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Oceanic uptake of $CO₂$ re-estimated through $\delta^{13}C$ in WOCE samples

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Abstract

In addition to ¹⁴C, a large set of $\delta^{13}C$ data was produced at NOSAMS as part of the World ocean circulation experiment (WOCE). In this paper, a subset of 973 δ^{13} C results from 63 stations in the Pacific Ocean was compared to a total number of 219 corresponding results from 12 stations sampled during oceanographic programs in the early 1970s. The data were analyzed in light of recent work to estimate the uptake of $CO₂$ derived from fossil fuel and biomass burning in the oceans by quantifying the $\delta^{13}C$ Suess effect in the oceans. In principle, the $\delta^{13}C$ value of dissolved inorganic carbon (DIC) allows a quantitative estimate of how much of the anthropogenic $CO₂$ released into the atmosphere is taken up by the oceans, because the δ^{13} C of CO₂ derived from organic matter (\sim –27%) is significantly different from that of the atmosphere ($\sim -8\%$). Our new analysis indicates an apparent discrepancy between the old and the new data sets, possibly caused by a constant offset in δ^{13} C values in a subset of the data. A similar offset was reported in an earlier work by Paul Quay et al. for one station that was not included in their final analysis. We present an estimate for this assumed offset based on data from water depths below which little or no change in $\delta^{13}C$ over time would be expected. Such a correction leads to a significantly reduced estimate of the $CO₂$ uptake, possibly as low as one half of the amount of 2.1 GtC yr^{-1} (gigatons carbon per year) estimated previously. The present conclusion is based on a comparison with a relatively small data set from the 70s in the Pacific Ocean. The larger data set collected during the GEOSECS program was not used because of problems reported with the data. This work suggests there may also be problems in comparing non-GEOSECS data from the 1970s to the current data. The calculation of significantly lower uptake estimates based on an offset-related problem appears valid, but the exact figures are tentative because the data set is small and the cause for an offset remains unknown. Therefore, it would be desirable to extend this comparison to the Indian Ocean where it is believed that better GEOSECS δ^{13} C data are available. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Human-induced $CO₂$ is a major concern in regards to possible effects on the global climate. It

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has, however, not been possible to establish a consistent scenario of the fate of this $CO₂$. Atmospheric measurements [1] indicate that the increase of $CO₂$ is only about 57% of the $CO₂$ emitted; the remainder must be dispersed into the oceans and/or the biosphere. The percentage of this has been subject to intense research efforts, with estimates ranging from a predominant uptake by the oceans to a negligible ocean $CO₂$ sink [2–6]. Since the two reservoirs differ significantly in size, turnover rate, and feedback characteristics, it would be desirable to have a firm estimate of this quantity. Measurements of δ^{13} C signatures in the various reservoirs in principle provide means to achieve this.

Atmospheric CO₂ currently has a δ^{13} C value of about -8% and the δ^{13} C of CO₂ released by combustion of fossil fuel and biomass is about -27% . The reason for this difference is the isotopic fractionation during photosynthetic fixation of $CO₂$ by (predominantly terrestrial) $C₃$ plants [7]. The massive combustion of fossil fuel starting at about 1800 has led to a gradual decrease over time of the δ^{13} C in the atmosphere known as atmospheric δ^{13} C Suess effect [8]. Presently, it follows an exponential trend with similar time constants as the change in atmospheric $CO₂$ [4]. Exchange between the atmosphere and the ocean surface (aided by less dominant fluxes caused by river runoff and precipitation) causes this decrease in δ^{13} C to propagate into the oceans as well.

Three major δ^{13} C-based models are currently known: the `inventory method', proposed by Quay et al. [2], the `isotopic disequilibrium method', proposed by Tans et al. [3] and the `dynamic constraint method' published by Heimann and Maier-Reimer [4]. Based on data available at the time of publication, the models yielded estimates for the oceanic carbon uptake in the time period 1970±1990 of 2.1, 0.2 to 1.1 (depending on parameter chosen for fractionation factors, discussed in more detail below), and 3.1 GtC yr^{-1} (gigatons carbon per year), respectively $(1 \text{ Gt} = 10^{15} \text{ g})$. Another method introduced by Bacastow et al. [5] uses a surface ocean δ^{13} C time series at Bermuda in combination with models for extrapolating to a global carbon uptake estimate. A new method proposed by Sonnerup et al. [6] utilizes synoptic

 δ^{13} C data for calculating 'preformed' δ^{13} C along isopycnal surfaces. The rate of δ^{13} C change is determined by using water ages calculated from concurrent CFC measurements. This method has the advantage of not relying on data from the past, but it is sensitive to mixing biases and dating issues. The estimate derived from this method is 1.9 ± 0.9 GtC yr⁻¹. The current intergovernmental panel on climate control (IPCC) estimate is 2.0 ± 0.8 GtC yr⁻¹ [9]. A short introduction to the disequilibrium and dynamic constraint method will be given when the results of our investigations are discussed. The inventory method is re-evaluated in this paper in the light of our larger data set.

2. Discussion of input data

The Inventory Method proposed by Paul Quay et al. [2] (in short called PQ) is based on the global averaged depth-integrated change of δ^{13} C in the oceans. PQ estimated this quantity from changes in depth-profiles over 20 years in the Pacific ocean. The earlier set was measured during the HUD-SON-70 (1970) cruise along 150° W, SCAN-X (1970) along 90°W±114°W, Tow-7 (1970) along $165^{\circ}W-171^{\circ}W$ and Antipodes-15 (1971) along 172°W [10,11]. The larger data set of the GEO-SECS δ^{13} C Pacific data (1977) is considered problematic [10,11] and, therefore, excluded from further analysis, leading to a sparse 1970s δ^{13} C data set in the Pacific. The later set of data used by PQ was measured during three National Oceanographic and Atmospheric Administration (NOAA) research cruises (1989, 1990, 1991) [2]. For this analysis, 17 profiles from the 1970s (293) samples) of the Pacific were available, seven of which (127 δ^{13} C measured samples total) were considered suitable for comparison with seven corresponding 1990s NOAA profiles (124 δ^{13} C samples).

The Pacific World Ocean Circulation Experiment (WOCE) program provides oceanic δ^{13} C data, because δ^{13} C was measured with high precision at NOSAMS in addition to Δ^{14} C [12,13]. These data can be used to revisit the inventory method. The WOCE Pacific seawater data includes a total of 6423 δ^{13} C measurements carried

out by NOSAMS on 6332 distinct samples from 358 stations. Details of the sample preparation are given in [14]. The samples were collected between March 1991 and November 1993, with the exception of 185 measurements at 11 test stations (cruise P01). This enhanced data set allows comparison with a larger subset of the 1970s profiles, and the geographical distance between the old and new profiles can be reduced. Moreover, there are several WOCE profiles suitable for comparison, so that groups of 1990s profiles can be averaged for enhanced accuracy. In addition to the geographical distance, the salinity and temperature data are important for the selection of profiles to identify the stations within the same body of water. As a result of this preselection a total of 63 WOCE stations were found to be usable, featuring 973 data points; these are compared to 12 stations from the 1970s including a total of 219 data points (Fig. 1).

3. Data evaluation

The evaluation is based on the integration of the difference in δ^{13} C between 1970 and 1990 over the entire water column. Human-induced changes should vanish for water masses below the penetration depth corresponding to a time span of 200 years. Because of this, values of δ^{13} C measured in the 1970s and 1990s should approach each other as the water column gets deeper and the integrated

Fig. 1. The location of the stations in the Pacific Ocean used for analysis. (\bullet : 1970s, X: PQ, \circ : WOCE data).

result should approach a constant value with increasing integration depth. Unexpectedly, a remarkably constant offset of about 0.2% between the δ^{13} C values is visible in most of the compared profiles (Fig. 2). The offset is best visible at depths between 800 and 1400 m (the possibility of a layer

Fig. 2. The change in the depth distribution of the $\delta^{13}C$ of the dissolved inorganic carbon (DIC) in the Pacific Ocean. The individual figures (a-l) are ordered by latitude going from north to south. All 1970 data are marked by full diamonds (\bullet). An offset of ~0.2‰ is apparent in the data, except for SCAN-X 43 (h) and SCAN-X 30 (i). HUDEQT (f) and SCAN-X 20 (g) do not permit firm conclusions in respect to a possible offset.

of stability in this depth range is mentioned in [15]). Although in some instances a change in δ^{13} C over a 20-year time period induced by the Suess effect may extend to this depth range, such a change would be expected to be small compared to an assumed 0.2% offset. A real change over time is expected in the water bodies above this depth. There is no strong correlation between the 1970s and 1990s deep water data. Possible explanations

for this are sparse data for these depths or sampling of different water masses [15]. Problems are imposed by the three analyzed profiles of the SCAN-X cruise, where no offset is visible for two profiles (Fig. 2(h) and (i)) and one, $SCAN-X$ 20 remains unclear (Fig. $2(g)$). A tentative explanation could be that there was no offset problem with SCAN-X cruise data. An analysis of the individual WOCE stations compared to SCAN-X 20 (0.6°N, 86°W) reveals a large variability of up to 0.4% within those stations at a proximity of less than one degree latitude, likely caused by the equatorial current system. For this reason, the station is not used in our calculations. As for the HUDEQT profile (Fig. $2(f)$) data exists only above 240 m, no conclusive offset determination is possible. For the other seven HUDSON and the one TOW stations a consistent offset of about 0.2% is visible.

The magnitude of the offset was estimated based on δ^{13} C data between 800 and 1400 m for a subset of five station pairs where the 1970 and 1990 δ^{13} C data appear completely parallel: HUD 300, HUD 297, TOW7 146, HUD 282, HUD 277 (any deviation from a parallel shift may be an indication for a true δ^{13} C change). The average offset estimated from these five stations is 0.206. If there is a Suess effect between these depths, then it would make our estimate of the offset a maximum value. To estimate the uncertainty, we calculate the external error of the five offsets obtained for the individual stations. The standard deviation is $\pm 0.027\%$, the external uncertainty of the average, therefore, $\pm 0.012\%$. To check how the selection of the stations influences this result, we also calculated the offset for all stations except SCAN-X 20, SCAN-X 30, SCAN-X 43, and HUDEQT in the same depth interval. The result, $0.204 \pm 0.015\%$ is approximately the same (Fig. 3).

The results of the depth integration with a 0.206 offset correction applied to all 1970s stations except SCAN-X 30 and SCAN-X 43 data (under the preliminary assumption that the offset is in the 1970s data) are given in Table 1. Fig. 3(a) shows the integration of the individual profiles, Fig. $3(b)$ the same data corrected for a 0.206% offset.

In order to relate the unexpected results to [2], a recalculation using the data of [2] was performed (Fig. 3(c)). The results reported in [2] are marked by vertical bars in Fig. 3(c). Even with the additional help of temperature-salinity plots (where available) it was not possible to establish the criteria the authors used to select the integration depths.

3.1. Additional evidence for a 0.2% $\delta^{13}C$ offset

(1) A real change in the δ^{13} C (as opposed to an offset) is likely to be accompanied by a corresponding change in the Δ^{14} C data as both the characteristic penetration times of fossil fuel $CO₂$ and bomb radiocarbon during the early 90s are about 30 years $[16]$. Analyzing the difference between GEOSECS and WOCE Δ^{14} C data revealed no such changes for the examined stations, except for the one station close to Hawaii (TOW-7 146, 20N 166W). GEOSECS data for this station is located west of Hawaii $(161-177W)$, WOCE data east of Hawaii (152-155W) with data for both stations located at latitudes between 14N and 22N. This could be a contributing factor to the observed difference in Δ^{14} C.

(2) A constant offset of 0.2% between the stations HUDSON 301 and CGC91-58 is also reported in [2]. The data of these stations were discarded because of the observed offset.

(3) Two large surface water δ^{13} C data sets are available for the meridional transect at 150°W, Hudson-70 (1970) and WOCE P16 (1991/92) (Fig. 5). Apparent is a remarkably constant decrease of about 0.5% over almost the entire transect. For moderate latitudes, Δ^{14} C models predict a surface water change of about 0.7 times the observed δ^{13} C change of 0.4% in the atmosphere [17]. The geographically weighted averaged δ^{13} C decrease is 0.49% . If this value is assumed to be 0.20% too high due to an offset the corrected

Fig. 3. Depth integrated $\Delta\delta^{13}C$ plotted as a function of integration depth. (a) exibits the result without any offset correction. (b) shows the same data corrected for a presumed $\sim 0.20\%$. (c) is a plot of the original data used in [2] (the vertical bars indicate the results reported in this paper).

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change of 0.29% is in close agreement with this predicted 0.28% . The corresponding -0.013% yr^{-1} average surface ocean δ^{13} C decrease however appears to be somewhat low compared to current estimates (global ocean average surface δ^{13} C rate of change of $-0.15 \pm 0.4\%$ decade⁻¹ [6], -0.171% decade⁻¹ [5], -0.018_{/00} yr⁻¹ [18]). Another argument is possibly supplied by regions of upwelling water (subpolar ocean, equatorial zone, see also Fig. 5), where the human-induced change in $\delta^{13}C$ would be expected to be small. This is only the case if the 0.20% offset correction is applied to the Hudson-70 data (Fig. 5). A limiting factor to onetime surface data analysis are short-term, seasonal and interannual variability of surface ocean $\delta^{13}C$ [5,19].

 (4) The possibility of an offset has been pointed out before [3], making an offset not entirely unexpected, although the magnitude would be much higher than expected.

(5) The fact that the change is practically constant at about 0.20 over a significant depth interval makes it unlikely to be attributed to real changes as those would be expected to be varying to some degree as those observed in the main thermocline.

4. Estimation of the worldwide $\Delta\delta^{13}$ C average

The average of all stations, weighted by the Pacific area of the respective 10° latitude belt and with all stations normalized to a Δt of 20 years is $-93 \pm 7\frac{\nu}{\omega}$ m. This, however, cannot be used to reliably infer the average change in the Pacific as it is unknown how well these stations represent the average Pacific. A reasonable assumption appears to be to use all data except those from the highest latitudes and those of questionable quality $SCAN-X 20$ where the existence of the offset is uncertain and HUDEQT where data are available only for a depth of 240 m, also see Fig. 2). This leads to $114 \pm 8\%$ m for the area-weighted average. Without correlation to another tracer with a globally measured distribution it is difficult to reliably infer the average Pacific or World Ocean change from these data. For example, another tracer, Δ^{14} C, exhibits a longitudinal trend with a higher 14 C burden in the western Pacific [20]. If a

similar trend applied to δ^{13} C, our calculations would be biased since 8 of 12 stations of our data set are located at 150°W. Also because there are no data between 10°S and 40°S, only three stations (at 40, 50 and 63° S) represent the Pacific south of 10°S.

Following [2], the $\Delta\delta^{13}$ C values for 11 profiles were correlated with the bomb-test induced Δ^{14} C changes by means of linear regression (Fig. 4). This allows one to estimate the global δ^{13} C change from the worldwide average ${}^{14}C$ burden, assuming a linear correlation between $\Delta \delta^{13}$ C and Δ^{14} C. The analysis was performed using 14C data reported in [20] (as this data set was used in [2]) and a later, improved data set [16]. The worldwide 14C burden averages reported are 8.4×10^9 atoms cm⁻² [20] and 9.2×10^9 atoms cm⁻² [16], respectively. The mean inventory values reported for the Pacific were on average 14.7% higher in [20] than in [16]. This relationship is used to avoid problems arising from extrapolating from the relatively small number of δ^{13} C profiles in the 1970s to a global estimate [2].

In our data, the correlation between the bomb ¹⁴C burden and depth-integrated δ^{13} C changes using the method of [2] is weak (Fig. 4). The following explanations are conceivable: (1) the offset correction, as applied, is not correct. The data in the 800–1400 m depth range provide strong evidence for an offset, but it is impossible to tell whether the offset is constant throughout the water column and, in particular, in the surface ocean where time changes are expected. Although it may seem reasonable to assume the offset is an analysis or calibration problem, this may not be the case and could be a factor in the observed relationship. (2) The locations and times of δ^{13} C and Δ^{14} C sampling and analysis differ too much. The δ^{13} C analysis covers a time frame between about 1970 and 1990, whereas the Δ^{14} C analysis estimates the change between pre-bomb (about 1955) and 1970. Also the δ^{13} C and Δ^{14} C data for the early 1970s data set were not sampled at the same locations, in some cases no nearby stations are available as the spatial resolution for Δ^{14} C is not very dense. As there is frequently noticeable variance in Δ^{14} C between nearby stations, issues of interpolation between adjacent stations are

Fig. 4. The correlation between the depth-integrated changes in $\Delta\delta^{13}C$, normalized to $\Delta t = 20$ yr, and bomb-produced $\Delta^{14}C$ where both the ¹³C and ¹⁴C changes have been determined. The ¹⁴C data from this plot is adopted from [15]. The ¹⁴C was interpolated by building the arithmetic mean within 10° latitude belts and interpolating between two adjacent belts. It is apparent that the $^{13}C^{-14}C$ correlation in the data is weak. The linear regression, chosen as the fit method (represented by the long-dashed line), is defined by $\Delta \delta^{13}C = -15.7\Delta^{14}C + 37.0$, where $\Delta \delta^{13}C$ is in units of $\frac{9}{20}$ m and $\Delta^{14}C$ in atoms cm⁻² (r², the usual measure of goodness-of-fit is 0.33, the standard error of the y estimate is 55.3% m). Due to the weak correlation the linear regression fit exhibits a bias: also shown is the linear regression with $\Delta^{14}C$ as a function of $\Delta\delta^{13}C$ (short-dashed line) which leads to a fit of $\Delta\delta^{13}C = -48.1\Delta^{14}C + 279.0$.

Fig. 5. The decrease of the δ^{13} C values of dissolved inorganic carbon in the mixed layer (the surface water exchanging CO₂ with the atmosphere) of the Pacific Ocean between 1970 and 1991/92.

raised. (3) There is indeed only a weak correlation. The likelihood of (3) has been previously pointed out in [3] (different uptake history of

bomb ¹⁴C versus continuous δ^{13} C input and δ^{13} C fluxes for which there is no 14 C equivalent [21,22]). This weak correlation can also be

observed in three-dimensional ocean carbon cycle model simulations [4]. Tentative results of analyzing the WOCE-GEOSECS Δ^{14} C change with respect to δ^{13} C may indicate that these changes during the relative short time interval of about 10 years between the bomb peak and the GEOSECS Δ^{14} C inventory may indeed not well track longterm distribution phenomena driving the δ^{13} C changes over several decades. The combined effect of (2) and (3) may contribute to explain the weak correlation shown in Fig. 4.

In the plot in Fig. 4, the δ^{13} C change associated with the global bomb 14 C burden of 9.2×10^9 atoms cm⁻² is $107 \pm 23\%$ m. However, further analysis revealed that this value depends on the method used to select the 14C data and even more so on the method of fitting the data. The problems with the linear regression can be best demonstrated by recalculating the linear regression using ¹⁴C as the dependent and δ^{13} C as the independent variable. This yields a significantly different fit $(Fig. 4)$. The difference between the two fits is an indication for a weak correlation [23]. An additional uncertainty arises from the method which is used to estimate the Δ^{14} C values from the available Δ^{14} C data. We applied different methods: (1) simply select the closest station for comparison, (2) calculate the average of a handselected set of nearby stations, (3) use linear interpolation between the two closest stations, (4) calculate the arithmetic mean 14C burden for the neighboring 10° latitude belts and interpolate between these results, (5) use the Δ^{14} C data reported in [20] and [16] for (3) and (4). This sensitivity check suggests an average δ^{13} C change of -138% m. From the scatter of the results obtained with different methods, and linear fits using both δ^{13} C and Δ^{14} C as the dependent variable, we adopt an uncertainty of $\pm 50\%$ m. This translates to a worldwide average depth-integrated δ^{13} change of $-6.9 \pm 2.5\%$ m yr^{-1} . Due to the weak correlation this result is tentative.

The contribution to the overall uncertainty introduced by the uncertainty of the offset correction, which is correlated for all stations, is estimated by multiplying the typical integration depth (1100 m) with the uncertainty of the offset $(\pm 0.015\%_{\odot})$: $\pm 17\%_{\odot}$ m.

To obtain the global carbon uptake we redo the calculations in [2] with $-138 \pm 50\%$ m for the average worldwide depth-integrated δ^{13} C change. Without modifying any other parameter and applying a Monte Carlo method for estimating the uncertainty as in [2] this results in an uptake estimate of 1.0 ± 0.9 Gt C yr⁻¹ which is adopted as the final estimate.

A correct method for estimating the worldwide average from the given δ^{13} C data involves satisfactory modeling of high-resolution oceanographic processes which likely cannot be done satisfactorily with a simple linear fit. Regardless of the difficulties to give an exact estimate we wish to emphasize the fact that any reasonable figure must be significantly lower than the figure given in [2].

4.1. Comparison with [2] and other methods

For the five stations (HUD 297, TOW 146, HUD EQT, HUD 282, HUD 280), where the same 1970s data set is used as in [2] and which, therefore, can be directly compared to the results reported here, the depth-integrated δ^{13} C change is on average 47% of the depth-integrated change reported in [2], with HUD 282 and HUD 280 comparatively low at only about 20%. Generally, the depth-integrated δ^{13} C change for the individual stations, after the offset correction, appears to be roughly 50% of the values reported in [2]. Based on the offset correction and the correlation to the bomb 14 C burden, the worldwide depthintegrated δ^{13} C average change (-138 ± 50% m between 1970 and 1990) is 66% of the result reported in [2] $(-208 \pm 45\%_{\text{oo}})$. By repeating the model calculations and the error estimate, this results in an worldwide oceanic uptake of 1.0 ± 0.9 GtC yr⁻¹, 48% of the 2.1 \pm 0.8 reported in [2]. While this appears to be low it is still within one standard deviation of current estimates. Table 2 summarizes other estimates for the oceanic uptake of $CO₂$. We note that the dynamic constraint method [4] also uses the global $\Delta\delta^{13}$ C as an input parameter and should therefore be revisited, as it results in a lower uptake estimate as well.

Table 2 Oceanic uptake of CO_2 deduced from δ^{13} C using different approaches

Group	Year	Ref.	Approach	Oceanic uptake (Gt C yr^{-1})
Quay et al. Tans et al.	1992 1993	$[2]$ $[3]$	Inventory method Isotope disequilibrium method Fractionation from [24]	$2.1 + 0.8$ 0.2 1.1
Houghton et al. Heimann and Maier-Reimer	1995 1996	$[9]$ [4]	IPCC, Climate change 1994 Dynamic constraint method HAMOCC3	2.0 ± 0.8 3.10 ± 1.63 1.29, 1.33
Bacastow et al.	1996	$[5]$	Extended surface ocean time series	$0.8 + 0.2$ $1 - 4$
Sonnerup et al. This work	1999 2000	[6]	Preformed δ^{13} C Inventory method	$1.9 + 0.9$ 1.0 ± 0.9

5. Summary and outlook

Our findings provide evidence that there is a systematic offset of about 0.2 between a large subset of the δ^{13} C data of seawater samples analyzed in the early 1970s and in the early 1990s (WOCE). The reason for this offset is unknown and we refrain from speculation about the cause, yet there are indications that the problem lies in the 1970s HUDSON (and possibly TOW) data. Applying a correction for this offset drastically changes the results of two different methods for estimating the oceanic $CO₂$ uptake. The results of the inventory method are reduced to possibly as much as one half. This result implies a much larger non-oceanic $CO₂$ sink (plants and soil) than suggested by current estimates. Due to the limited amount of data for the 1970s, remaining doubts relating to corrections made for an assumed offset of unknown cause and uncertainties in estimating the worldwide average of the depth-integrated δ^{13} C change from the available stations the findings are preliminary at their exact figures. Further research is needed to refine the data. There is a data set for the Indian Ocean which is well suited for testing the accuracy of the present findings.

In conclusion, estimates about oceanic uptake of $CO₂$ as depicted in Table 2 are still dominated by large uncertainties. This may reflect primarily our limited knowledge about the differences in regional uptakes as one goes around the globe. Improved data combined with improved modeling taking these differences into account is probably the most promising approach to arrive at more firm predictions.

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