



Evaluating Changes of the Anthropogenic Carbon Inventory in the Northwestern Pacific using $\delta^{13}\text{C}$

Solomon Wilson

¹*University of Washington, School of Oceanography,
Box 355351, Seattle, Washington 98195
Solomonw2@gmail.com
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NONTECHNICAL SUMMARY

Carbon dioxide concentrations in both the atmosphere and ocean have been increasing recently due to the burning of fossil fuels. In order to predict and understand future climate the inventory of carbon dioxide from fossil fuels needs to be assessed. The isotopic composition of carbon from fossil fuels is distinct from that of carbon found in the ocean, and is therefore useful as a tracer of anthropogenic carbon. Two sets of carbon isotope data are available along a latitudinal transect of the Northwestern Pacific from the years 1994 and 2005. Using an equation to predict the carbon isotopic composition from one data set to the other, we can calculate the change in carbon isotopes. The carbon isotope changes can then be calculated into changes in anthropogenic carbon inventory in the study area.

ABSTRACT

Because carbon dioxide from fossil fuels is depleted in ^{13}C , it can be used to track ocean uptake of anthropogenic carbon. The P10 track of the World Ocean Circulation Experiment (WOCE) runs from 4°S to 42°N in the western Pacific, and has two data sets containing nutrient and $\delta^{13}\text{C}$ data. The P10 track has over 100 stations, and CTD casts are either done down to 2000m or a full cast of the water column. An isopycnal Multiple Linear Regression (isoMLR) was used to separate anthropogenic changes and natural variability. The mean depth integrated $\delta^{13}\text{C}$ change is calculated to be $-12.04 \text{ ‰ m yr}^{-1}$ over the P10 cruise track. Using an RC value of $-0.021 \pm 0.004 \text{ ‰ } (\mu\text{molC/kg})^{-1}$ from the Hawaii Ocean Time Series (HOTS) the rate of anthropogenic carbon uptake is calculated to be $0.20 \mu\text{molC } (\text{m}^2 \text{ yr})^{-1}$.

INTRODUCTION

The globally averaged temperature is expected to increase by $2\text{-}3^{\circ}\text{C}$ (IPCC, 2007), which is primarily attributed to increase in carbon dioxide concentrations in the atmosphere. Growth of emissions has increased from 1.3% to 3.3% each year over recent decades (Canadell et al., 2007). Increases in pCO_2 in the atmosphere throw

the air-sea gas exchange out of equilibrium, leaving the oceans undersaturated in CO_2 (Sabine and Tanhua, 2010). This leads to a net flux of CO_2 into the ocean from the atmosphere. Currently, the ocean pCO_2 is increasing at an average rate of $1.5 \mu\text{atm yr}^{-1}$ (Takahashi et al., 2009). The uptake flux of anthropogenic CO_2 by the ocean has been estimated to be around 2.2 Pg C yr^{-1} (Gruber et al. 2009). Since preindustrial times it has been estimated that the ocean has taken up 41% of emitted fossil fuels (Sabine and Tanhua, 2010). As

the $p\text{CO}_2$ of the atmosphere continues to rise, the ocean becomes ever more important in the uptake of carbon. The ocean is an important buffer for the atmosphere, but it is not able to continue storing carbon at the same rate that emissions are increasing.

The ratio of $^{13}\text{C} / ^{12}\text{C}$ (henceforth referred to as $\delta^{13}\text{C}$) is a useful tracer for anthropogenic carbon in the ocean. $\delta^{13}\text{C}$ is calculated by dividing a measured $^{13}\text{C} / ^{12}\text{C}$ ratio divided by a standardized $^{13}\text{C} / ^{12}\text{C}$ ratio minus 1 all multiplied by 1000. The units are reported in parts per thousand, or per mil (‰). During photosynthesis plant fractionate carbon, using $^{12}\text{CO}_2$ preferentially over $^{13}\text{CO}_2$ (O’Leary, 1981). This leads to a smaller ratio of $^{13}\text{C} / ^{12}\text{C}$ of the organic matter produced by photosynthesis. Stated another way, the carbon being fixed during photosynthesis is depleted of ^{13}C . When combusting fossil fuels, which are derived from plant material, ^{13}C depleted carbon is introduced into the atmosphere which decreases the atmospheric $\delta^{13}\text{C}$. When the burned fossil fuels in the atmosphere sink into the ocean as DIC, the isotopic signature is introduced into seawater. Takahashi et al. (2000) observed depletion of ^{13}C from 15°S to 48°N along 175°W in the Pacific. Quay et al. (2007) makes this same observation in the Atlantic.

The Northwestern Pacific Ocean is a significant source of anthropogenic carbon uptake. The Kuroshio lies with this region and combined with the Kuroshio extension is the greatest sink of anthropogenic carbon in the entire Pacific Ocean (Hideyuki et al., 2011). In addition to being a significant sink of carbon in the North Pacific, the Kuroshio also forms Mode Waters, which have the largest quantity of anthropogenic CO_2 in the Pacific (Sabine et al., 2004; Takahashi et al., 2010). The anthropogenic inventory of carbon in the Pacific ranges from nearly 0 molC m^{-2} to 25 molC m^{-2} .

The World Ocean Circulation Experiment (WOCE) has a compilation of tracks that run all over the world’s oceans. These tracks are often repeated on a decadal time scale to provide more recent data. On these cruises various sea-water properties are measured and recorded. In our region of interest, the P10 line will be of the most use (Fig. 1). This line runs from the Northern tip of Papua New Guinea near the equator to the eastern coast of Japan. With the release of WOCE $\delta^{13}\text{C}$ data along the P10 line from 1993 and 2005, there is an exciting new opportunity to quantify the increase of anthropogenic carbon using changes in $\delta^{13}\text{C}$ over recent decades in this region.

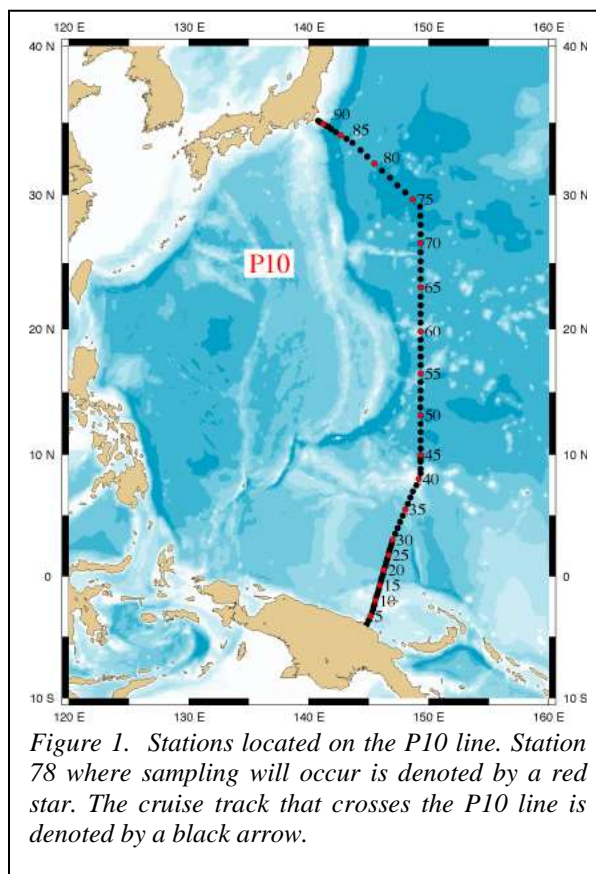


Figure 1. Stations located on the P10 line. Station 78 where sampling will occur is denoted by a red star. The cruise track that crosses the P10 line is denoted by a black arrow.

METHODS

WOCE P10 repeat hydrography – WOCE is a part of the World Climate Research Program. Under WOCE, many transects of different ocean

basins have been surveyed for hydrographic properties. The P10 transect runs north to south along the western North Pacific (Fig. 1). P10 was surveyed in 1994 and 2005. There are 136 stations listed in P10, but only 19 were sampled in 2005 and 38 stations in 1994. The 1994 cruise consists of 5 deep casts reaching a maximum depth of 6070db and the rests are shallow (<2000m) casts. In the 2005 cruise 9 of the 19 stations were deep casts. This information can be found at http://cdiac.ornl.gov/oceans/woce_p10.html

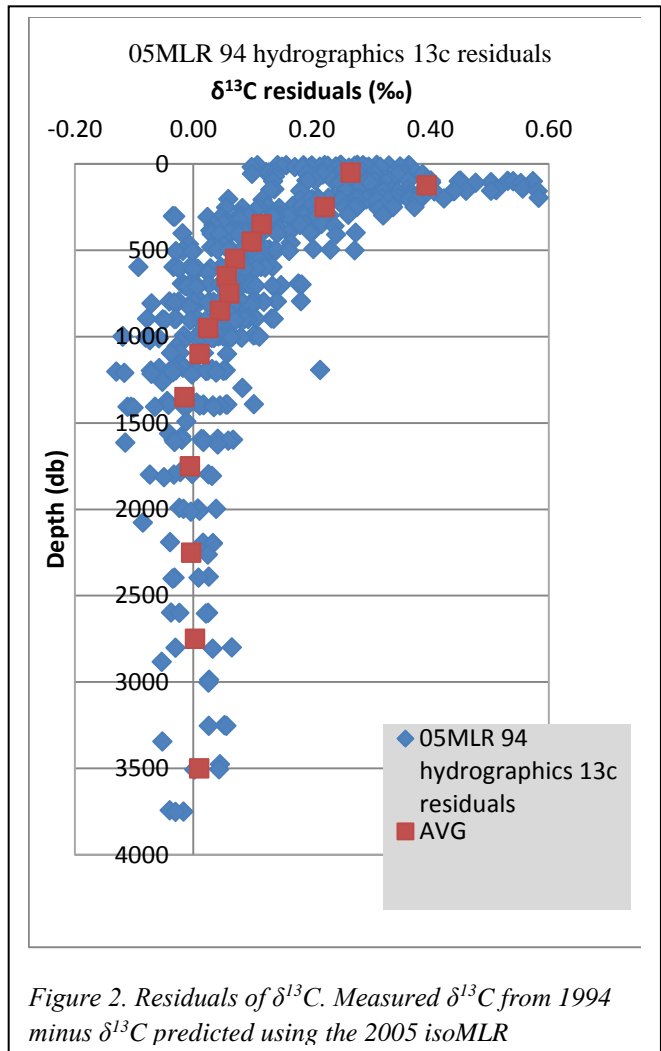
d¹³C MLR Analysis – A Multiple Linear Regression (MLR) is used to differentiate between natural variability in *d¹³C* and anthropogenic forcing. The equation used for the MLR is

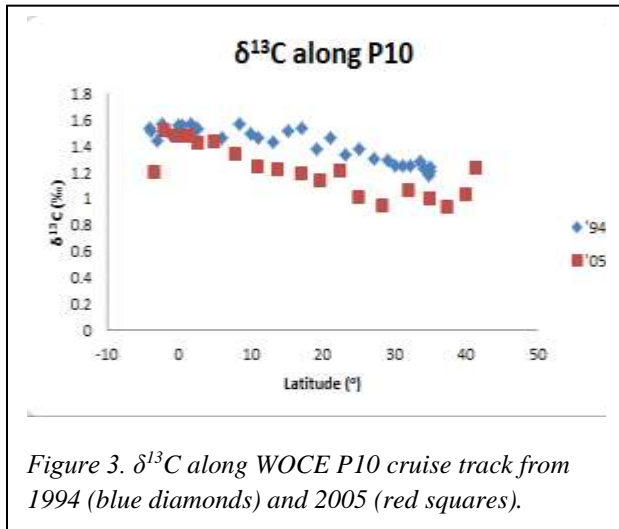
$$\delta^{13}\text{C} = \beta + m_1\theta + m_2S + m_3\text{AOU} + m_4\text{NO}_3 + R \quad (1)$$

from Sonnerup et al. (2000). β represents the intercept, θ is potential temperature, S is salinity, AOU is apparent oxygen utilization, and NO_3 is nitrate. R is the residuals, which represent how well the MLR model fits the data. m_1 , m_2 , m_3 , and m_4 are empirically derived regression coefficients for each respective variable. The MLR predicts $\delta^{13}\text{C}$ based off of these properties. An assumption is made that the relationship between $\delta^{13}\text{C}$ and our coefficients is constant throughout time. If hydrographic properties have a constant relationship to $\delta^{13}\text{C}$ then variations from what the MLR predicts must be from anthropogenic input and not changes in water properties. A MLR is constructed from the 1994 data set. Then the hydrographic properties from the 2005 data set are used in the 1994 MLR to predict $\delta^{13}\text{C}$. The differences between the predicted $\delta^{13}\text{C}$ using the '94 MLR and the measured $\delta^{13}\text{C}$ in 2005 is assumed to be due to anthropogenic carbon input. A MLR is also constructed from 2005 data and then that MLR is used with the 1994 hydrographic properties. Calculating the change in $\delta^{13}\text{C}$ going

forward as well as backwards through time is a valuable check to see if both methods are consistent.

To increase the accuracy of the MLR, an isopycnal MLR (isoMLR) approach is used (Quay et al., 2007). Instead of using one MLR for an entire dataset, a distinct MLR is created for each separate density bracket. This is effective because the MLR predicts $\delta^{13}\text{C}$ from hydrographic properties, and an assumption is made that water of similar density will also have more similar water properties. Quay et al. (2007) found that this method increased accuracy from $\pm 0.09\text{‰}$ to ± 0.04 . In this study 10 density brackets are used (Table 1).





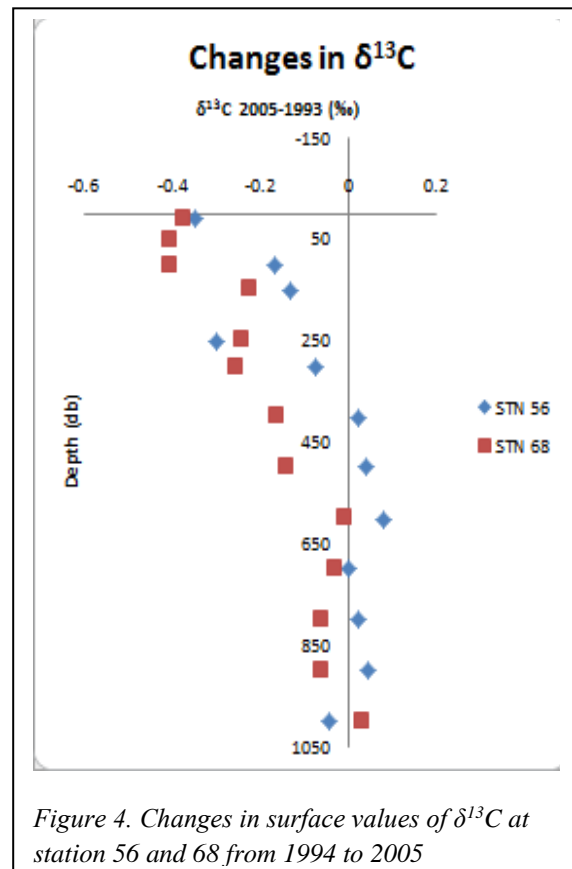
The isoMLRs have an average of 43 and 50 data points per density bracket in the 2005 and 1993 isoMLRs respectively. Deep water comparisons are made below 1500m to compare hydrographic properties which should remain constant through data sets. calculating $\delta^{13}\text{C}$ residuals of the 2005 MLR and 1994 hydrographic properties (Fig. 2). Below 1750m the residuals are centered at -0.034‰ with a standard deviation of 0.0063‰ . The residuals have been corrected by adding 0.034‰ so that the deep water residuals average at 0‰

From $\delta^{13}\text{C}$ to anthropogenic carbon using an RC value – From the MLR we are able to calculate a depth integrated value for $\delta^{13}\text{C}$ change. $\delta^{13}\text{C}$ is merely a tracer for anthropogenic carbon so a method is needed to convert from $\delta^{13}\text{C}$ to anthropogenic carbon. To do this an RC value of $-0.021 \pm 0.004\text{‰} (\mu\text{molC/kg})^{-1}$ from the Hawaii Ocean Time Series (HOTS) is used. The RC value is simply a ratio of anthropogenic $\delta^{13}\text{C}$ change to anthropogenic CO_2 change. Having calculated the change in anthropogenic $\delta^{13}\text{C}$ from the MLR, the depth integrated $\delta^{13}\text{C}$ residuals are simply divided by the RC value.

RESULTS

Surface values of $\delta^{13}\text{C}$ at stations have decreased between 1994 and 2005 (Fig. 3). The decrease in $\delta^{13}\text{C}$ is more pronounced at higher latitudes, ranging from -0.22‰ at 11.16°N to -0.37‰ at 25.16°N . Between station 56 and 68 we can see that at the higher latitude station has a larger decrease of $\delta^{13}\text{C}$ which averages to be -0.16‰ less (Fig. 4). Not only does the change in $\delta^{13}\text{C}$ increase, but the penetration of $\delta^{13}\text{C}$ change increases from 300db at station 56 to 500db at 68.

The residuals that were calculated for the 2005 dataset using the 2005 isoMLR method had an average of 0.00‰ with a standard deviation of 0.055‰ (Fig. 5). In the top 1000m there was a spread of $\pm 0.2\text{‰}$. Below 1000m the spread of residuals is $\pm 0.1\text{‰}$. The 1994 data set had residuals using the 1994 isoMLR also had an average of 0.00‰ with a standard deviation of 0.042‰ (Fig. 6).



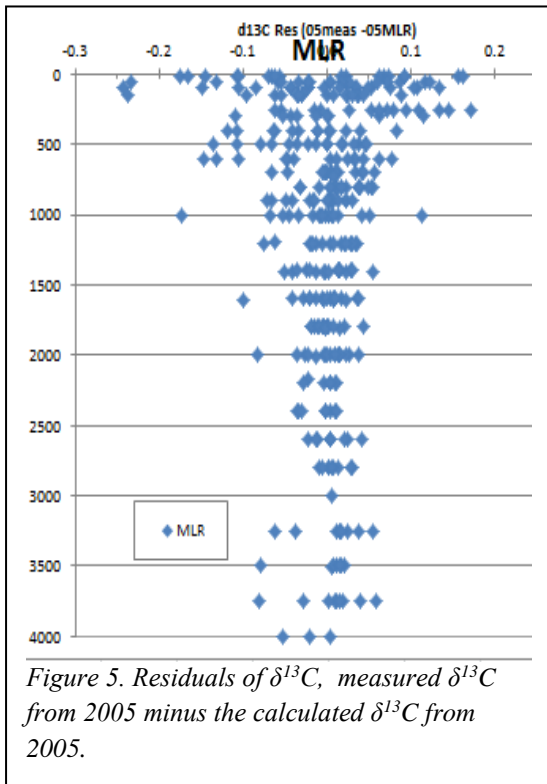


Figure 5. Residuals of $\delta^{13}\text{C}$, measured $\delta^{13}\text{C}$ from 2005 minus the calculated $\delta^{13}\text{C}$ from 2005.

Looking at residuals from the isoMLR (Fig. 5, Fig 6) there is a very similar spread to Quay et al. (2007). These plots of residuals display the accuracy of the isoMLR by predicting the $\delta^{13}\text{C}$ from 1994 and 2005 and subtracting the measured

$\delta^{13}\text{C}$ from 1994 and 2005 respectively. This results in the $\delta^{13}\text{C}$ residuals, which are the difference between the isoMLR predicted $\delta^{13}\text{C}$ and the known $\delta^{13}\text{C}$ values. If the isoMLR were to accurately predict the $\delta^{13}\text{C}$ values perfectly, then the residuals would all be 0. As figure 13 shows, the isoMLR is not perfect. However, the residuals are all centered at 0‰ which shows that the average predicted $\delta^{13}\text{C}$ is accurate.

When applied to the 2005 data set, the 1994 isoMLR predicted residuals to be -0.2‰ at surface (Fig. 7). They increased with depth and remain centered at 0‰ deeper than 800m. Using the 2005 isoMLR to predict residuals in 1994 the residuals were observed to be centered at 0‰ below 850m. Above 850m the residuals increase to about 0.2‰ (Fig. 2). The depth integrated residual is 132.5 ‰ m. Over the period between 1994 and 2005, this is a rate of 12.04 ‰ m yr⁻¹. The flux of anthropogenic carbon over this time is 0.20 μmolC (yr m²)⁻¹.

	Intercept	Pot Temp	Sal	AOU	NO3	PO4	SiO3	Degr Free	Error
<22.5	-0.18	0.11	-0.05	0.00	0.05	0.00	0.00	20	0.094
22.5-25	-6.01	0.03	0.18	-0.01	0.03	0.00	0.00	29	0.069
25-26.4	-30.40	-0.11	0.95	-0.01	0.00	0.00	0.00	45	0.108
26.4-26.8	-34.13	0.05	0.98	-0.01	0.10	0.00	0.00	28	0.070
26.8-27.2	-31.29	0.06	0.91	-0.01	0.07	0.00	0.00	51	0.055
27.2-27.4	-15.30	0.08	0.45	-0.01	0.05	0.00	0.00	37	0.035
27.4-27.6	-11.01	0.08	0.38	-0.01	-0.03	0.00	0.00	45	0.030
27.6-27.7	-11.09	0.13	0.37	-0.01	-0.02	0.00	0.00	42	0.02
27.7-27.75	191.92	0.14	-5.47	-0.01	0.01	0.00	0.00	44	0.04
27.75-27.8	299.47	0.01	-8.57	-0.01	0.00	0.00	0.00	55	0.02
								426	0.05
WOCE 1994									
	Intercept	Pot Temp	Sal	AOU	NO3	PO4	SiO3	Degr Free	Error
<22.5	2.96	0.02	-0.06	0.00	0.09	0.00	0.00	29.00	0.06
22.5-25	-0.72	0.04	0.03	0.00	0.01	0.00	0.00	70.00	0.06
25-26.4	-23.84	-0.03	0.73	0.00	0.00	0.00	0.00	104.00	0.06
26.4-26.8	-35.22	0.00	1.04	-0.01	0.04	0.00	0.00	42.00	0.05
26.8-27.2	-29.46	0.06	0.86	-0.01	0.07	0.00	0.00	92.00	0.05
27.2-27.4	-4.84	0.12	0.12	-0.01	0.09	0.00	0.00	64.00	0.05
27.4-27.6	5.06	0.08	-0.17	-0.01	0.10	0.00	0.00	52.00	0.05
27.6-27.7	120.19	-0.10	-3.43	-0.02	0.06	0.00	0.00	17.00	0.03
27.7-27.75	59.06	0.33	-1.60	0.00	-0.08	0.00	0.00	15.00	0.03
27.75-28	-304.85	0.00	8.82	0.00	-0.03	0.00	0.00	17.00	0.02

Table 1. Table of Intercept, MLR coefficients, degrees free, and error for 2005 isoMLR (top) and 1994 isoMLR(bottom) for each density bracket.

DISCUSSION

Surface trends in $\delta^{13}\text{C}$ along P10 (Fig. 3) shows that there has been a significant decrease across the entire transect from 1994 to 2005. This trend can also be seen in depth profiles of $\delta^{13}\text{C}$ at certain stations (Fig. 4). Because carbon dioxide from fossil fuels has a $\delta^{13}\text{C}$ of -28‰ (O'Leary, 1981) we would expect that an input of anthropogenic carbon dioxide to decrease the overall $\delta^{13}\text{C}$ value of seawater. Station 56 and 68 best represent this decrease in $\delta^{13}\text{C}$.

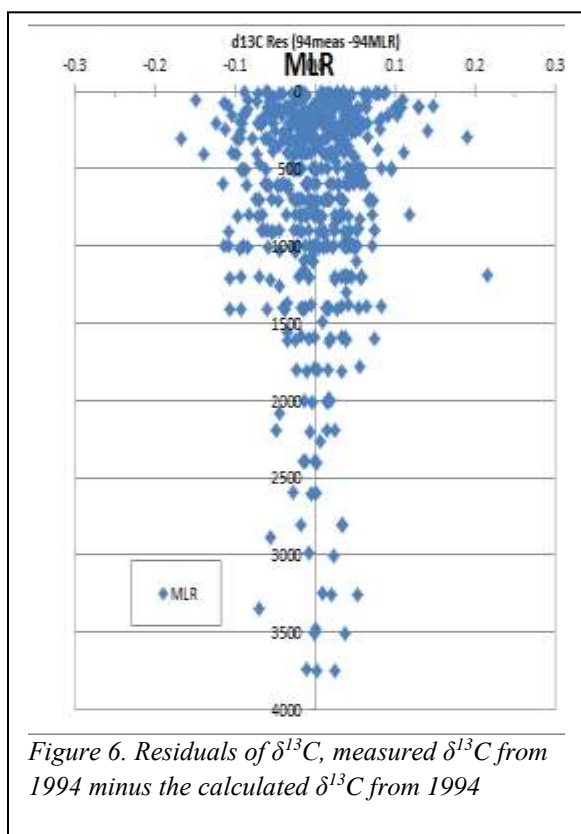


Figure 6. Residuals of $\delta^{13}\text{C}$, measured $\delta^{13}\text{C}$ from 1994 minus the calculated $\delta^{13}\text{C}$ from 1994

Stations 56 and 68 also display a latitudinal trend of both increasing $\delta^{13}\text{C}$ change and $\delta^{13}\text{C}$ penetration (Fig. 4, Fig. 8). At more northern latitudes the ocean is typically colder. Colder temperatures increase the capacity for carbon dioxide storage in water, which could partially explain the increase of $\delta^{13}\text{C}$ change. The Kuroshio and Kuroshio extension are a known significant

sink of carbon dioxide (Hideyuki et al., 2011). It is possible that as stations become closer to the Kuroshio extension and eventually cross through it, we see the strong sink of CO_2 signal in the depth profiles of $\delta^{13}\text{C}$. Depth profiles of $\delta^{13}\text{C}$ at P10 certain stations that were sampled in both 1993 and 2005 are not as clear (Fig. 8). Stations 34 and 47 have trends that do not follow the general decrease of $\delta^{13}\text{C}$ over time. This could be due to several factors. One reason is that the two data sets were sampled in different seasons, which could mask the trend in seasonal variability. These two stations are located at 5° and 11° N. Sabine et al. (2002) showed that in the tropics around these stations there is exceptionally low anthropogenic carbon inventory. With little anthropogenic carbon we would not expect to a large $\delta^{13}\text{C}$ decrease.

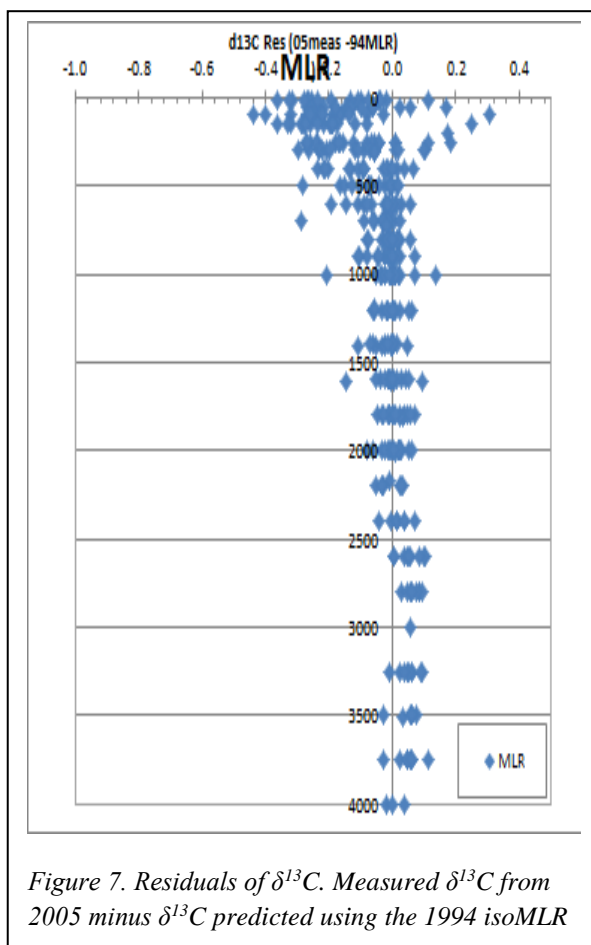


Figure 7. Residuals of $\delta^{13}\text{C}$. Measured $\delta^{13}\text{C}$ from 2005 minus $\delta^{13}\text{C}$ predicted using the 1994 isoMLR

When applying the isoMLR constructed from the 2005 dataset to 1994 hydrographic data, we see the residuals rise to a maximum at the surface of 0.27‰ (Fig. 2). Since we want to know the change going forward in time instead of backwards, we simply switch the sign to -0.27‰ to properly represent the changes. This shows that any given water parcel at the surface along P10 is expected to have a $\delta^{13}\text{C}$ value that has changed by

-0.27‰ from 1994 to 2005 due to anthropogenic input. Takahashi et al. (2000) found a change of -0.41‰ and -0.28‰ along the 1026.0 kg/m³ and 1026.4 kg/m³ isopycnals respectively along 175°E. This change was calculated using preformed $\delta^{13}\text{C}$. It is difficult to make a comparison because both the time scales and transect areas differ significantly.

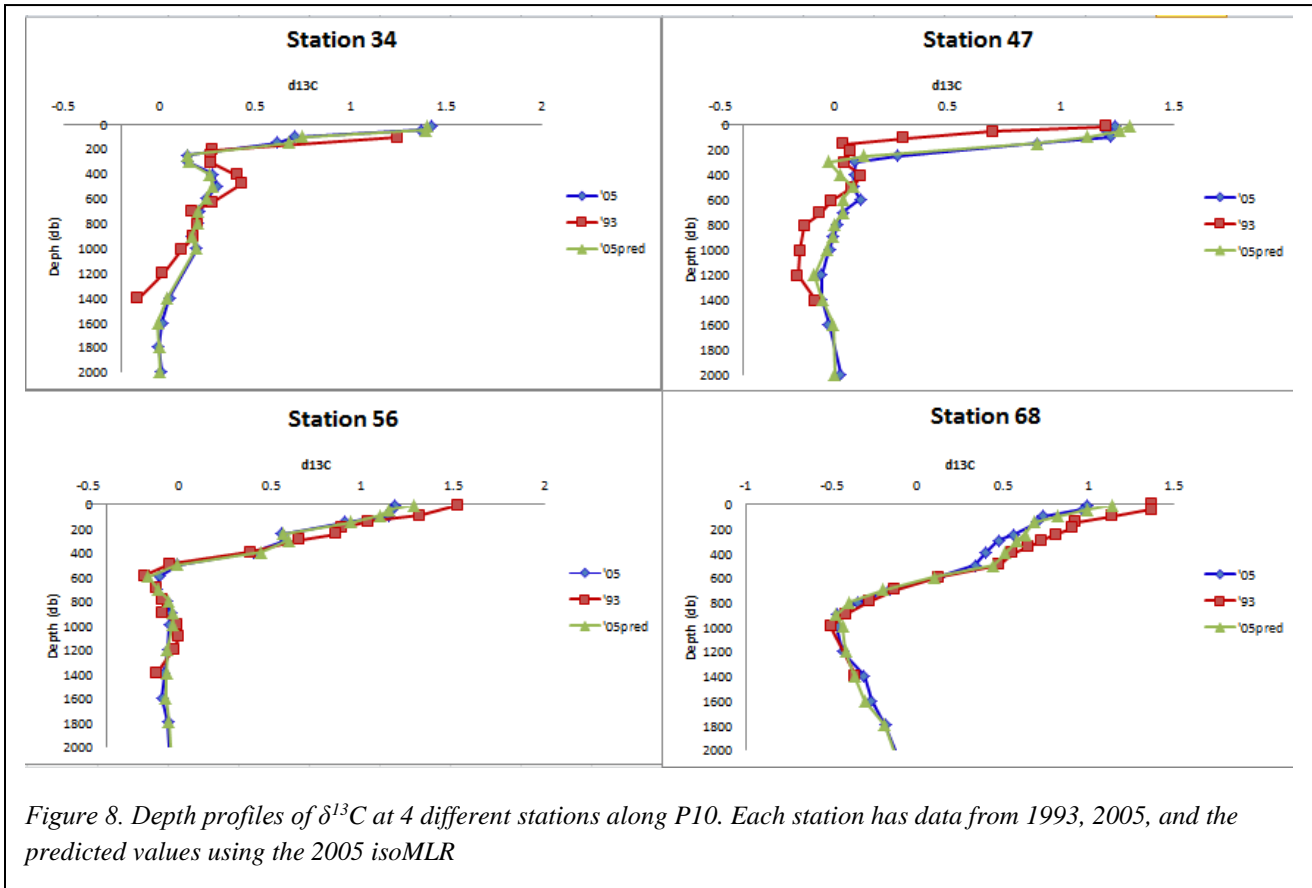


Figure 8. Depth profiles of $\delta^{13}\text{C}$ at 4 different stations along P10. Each station has data from 1993, 2005, and the predicted values using the 2005 isoMLR

In the Indian ocean the depth integrated $\delta^{13}\text{C}$ change between 1978 and 1995 was -6.9‰ m yr⁻¹ (Sonnerup et al., 2000). It is surprising that the change in $\delta^{13}\text{C}$ is so much smaller than the -12.04‰ m yr⁻¹ observed in the Pacific over a smaller time period. The Indian ocean is also in contact with the southern ocean, which is a known strong sink for anthropogenic carbon.

Using the same isoMLR method, Quay et al. (2007) had a similar trend which reached a

maximum of -0.45‰. The difference in magnitude of residuals is likely due to the different ocean basins. The Atlantic ocean has a much greater anthropogenic carbon inventory (Sabine et al., 2004). This is due to the formation of North Atlantic Deep Water which subducts water rich in anthropogenic carbon. Anthropogenic carbon can be found at depths of over 3000m in the North Atlantic (Sabine et al., 2004). Sabine et al. (2010) estimated that ~23% of the anthropogenic carbon

storage in the ocean is located in the North Atlantic Ocean. The North Pacific has no such deep water formation which prevents anthropogenic carbon from being stored at depth.

Quay et al. (2007) found that the depth integrated residuals of the OACES cruise in the Atlantic to be -15.0‰ m yr^{-1} . It is not a surprise that the $\delta^{13}\text{C}$ change is greater in the Atlantic because of the greater anthropogenic inventory. While the Pacific certainly has a lesser anthropogenic input, the change of $\delta^{13}\text{C}$ residuals show that the change is on the same scale as the Atlantic and only slightly less. It is also interesting to note that the $\delta^{13}\text{C}$ residuals in figure 2 were statistically significant down to 850m, showing that there is significant penetration of the anthropogenic $\delta^{13}\text{C}$ signal.

Using the depth integrated $\delta^{13}\text{C}$ residuals, the calculated flux of anthropogenic carbon into the ocean is $0.52 \text{ molC (m}^2 \text{ yr)}^{-1}$. Jarecki (2013) used the C^* method to estimate the anthropogenic carbon flux to be $0.94 \text{ molC (m}^2 \text{ yr)}^{-1}$. Jarecki (2013) focused on the Kuroshio extension region. In this study the whole P10 line was used, not just the Kuroshio extension. As mentioned before, the Kuroshio is an exceptionally large sink of carbon in the Pacific. It is expected that a latitudinal composite of anthropogenic carbon flux in the Pacific to be less than that of just the Kuroshio.

In the Atlantic Quay et al. (2007) estimated the flux of anthropogenic carbon to be $0.63 \text{ molC (m}^2 \text{ yr)}^{-1}$. Considering that the Atlantic is a much stronger anthropogenic carbon sink, the rate calculated in the Pacific is surprisingly similar to that of the Atlantic. This might lead us to believe that the Northwestern Pacific is a stronger sink for anthropogenic carbon than previously thought. Since the P10 transect is in the west Pacific, it cannot be assumed that the anthropogenic carbon flux of $0.52 \text{ molC (m}^2 \text{ yr)}^{-1}$ is representative of the entire Pacific, let alone the Northern Pacific.

CONCLUSIONS

Increased inventory of anthropogenic carbon dioxide into the Northwestern Pacific has been accompanied by a clear decrease of $\delta^{13}\text{C}$ in the surface layer of the WOCE P10 line from 1994 to 2005. This decrease can be observed in depth profiles down to 850m and the magnitude of decrease is 0.15‰ greater at 25.16°N than 11.16°N . Using isoMLR to separate anthropogenic forcing from natural variability, the change in surface $\delta^{13}\text{C}$ was calculated to be -0.2‰ . The depth integrated $\delta^{13}\text{C}$ change is -12.04‰ m yr^{-1} . Using an RC value, the depth integrated $\delta^{13}\text{C}$ change is converted to $0.52 \text{ mol C m}^{-2} \text{ yr}^{-1}$. The uptake rate in study area is significantly less than the $0.94 \text{ molC (m}^2 \text{ yr)}^{-1}$ Jarecki (2013) found using the C^* method in a similar region. $0.63 \text{ molC (m}^2 \text{ yr)}^{-1}$ was found in the Atlantic which is known to be a much greater anthropogenic carbon sink. This suggests that the Northwestern Pacific is a greater carbon sink than previously thought, especially the Kuroshio region that Jarecki (2013) focused on.

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