean

At present, the central Arctic Ocean is covered by sea-ice throughout the year. The halocline is about 200 m thick, with temperatures close to the freezing point of sea water (ca. -1.8° C). It is maintained by waters from the shelves, e.g. rivers, meltwater from sea-ice, and brines released during sea-ice formation, all which have a wide range of salinity (Aagaard and Carmack, 1989). Atlantic waters flow into the Arctic Ocean via the West Spitsbergen Current through Fram Strait and cause an Atlantic-derived temperature and salinity maximum at 200-500 m water depth. These waters describe a cyclonic circulation within the Arctic Ocean with several branches turning back towards Fram Strait (Coachman and Barnes, 1963; Rudels, 1995). There is a north/south hydrographic boundary within the Nansen Basin, which is well characterized by the occurrence of river runoff to the north and the cross-over between net sea-ice melting towards the south and net sea-ice formation towards the north (Bauch et al., 1995). In the southern part of the Nansen Basin (station 276 to 340) N. pachyderma (sin.) prefers a habitat in the advected Atlantic water between 100 and 200 m depth. In the northern Nansen Basin (station 358-371) the habitat of N. pachyderma (sin.) lies on average at about 75 m water depth within the Arctic Ocean halocline waters, which overlies the Atlantic layer there. An average depth of calcification can be derived from the standing stock data, in conjunction with the proportion of living specimens and individuals showing a secondary calcite crust in each depth interval (see Table 1). The depth of calcification varies in both regimes

Plankton tov	w averages			Core top data		
Habitat depth (m)	Calcific. depth (m)	δ ¹⁸ O (160-250 μm) (%)	δ ¹³ C (160-250 μm) (%)	δ ^{1 8} O (125–250 μm) (%o)	δ ¹³ C (125-250 μm) (%)	Name of core(s)
200	211	3.42	- 0.01 (0.16)	3.33	0.55	1513-9
116	130	3.52	-0.16(0.15)	3.50	0.50	1515-10, 1519-11
128	182	3.42	-0.07(0.03)	3.50	0.50	1515-10, 1519-11
145	157	3.54	0.14(0.09)	3.71	0.53	1519-11
75	141	3.44	-0.11(0.07)	3.50	0.00	1519-20, 1522-19
84	163	2.76	-0.12	3.12	1.01	1522-19
99	205	2.77	-0.30(0.12)	2.64	0.87	1523-15
64	92	2.27	0.10(0.11)	2.72	0.80	1529-7
65	111	2.81	0.10(0.06)	2.60	0.80	1525-2, 1527-10
73	108	2.41	0.24 (0.27)	2.63	0.90	1527-10
g for the data esents the frac standing stocl	of <i>N. pachyderma</i> ction of calcite fo k (concentration of	<i>i</i> (sin.) from the water c prmed at each depth in of foraminifera in interv	olumn was determined iterval. $f_{cw}(%) = [SSL]$ all sum of concentration	according to the "calc CI(1 – D1)] Σ [SSLCI(ns in all intervals); CI tl	ification weighing facto $1 - DI$], where L is the proportion of individ	rs", <i>f</i> _{CW} (Bauch et al., e length of sampling uals with a secondary
	Habitat depth (m) (m) 200 200 116 128 145 75 84 66 64 65 63 73 73 73 73 73 73 73 73 73 73 73 73 73	HabitatCalcific.depthdepthdepth(m)(m)(m)(m)(m) (m) (m) (m) (m) (m) (m) (m)	Habitat Calcific. δ^{18} O depth depth (160–250 µm) (m) (m) (m) (m) (m) (%) (m) (m) (m) (m) (m)	Habitat Calcific. δ^{18} O δ^{13} C depth depth (160–250 µm) (160–250 µm) (m) (m) (m) (m) (m) (m) (m) (m) (m) (m) (m) (m) (m) (m) (m) 200 211 3.42 -0.01 (0.16) 116 130 3.52 -0.01 (0.03) 128 182 3.42 -0.01 (0.03) 141 3.42 -0.01 (0.03) 157 3.44 -0.11 (0.07) 66 205 2.77 -0.12 (0.01) 67 92 2.76 -0.12 (0.01) 67 111 2.81 0.10 (0.01) 65 111 2.81 0.10 (0.06) 73 108 2.41 $0.10 (0.06)$ 73 108 2.41 $0.24 (0.27)$ 73 108 0.24 (0.27) $0.24 (0.27)$ 73 108 2.41 $0.24 (0.$	HabitatCalcific. δ^{18} O δ^{13} C δ^{13} C δ^{18} Odepthdepth(160-250 \mum)(160-250 \mum)(125-250 \mum)(m)(m)(m)(m)(m)(m)(m)(m)(m)(m)(m)(m)(m)(m)(m)(m)(m)(m)200211 3.42 -0.01 (0.16) 3.33 200211 3.42 -0.01 (0.16) 3.33 116130 3.52 -0.01 (0.15) 3.50 128182 3.42 -0.01 (0.03) 3.71 75141 3.44 -0.11 (0.07) 3.70 75141 3.44 -0.11 (0.07) 3.70 75141 3.44 -0.11 (0.07) 3.70 75141 3.44 -0.11 (0.07) 3.70 75111 2.77 -0.30 0.12 2.64 66205 2.77 -0.12 0.10 0.01 73108 2.41 0.10 0.06 2.63 73108 2.41 0.24 0.27 2.63 73108 2.41 0.10 0.06 2.63 73108 0.24 0.27 2.63 732.41 0.10 0.06 2.63 74 2.77 0.10 0.06 2.63 75 0.10 0.06 2.64 75 0.10 0.06 2.63 <	HabitatCalcific. δ^{18} O δ^{13} C δ^{18} O δ^{13} C δ^{13} O δ^{13} Cdepthdepth $(160-250 \ \mum)$ $(160-250 \ \mum)$ $(125-250 \ \mum)$ $(125-250 \ \mum)$ (m)(m)(m) $(\%_{oo})$ $(\%_{oo})$ $(\%_{oo})$ $(\%_{oo})$ (m) (m) $(\%_{oo})$ $(\%_{oo})$ $(\%_{oo})$ $(\%_{oo})$ 200 211 3.42 $-0.01 (0.16)$ 3.33 0.55 116 130 3.52 $-0.01 (0.16)$ 3.33 0.55 128 182 3.42 $-0.01 (0.03)$ 3.71 0.53 145 157 3.54 $0.01 (0.03)$ 3.71 0.53 145 157 3.54 $-0.01 (0.07)$ 3.50 0.50 145 157 3.54 $-0.01 (0.07)$ 3.50 0.50 145 157 3.54 $0.01 (0.07)$ 3.50 0.90 84 163 2.76 $0.01 (0.01)$ 3.71 0.53 66 205 2.77 $-0.01 (0.01)$ 3.72 0.90 65 111 2.72 0.010 0.26 0.80 65 111 2.81 $0.24 (0.27)$ 2.64 0.80 73 108 2.41 $0.24 (0.27)$ 2.63 0.90 73 108 2.41 $0.24 (0.27)$ 2.63 0.90 75 100 0.060 2.60 0.80 75 101 0.060 2.64 0.90 <td< td=""></td<>

average calcification depth was calculatd from the depth distribution of the calcification weighing factors. The values in parenthesis are the standard deviation of

the weighted mean δ^{13} C values of N. pachyderma (sin.) from plankton tows.

Table 1



Fig. 1. Location map showing the geographical position of stations taken during Ark IV/3 in 1987, where shells of *N. pachyderma* (sin.) from plankton tows were analysed for their δ^{13} C composition. The arrows indicate the surface currents of Atlantic waters entering through Fram Strait and the Arctic halocline waters moving southwards with the Transpolar Drift.

between 100 and 200 m water depth (Bauch et al., 1997) and represents the mean depth at which a foraminifer forms its calcite shell. The absolute concentration of planktic foraminifera in the upper 500 m of the water column shows a maximum of 60 ind m^{-3} (for tests 160–250 µm) at station 296, which was located near an area with only partial sea-ice cover. Farther north, concentrations decrease rapidly (Carstens and Wefer, 1992) (Fig. 1).

Oxygen isotope data of *N. pachyderma* (sin.) from plankton tows show a consistent offset with depth of about 1% relative to δ^{18} O values calculated for inorganic calcite precipitated under equilibrium conditions from in situ δ^{18} O and temperature measurements of the water column (Shackleton, 1974; O'Neil et al., 1969). A comparison of δ^{18} O values of *N. pachyderma* (sin.) from plankton tows and surface sediments shows good agreement (Figs. 4a and b) and shows that significant calcification of *N. pachyderma* (sin.) does not occur below the towing depth of 500 m (Bauch et al., 1997).

4. Results

4.1. Carbon isotope data of N. pachyderma (sin.)

Carbon isotopes of *N. pachyderma* (sin.) in plankton tows show an average offset of 0.25% ($\pm 0.04\%$ standard error) between the size fractions 160–250 and $> 250 \mu m$ (Fig. 2). This phenomenon agrees with observations made for various species as well

as for *N. pachyderma* (sin.) (Aksu and Vilks, 1988; Donner and Wefer, 1994; Kohfeld et al., 1996). This consistent offset was used to extend the data base of the size fraction 160–250 µm by using the data of the coarser size fraction and the observed offset whenever data from the finer fraction are missing (6 cases; see Fig. 2). There is no systematic change of δ^{13} C values of *N. pachyderma* (sin.) with depth. While phosphate concentrations in the water column reflect the distribution of river water within the northern Nansen Basin (Fig. 2; Stations 340–371), this phenomenon is not reflected in δ^{13} C values of *N. pachyderma* (sin.) (Figs. 2 and 5). Average carbon isotopes of *N. pachyderma* (sin.) from the water column for the size fraction 160–250 µm show δ^{13} C values between -0.3 and 0.2% in the Nansen Basin.

In order to compare plankton with sediment data, a weighting of the plankton samples was conducted using the "calcification weighting factors" f_{CW} , which represent the fraction of calcite formed at each depth interval (Bauch et al., 1997)

$$f_{\rm CW}(\%) = [SSLCI(1 - DI)]/\Sigma[SSLCI(1 - DI)],$$

where L is the length of sampling interval, SS the standing stock (concentration of foraminifera in interval/sum of concentrations in all intervals), CI the proportion of individuals with a secondary calcite crust, and DI the proportion of individuals sampled dead. The weighed average δ^{13} C from the plankton data therefore represents δ^{13} C of *N. pachyderma* (sin.) as it is imprinted in the sediments.

In the Nansen Basin δ^{13} C values of *N. pachyderma* (sin.) from core top sediments increase from south to north from about 0.4‰ at the Barents Sea slope to about 0.9‰ at the Gackel Ridge (Fig. 3). In the Amerasian part of the Arctic Ocean δ^{13} C values further increase to about 1.1‰ just west of the Lomonosov Ridge (Spielhagen and Erlenkeuser, 1994). Relative to core top data, the weighted values of *N. pachyderma* (sin.) from the water column are depleted in δ^{13} C by 0.4–1.2‰ (Fig. 3).

4.2. Estimates of carbon isotopes of total dissolved carbonate (DIC)

Direct measurements of δ^{13} C of the DIC within the Arctic Ocean are not published. Measurements were conducted for samples taken during ARK IV/3 in 1987 (Kromer, 1998, pers. comm.), which were processed for ¹⁴C-AMS analysis; therefore no quantitative extraction of the DIC was intended, and the data are of insufficiant quality with respect to $\delta^{13}C_{DIC}$. The $\delta^{13}C$ values range between 0.3 and 1.2‰ (omitting 1 data point) within the upper 600 m for stations in the Nansen Basin, and no consistent trend with depth was detected. Considering that incomplete extraction of the samples will lower the measured $\delta^{13}C$ value, due to fractionation processes during the extraction, a rough estimate for the $\delta^{13}C_{DIC}$ in the Atlantic layer of about 1‰ is made. This estimate is close to values of $\delta^{13}C_{DIC}$ reported for samples taken within the upper 600 m and below the euphotic zone at locations "upstream" of our study area in the Norwegian and Greenland seas of 1 to 1.3‰ observed in 1972 (Kroopnick, 1985; Nydal et al., 1992) and "downstream" values made in the Atlantic layer near the shelf break of the Laptev Sea of 0.8 to 1‰ observed in 1997 (Erlenkeuser, 1999, pers. comm.).



Fig. 2. δ^{13} C values of *N. pachyderma* (sin.) from plankton tows ((\bigcirc) = tests 160–250 µm; (\blacktriangle) = tests > 250 µm); the values are indicated in the middle of each sampling interval (0–50, 50–100, 100–200, 200–300 and 300–500 m). The vertical lines represent δ^{13} C values of *N. pachyderma* (sin.) from core top sediments. Phosphate measurements in the water column are indicated by grey diamonds.



Fig. 3. δ^{13} C values of *N. pachyderma* (sin.) versus latitude of the station location. (\bigcirc) Weighted average δ^{13} C values of *N. pachyderma* (sin.) from plankton tows of the size fraction 160–250 µm. ($\textcircled{\bullet}$) δ^{13} C values of *N. pachyderma* (sin.) from core top sediments of the size fraction 125–250 µm. Solid black line: Difference of δ^{13} C values of *N. pachyderma* (sin.) from plankton tows and core top sediments (surface ocean Suess effect). (\bigtriangledown) Theoretical δ^{13} C values in equilibrium with the modern atmospheric CO₂ (δ^{13} C = $-7.8_{\infty 0}^{*}$) calculated at the temperature measured at the average depth of calcification of *N. pachyderma* (sin.) (Zhang et al., 1995). The grey-shaded area marks our estimate of δ^{13} C_{DIC} in the Nansen Basin.

This estimate of $\delta^{13}C_{DIC}$ in the Nansen Basin of 1% is far off the isotopic the atmosphere. equilibrium with Assuming equilibrium fractionation $(\varepsilon(DIC - g) = -0.105T + 10.51;$ Zhang et al., 1995) at temperatures measured at the depth of calcification for N. pachyderma (sin.) in the Arctic Ocean (0.8 to -1.4° C; Bauch et al., 1997) values of $\delta^{13}C_{DIC}$ in equilibrium with the atmosphere of about 2.5_{∞}° for modern conditions and about 4_{∞}° for preindustrial conditions are derived (taken δ^{13} C of atmospheric CO₂ as -7.8 and -6.4%, respectively). Because equilibration with the atmosphere occurs at the sea surface, this estimate implies that the water parcel at the depth of calcification had the same temperature when it was formed at the surface of the Arctic shelf areas. Since the calculations are not very sensitive within the applicable temperature range, this estimate seems reasonable.

5. Discussion

5.1. Reflection of $\delta^{13}C_{DIC}$ values in $\delta^{13}C$ data of N. pachyderma (sin.)

Inorganic calcite precipitated in equilibrium with seawater was found to be 1_{00}^{\prime} heavier in δ^{13} C than the δ^{13} C of the DIC (Romanek et al., 1992). Our estimate of δ^{13} C_{DIC} suggests that *N. pachyderma* (sin.) precipitates its calcite out of isotopic equilibrium and is by roughly 1_{00}^{\prime} lighter than δ^{13} C_{DIC} (Fig. 3). A study in the new polynia on the Greenland shelf (Kohfeld et al., 1996) reveals no offset between δ^{13} C_{DIC} and δ^{13} C of *N. pachyderma* (sin.) This study as well as that by Kohfeld et al. (1996) indicate that *N. pachyderma* (sin.) does not form its calcite crust according to equilibrium fractionation of inorganic calcite, thus revealing a "vital effect" of -2 and -1_{00}^{\prime} , respectively. The inconsistency between the observed vital effects might only partially be explained by the roughness of the estimate of our study, but has to be caused by other reasons as well. Further studies are needed to investigate influences on vital effects of *N. pachyderma* (sin.); so far results indicate regional differences in controlling factors (Kohfeld et al., 1996; Kohfeld, 1998).

5.2. Comparison of water column and sediment data of N. pachyderma (sin.)

For a comparison of the core top and the water-column data it is important to keep in mind that the core top values of *N. pachyderma* (sin.) represent mean ocean conditions, averaging about 3 kyrs within the Arctic Ocean (Köhler, 1992), and reflect pre-industrial conditions with respect to δ^{13} C values. Data from living *N. pachyderma* (sin.) from the water column, on the other hand, reflect modern, anthropogenically, influenced values. Additionally, a shift in surface water properties has been detected during the last decade (Macdonald, 1996; Steele and Boyd, 1998). However, oxygen isotopes are not influenced by the anthropogenic CO₂ increase and δ^{18} O is a non transient tracer. The plankton and sediment δ^{18} O values of *N. pachyderma* (sin.) in the Arctic Ocean (Figs. 4a and b) corespond within $\pm 0.09\%$, constant within the statistical variability. Therefore, we conclude that our data set, taken in 1987, is representative for conditions in that area in terms of annual variability and with respect to climate changes over the last 3 kyrs. But it also can be seen that the northern most stations (362–370) show higher variations ($\pm 0.20\%$), probably an artifact due to the low sampling density at this stations.

5.3. Surface ocean ¹³C Suess effect

Since the comparison of δ^{18} O values of *N. pachyderma* (sin.) from plankton tows and surface sediments shows good agreement (Fig. 4), we assume that the changes between plankton tows and surface sediments δ^{13} C data of *N. pachyderma* (sin.) are solely caused by the transient characteristic of this parameter caused by the penetration of anthropogenic CO₂ into the ocean during the last 150 yrs. Thus, the decrease of δ^{13} C values of *N. pachyderma* (sin.) from the water column relative to core top



Fig. 4. (a) δ^{18} O values of *N. pachyderma* (sin.) from plankton tows (\blacksquare) and core top sediments (\square). (b) Difference of isotope values of *N. pachyderma* (sin.) from plankton tows and core top sediments. (\blacktriangledown) δ^{13} C values (plankton-core top) (\bigcirc) δ^{18} O values (plankton-core top). The horizontal lines represent calculated average values.

sediments values reflects the surface ocean ${}^{13}C$ Suess effect at the depth range of calcification of *N. pachyderma* (sin.).

The surface ocean ¹³C Suess effect changes along the section over the Nansen Basin (Fig. 3). In the southern regime (stations 276 to 296), reflecting the waters of the Atlantic layer, the decrease amounts to about -0.6% ($\pm 0.1\%$). In the northern regime (stations 340 and 358–371), reflecting the waters of the Arctic Halocline, the decrease is variable and shows values between -1.0 and -1.2% for stations 340, 358 and 362, and farther north -0.7% for station 376, 370 and 371. The average surface ocean ¹³C Suess effect for the entire northern regime is -0.9% ($\pm 0.2\%$).

An anthropogenic decrease of -0.9% in $\delta^{13}C_{DIC}$ for the Arctic Ocean halocline waters is relatively high compared to values measured in other areas of the world oceans. For the upwelling regime off West Africa about -0.6% was determined using also planktic foraminifera from core tops and water column (Beveridge and

Shackleton, 1994). From demosponges from the Caribbean and Coral seas a decrease of $\delta^{13}C_{DIC}$ in surface water of -0.9 and -0.7% was derived, respectively (Böhm et al., 1996). Data of sinking organic matter in comparison to data from surface sediments suggest relatively high storage rates of anthropogenic CO₂ in Southern Ocean surface waters, exceeding the value derived by Beveridge and Shackleton (Fischer et al., 1997).

The amount of the surface ocean ¹³C Suess effect represents the degree of equilibration with the atmosphere and is thus a measure of ventilation of the surface water in respect to CO_2 . Therefore, it can be concluded that Arctic Ocean halocline waters are better equilibrated with the atmosphere than the Atlantic-derived waters in the southern Nansen Basin. Because the Arctic Ocean is permanently covered with sea-ice and the shelf regions are only free of sea-ice during the summer season, this relatively good degree of equilibration with the atmosphere seems surprising. On the other hand, the Arctic Ocean halocline waters are formed on the broad and shallow Siberian shelf regions (Aagaard and Carmack, 1989) and river water is added, which serves for lowering preformed $\delta^{13}C$ (Erlenkeuser, 1995). Apparently the spread of the halocline waters in a relatively thin layer over the shelf areas with an approximate three-month sea-ice-free summer season and the input of river water, allows a better isotopic exchange with the atmosphere than the conditions experienced by the Atlantic-derived waters, which flow with the West Spitzbergen Current and over the Barents Sea into the Arctic Ocean (Rudels, 1995).

5.4. Interpretation of absolute $\delta^{13}C$ values of N. pachyderma (sin.)

Parallel to the increase in offsets between sediment and water $\delta^{13}C$ data between the southern and the northern regime, there is also an increase in absolute value of sediment $\delta^{13}C$ of *N. pachyderma* (sin.) from about 0.5% at 81.5°N to about 0.9% at 84°N (Fig. 3). This could mean that the absolute $\delta^{13}C$ values of *N. pachyderma* (sin.) of core top sediments within the Nansen Basin are correlated with the degree of equilibration with the atmosphere.

The estimated value of $\delta^{13}C_{DIC}$ in the Arctic Ocean (estimated to roughly 1‰ at 100–500 m waterdepth) is about 1.5‰ lower than expected for theoretically calculated $\delta^{13}C_{DIC}$ in isotopic equilibrium with the atmosphere (Fig. 3). Therefore, enhanced isotopic equilibration will tend to raise absolute values of $\delta^{13}C_{DIC}$ and thereby $\delta^{13}C$ of foraminiferal calcite. The trend in absolute values of sediment $\delta^{13}C$ data of *N. pachyderma* (sin.) is in line with the degree of ventilation within the waters of the Arctic Halocline. Due to the permanent sea-ice cover of the Arctic Ocean interior the change in isotopic equilibration or residence times on the Siberian shelf areas. The fact that such changes show up at the "downstream" location in the Nansen basin also demonstrates how well the Arctic Halocline is stratified in a lateral direction within the Transpolar Drift (e.g. Aagaard and Carmack, 1989).

When discussing absolute δ^{13} C values, possible alteration of the signal due to biological processes has to be taken into account. These processes are recorded in



Fig. 5. δ^{13} C values of *N. pachyderma* (sin.) from the water column versus phosphate concentration for stations in the Nansen basin, both taken during ArkIV/3. The phosphate concentrations are averaged over the depth intervals of the plankton samples. The line on the upper-right side reflect the expected functional relation between δ^{13} C_{DIC} and PO₄ caused by biological processes (δ^{13} C_{DIC} = 2.8 – (1.1 × PO₄); Lynch-Stieglitz et al., 1995), subsequent lines represent the same relationship with a constant offset (as indicated), and the little box represents the values found for North Atlantic Deep Water, NADW (see Lynch-Stieglitz et al., 1995). Note that δ^{13} C values of *N. pachyderma* (sin.) from the water column reflect modern δ^{13} C_{DIC} values with the addition of a vital effect, and are according to our data about 1‰ lighter than δ^{13} C_{DIC}.

 δ^{13} C changes as well as in nutrient concentrations. The δ^{13} C values of *N. pachyderma* (sin.) and in situ measured PO₄ values reveal some scatter, and no simple biologically controlled process can be detected (Fig. 5). We therefore conclude that δ^{13} C values within the Arctic Ocean halocline represent primarily a water mass signature and are dominated by the effect of the air-sea exchange.

5.5. Inventory of anthropogenic CO_2 in the Arctic Ocean

In the following paragraph a rough inventory for anthropogenic CO₂ uptake into the Arctic Ocean will be made. This serves as a first-order estimate and shows the consistency of our data. We apply an average δ^{13} C decrease for the DIC of surface water of about -0.9% ($\pm 0.2\%$); this is the average value found for our data set within the Arctic Ocean halocline waters at the average calcification depth of *N. pachyderma* (sin.) in that region (about 110 m water depth). Because the δ^{13} C signature of planktic foraminifera is strongly dependent on the carbonate chemistry of the ocean (Spero et al., 1997), a second scenario is calculated by applying a "carbonate ion effect" according to the estimated decrease in carbonate ion concentration $[CO_3^2^-]$ and the concomitant increase in δ^{13} C of *N. pachyderma* (sin.). Modelling results of Heimann and Maier-Reimer (1996) using the Hamburg model of the oceanic carbon cycle show that the ratio of isotopic change at the ocean surface and the change in its total carbonate content, $\Delta \delta_s / \Delta C_t$ is nearly constant, and a global range of -0.016 to -0.019% kg/µmol was derived. Keir et al. (1998) determine from their observations in the North Atlantic a ratio of $\Delta \delta_s / \Delta C_t$ of -0.016% kg/µmol. Adopting a mean value of -0.0175% kg/µmol for $\Delta \delta_s / \Delta C_t$ and -0.9% for $\Delta \delta_s$, a value of 51 µmol/kg for ΔC_t can be derived for the Arctic Ocean halocline waters. This estimate is a factor of two higher than the values estimated for that area from model calculations up to the year 1982 (Gruber, 1998). Total carbonate (C_t) and total alkalinity (A_t) measurements from the ARCTIC'91 expedition reveal a modern Revelle factor of about 14 within the Nansen Basin, and a maximal capacity for uptake of anthropogenic CO₂ of about 45 µmol/kg can be estimated (Körtzinger, 1998, pers. comm.).

We will use tritium measurements to calculate the mean penetration depth for this transient tracer within the Arctic Ocean and estimate a mean penetration depth for anthropogenic CO_2 by accounting for the different penetration times, according to the rough estimate that the exchange volume increases as the square root of time (Broecker and Peng, 1993). The mean tritium penetration depth within the Nansen Basin (11 stations; Ark IV/3, station 310-376, taken in 1987, Schlosser et al., 1995) amounts to about 920 m (\pm 70 m). Using the mean age of a fossil fuel CO₂ molecule of about 30 years in 1987 and that bomb tritium was released in 1962, the mean penetration depth of anthropogenic CO₂ is estimated to 920 m × $(30/25)^{1/2} = 1008$ m in 1987. From the mean penetration depth and area of the Arctic Ocean interior and addition of the volume of the Arctic shelf regions a total volume of $(5.1 \times 10^6 \text{ km}^2 \times 1008 \text{ m}) + 1.3 \times 10^6 \text{ km}^3 = 6.4 \times 10^6 \text{ km}^3$ is obtained. Using a ΔC_1 value of 51 µmol/kg, a total carbon inventory of about 3.9 GtC can be derived. For the estimate of the mean penetration depth Broecker and Peng (1993) suggest an uncertainty of about 20%. Adopting the range of values of $\Delta \delta_s / \Delta C_t$ as its error (about 20%), we derive an estimate of the a total anthropogenic carbon uptake of the Arctic Ocean of about 4 + 2 GtC.

In a second scenario the "carbonate ion effect" will be included. The δ^{13} C signature of planktic foraminifera is strongly dependent on the carbonate chemistry of the ocean (Spero et al., 1997). Culturing results of *G. bulloides* reveal an enrichment in δ^{13} C by 1.3‰ for every 100 µmol/kg decrease in carbonate ion concentration [CO₃²⁻]. If we assume a similar response for *N. pachyderma* (sin.), Lea et al. (2000) suggest an even stronger response, the Suess effect in the δ^{13} C_{DIC} is even larger than the observed -0.9‰ in *N. pachyderma* (sin.) and partly masked by the carbonate ion effect (CIE) in the planktic foraminifera. If we start off with an increase of C_t by 50 µmol/kg and constant total alkalinity at conditions observed in the Arctic Ocean halocline waters, the change in [CO₃²⁻] and concomitant CIE can be calculated (see Table 2), thus changing the value of the deduced Suess effect. By iteration a Suess effect of -1.6‰and a change in C_t of 90 µmol/kg is observed, implying a total anthropogenic uptake of the Arctic Ocean of about 7 ± 3 GtC. Adopting the CIE from culturing experiments of *G. bulloides* for *N. pachyderma* (sin.) leads to a surface ocean Suess effect, which is with -1.6‰ even larger than the estimate of the Suess effect in the

Iteration	$\Delta \delta_{ m s}$ (‰)	ΔC_{t} (µmol/kg)	Δ [CO ₃ ²⁻] (µmol/kg)	CIE (‰)	Suess effect (‰)
1	- 0.9	50	- 29	0.38	- 1.28
2	-1.28	73	- 43	0.56	-1.46
3	-1.46	83	- 49	0.64	-1.54
4	- 1.54	88	- 52	0.68	-1.58
5	-1.58	90	- 54	0.70	-1.60
6	-1.60	91	- 54	0.70	-1.60

Table 2 Iterative calculation of the carbonate ion effect $(CIE)^a$

^aFor the first iteration the isotopic change caused by the Suess effect $\Delta \delta_s$ is taken as the observed isotopic change in *N. pachyderma* (sin.) of -0.9%. ΔC_t is calculated using a constant ratio $\Delta \delta_s / \Delta C_t$ of -0.0175%. kg/µmol. Assuming constant alkalinity over the last 150 years and using mean values observed in the Nansen Basin ($A_t = 2280 \ \mu mol/kg$; $C_{t-akt} = 2150 \ \mu mol/kg$; $T = 0^{\circ}C$ and S = 34; e.g. Anderson et al., 1990) the concomitant change in carbonate ion concentration $[CO_3^2^-]$ is calculated (Ware et al., 1991). Adopting a carbonate ion effect of 1.3% increase in $\delta^{13}C$ for every 100 µmol/kg decrease in carbonate ion concentration $[CO_3^2^-]$ (Spero et al., 1997) a $\delta^{13}C$ change due to the CIE is derived. The Suess effect is calculated by subtracting the CIE value and $\Delta \delta_s$. In the following iterations the previously derived Suess effect is taken as the new $\Delta \delta_s$.

atmosphere of -1.4% (Friedli et al., 1986; Francey et al., 1999). This could be caused by the roughness of estimates and the assumptions made for our calculations. Nevertheless, the Revell factor in the Arctic Ocean halocline waters limits the anthropogenic CO₂ uptake to a smaller amount. A possible explanation is the formation of the halocline waters on the Siberian shelf areas. Measurements of C_t and estimates of A_t on the shelf areas (Erlenkeuser et al., 1995; Olsson and Anderson, 1997) indicate a smaller Revell factor, allowing a considerably higher CO₂ uptake from the atmosphere. There is a net flux of carbon from the shelf areas into the Arctic Ocean halocline waters (Anderson et al., 1990) caused by the input of C_t from river water and a possibly equally high fraction, which has its origin in decay of total organic carbon (DOC) (Olsson and Anderson, 1997). Since, most of the source of carbon incorporated in organic matter has to be atmospheric CO₂ and C_t added by seawater (Olsson and Anderson, 1997), the decay of DOC at the sediment surface in the shelf areas might be another suitable source to transport additional anthropogenic CO₂ into the Arctic Ocean halocline waters.

The Arctic Ocean covers about 2% of the worlds ocean area, and our estimate suggests that it accounts for about 4% (without CIE) to 6% (including the CIE) of the total uptake of anthropogenic CO₂ in the worlds ocean (taken as 110 GtC, Sarmiento et al., 1995). This value is much higher than other estimates: Carbon budgets derived for the Arctic Ocean (Anderson et al., 1998; Lundberg and Haugan, 1996) also can be used to deduce an uptake rate of anthropogenic CO₂. The results of Anderson et al. (1998) indicate that the uptake rate of the Arctic Ocean amounts to only about 1% of the globally averaged anthropogenic CO₂ uptake rate of about 2 GtC/yr (Siegenthaler and Sarmiento, 1993).

6. Conclusion

Values of δ^{13} C of *N. pachyderma* (sin.) from the water column in the Arctic Ocean are depleted relative to values from core top sediments on average by -0.9_{00}° ($\pm 0.2_{00}^{\circ}$) within the Arctic Ocean halocline waters and -0.6_{00}° ($\pm 0.1_{00}^{\circ}$) in the Atlantic-derived waters of the southern Nansen Basin. This decrease of δ^{13} C of *N. pachyderma* (sin.) reflects the decrease of δ^{13} C_{DIC} in ocean surface water and is caused by the addition of anthropogenic CO₂ to the atmosphere and the concurrent decrease of δ^{13} C values of atmospheric CO₂ by 1.4₀₀ (Suess effect). Addopting a "carbonate ion effect" (CIE) obtained for *G. bulloides* (Spero et al., 1997), the Suess effect might be partly masked and as high as -1.6_{00}° .

 $\delta^{13}C_{DIC}$ values are far off from isotopic equilibrium values with the atmosphere. Absolute values of *N. pachyderma* (sin.) covary with the surface ¹³C Suess effect and both reflect the degree of ventilation of the ocean water with respect to CO₂. This implies that the Arctic Ocean halocline waters are to a better degree ventilated with respect to CO₂ than the Atlantic-derived waters in the southern Nansen Basin. A comparison of our estimate of $\delta^{13}C_{DIC}$ data with $\delta^{13}C$ values of *N. pachyderma* (sin.) reveals an offset of about 1‰ within the Arctic Ocean, which conflicts with results of other studies (Kohfeld et al., 1996). Basic studies concerning carbon isotope fractionation in foraminiferal shells are of great importance for paleoceanographic studies and should be further conducted in respect to variations of the "vital effect" as well as the "carbonate ion effect".

An estimate of a total anthropogenic carbon inventory indicates that the Arctic Ocean, despite of its permanent sea-ice coverage, appears to take up more than its share of the anthropogenic CO_2 and is considerably higher than previous estimates. While the derived inventory value is only a rough estimate, we hope that our data will help future modelling studies that are necessary to give global estimates of the fate of fossil fuel-derived CO_2 and future climate.

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