

## Decadal changes in Pacific carbon

Christopher L. Sabine,<sup>1</sup> Richard A. Feely,<sup>1</sup> Frank J. Millero,<sup>2</sup> Andrew G. Dickson,<sup>3</sup>  
Chris Langdon,<sup>2</sup> Sabine Mecking,<sup>4</sup> and Dana Greeley<sup>1</sup>

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[1] This paper uses the extended multiple linear regression (eMLR) technique to investigate changes over the last decade in dissolved inorganic carbon (DIC) inventories on a meridional line (P16 along 152°W) up the central Pacific and on a zonal line (P02 along 30°N) across the North Pacific. Maximum changes in the total DIC concentrations along P02 are 15–20  $\mu\text{mol kg}^{-1}$  over 10 years, somewhat higher than the  $\sim 1 \mu\text{mol kg}^{-1} \text{a}^{-1}$  increase in DIC expected based on the rate of atmospheric  $\text{CO}_2$  increase. The maximum changes of 15–20  $\mu\text{mol kg}^{-1}$  along the P16 line over the 14/15-year time frame fit with the expected magnitude of the anthropogenic signal, but there is a deeper than expected penetration of the signal in the North Pacific compared to the South Pacific. The effect of varying circulation on the total DIC change, based on decadal alterations of the apparent oxygen utilization rate, is estimated to be greater than 10  $\mu\text{mol kg}^{-1}$  in the North Pacific, accounting for as much as 80% of the total DIC change in that region. The average anthropogenic  $\text{CO}_2$  inventory increase along 30°N between 1994 and 2004 was 0.43  $\text{mol m}^{-2} \text{a}^{-1}$ , with much higher inventories in the western Pacific. Along P16, the average Northern Hemisphere increase was 0.25  $\text{mol m}^{-2} \text{a}^{-1}$  between 1991/1992 and 2006 compared to an average Southern Hemisphere anthropogenic  $\text{CO}_2$  inventory increase between 1991 and 2005 of 0.41  $\text{mol m}^{-2} \text{a}^{-1}$ .

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### 1. Introduction

[2] The ocean plays a major role in the global carbon cycle because it is a vast reservoir of carbon, its surface waters exchange carbon with the atmosphere with equilibration times of about 1 year, and most importantly it absorbs and stores approximately one-third of the carbon dioxide released to the atmosphere annually from human activity [Sabine and Feely, 2007]. The ocean sink for anthropogenic  $\text{CO}_2$  has been estimated using a number of indirect approaches [e.g., Quay *et al.*, 1992, 2003; Sarmiento and Sundquist, 1992; McNeil *et al.*, 2003]. A lack of accurate ocean carbon measurements has prevented a direct global assessment of the change in ocean carbon inventories over time.

[3] In the 1990s, researchers from several countries worked together through two international programs, the World Ocean Circulation Experiment (WOCE) and the Joint Global Ocean Flux Study (JGOFS), to conduct an extensive survey of the chemical and physical properties of the global

ocean [Feely *et al.*, 2001; Wallace, 2001]. The WOCE/JGOFS data provided the first global inorganic carbon data set with sufficient accuracy and coverage to make robust estimates of modern carbon distributions. Using a combined data set of approximately 73,000 carbon measurements, together with the  $\Delta\text{C}^*$  calculation technique, Sabine *et al.* [2004a] estimated that the ocean had accumulated approximately 118 Pg C between 1800 and 1994. The uncertainty of this inventory was estimated to be about 20 percent. These results have been compared to a range of model results [e.g., Orr, 2004] and to estimates using a variety of back calculation techniques [e.g., Waugh *et al.*, 2006; Tanhua *et al.*, 2007]. Although the details vary between the different estimates, the distributions are consistent within the uncertainty of the estimates.

[4] To estimate the accumulation of ocean carbon based on a one-time global survey,  $\Delta\text{C}^*$  employs a back calculation technique that takes the difference between the measured DIC and the preindustrial carbon distributions calculated from the measured alkalinity, assuming that it has not changed, and the preindustrial atmospheric  $\text{CO}_2$  concentration [Gruber *et al.*, 1996]. This type of calculation is based on the premise that the preindustrial ocean was in a steady state with respect to the essentially constant atmospheric  $\text{CO}_2$  concentration for several thousand years (i.e., much longer than the overturning timescales for the ocean). There are many assumptions and corrections that must be applied for this approach to work. These assumptions have been thoroughly discussed in the literature [e.g., Gruber *et*

<sup>1</sup>Pacific Marine Environmental Laboratory, NOAA, Seattle, Washington, USA.

<sup>2</sup>Rosenstiel School of Marine and Atmospheric Sciences, University of Miami, Miami, Florida, USA.

<sup>3</sup>Marine Physical Laboratory, Scripps Institution of Oceanography, University of California, La Jolla, California, USA.

<sup>4</sup>Applied Physics Laboratory, University of Washington, Seattle, Washington, USA.

al., 1996; Sabine and Feely, 2001; Matsumoto and Gruber, 2005; Sabine and Gruber, 2005]. These assumptions are not unique to the  $\Delta C^*$  approach. All of the techniques for estimating anthropogenic  $\text{CO}_2$  have a number of implicit and explicit assumptions that limit the accuracy of the estimates.

[5] While Sabine et al. [2004a] provided an estimate of the total accumulation of anthropogenic  $\text{CO}_2$  in the ocean, this estimate does not say anything about how the rate of ocean carbon storage has changed over time. In an attempt to eliminate the need for some of the necessary assumptions with the back calculation technique and directly document the changes in ocean carbon inventory, the international ocean carbon community has begun reoccupying some of the ocean sections sampled during the WOCE/JGOFS global survey. Within the U.S., the CLIVAR/ $\text{CO}_2$  Repeat Hydrography Program has identified 19 lines that it will reoccupy over a 10-year repeating cycle (see <http://ushydro.ucsd.edu/>). The program started in 2003 with cruises in the North Atlantic and has been running 2–3 cruises per year since that time. Significant carbon system changes have been observed in the Atlantic based on recent repeat hydrography cruises by the U.S. and other countries [Feely et al., 2005; Friis et al., 2005; Sabine et al., 2006; Tanhua et al., 2006, 2007]. This paper describes the carbon system changes observed on three recent U.S. cruises in the Pacific Ocean. One of the primary observations from both oceans is that interpretation of carbon system changes on decadal timescales is more complicated than previously thought. Here we address some new approaches for isolating the effects of apparent organic matter decomposition rates and circulation changes on ocean carbon distributions. Separating these effects allows an improved estimate of the carbon storage that can be attributed to uptake of  $\text{CO}_2$  from the atmosphere.

## 2. Data Quality

[6] The first consideration when trying to identify decadal carbon inventory changes is measurement quality. The anticipated annual anthropogenic increase in surface water DIC is approximately  $1 \mu\text{mol kg}^{-1} \text{a}^{-1}$ , with smaller changes in the older subsurface waters. The highest quality measurements must be made to accurately quantify the decadal inventory changes.

[7] This study examines changes along two sections in the Pacific, one zonal section along  $30^\circ\text{N}$  and one meridional section along  $\sim 152^\circ\text{W}$ . In 2004 the CLIVAR/ $\text{CO}_2$  Repeat Hydrography Program conducted a zonal cruise (called P02) from Japan to the United States along  $30^\circ\text{N}$ . This line was last occupied in 1994 by Japanese researchers [Ono et al., 1998]. In 2005 the southern half of the P16 line (P16S  $\sim 152^\circ\text{W}$ ) from Tahiti to  $71^\circ\text{S}$  was sampled by the CLIVAR/ $\text{CO}_2$  Program. The remainder of the P16 line, P16N from Tahiti to Alaska, was completed in 2006. The range of the P16 line covered by the two CLIVAR/ $\text{CO}_2$  cruises was last occupied in 1991/92 as four segments P16A, P16S, P16C, and P16N. All data are publicly available from the Carbon Dioxide Information Analysis Center (<http://cdiac.esd.ornl.gov/oceans/>).

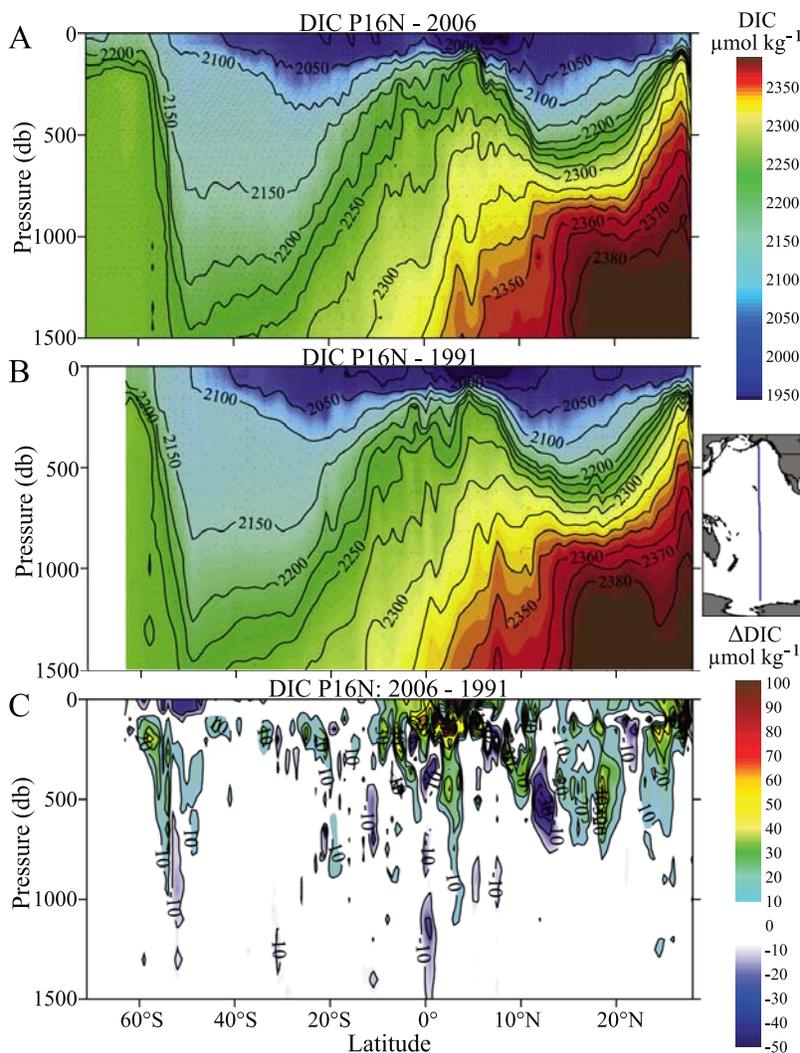
[8] Dissolved inorganic carbon (DIC) was analyzed on all cruises using a coulometric titration [Johnson et al., 1985,

1987; DOE, 1994; Ono et al., 1998]. Total alkalinity [Millero et al., 1993; DOE, 1994; Ono et al., 1998] was the second carbon parameter on all cruises except P16A and P16N which had  $\text{pCO}_2$  and pH, respectively, as their second parameter. Total alkalinity (TA) was calculated for these two cruises using the Mehrbach carbonate constants as refit by Dickson and Millero [1987]. The quality of the 1990s carbon data was evaluated by Lamb et al. [2002]. A set of adjustments for some cruises were recommended by the Global Ocean Data Analysis Project (GLODAP) based on many lines of evidence including comparison of calibration techniques, results from certified reference material analyses, precision of at-sea replicate analyses, agreement between shipboard analyses and replicate shore-based analyses, comparison of deep water values at locations where two or more cruises overlapped or crossed, consistency with other hydrographic parameters, and internal consistency with multiple carbon parameter measurements [Sabine et al., 2005]. They estimated that the overall accuracy of the DIC data after adjustments was  $\sim 3 \mu\text{mol kg}^{-1}$ . Total alkalinity, the second most common carbon parameter analyzed, had an overall accuracy of  $\sim 5 \mu\text{mol kg}^{-1}$  after adjustments.

[9] The CLIVAR/ $\text{CO}_2$  data quality was confirmed by daily analyses of Certified Reference Materials [Unesco, 1991; Dickson, 2001; Feely et al., 2001]. The consistency of the individual cruises was checked by comparing deep water ( $>2000 \text{ m}$ ) values at stations that overlapped on P16S and P16N and at the intersection of P16N and P02. These quality checks suggest that the DIC data are accurate to  $\sim 1 \mu\text{mol kg}^{-1}$  and the TA data are accurate to  $\sim 3 \mu\text{mol kg}^{-1}$ . Similar checks were made of the physical and nutrient data to confirm their consistency between the cruises. The CLIVAR/ $\text{CO}_2$  data were compared to the 1990s data along the two sections by examining values on isopycnal surfaces in deep water. The deep waters of the Pacific are among the oldest in the world and are not expected to change significantly over decadal timescales. The only observed offsets after the GLODAP corrections were found in the P02 TA data. The GLODAP assessment recommended an adjustment of  $+15 \mu\text{mol kg}^{-1}$  to the published 1994 P02 TA data [Lamb et al., 2002; Sabine et al., 2005]. Comparison with the CLIVAR/ $\text{CO}_2$  cruise suggested that this correction should have only been  $+10 \mu\text{mol kg}^{-1}$ . Although the difference in proposed corrections is within the stated uncertainty of the final GLODAP TA data set ( $\pm 5 \mu\text{mol kg}^{-1}$ ), the correction that minimized the deep water differences between these two occupations of P02 ( $+10 \mu\text{mol kg}^{-1}$ ) were used for this study.

## 3. Data Analysis

[10] A direct comparison of the DIC values can be accomplished by gridding both cruises to a regular interval and subtracting the two grids (Figure 1). Both the 1991/1992 and 2005/2006 sections show a very similar structure in the DIC distribution along P16. The difference plot, however, shows a patchy pattern of both positive and negative changes in the upper 1000 m. The observed pattern does not appear to be consistent with a regular uptake of  $\text{CO}_2$  from the atmosphere. This patchy distribution results from natural climate variability at interannual and shorter timescales. Aliasing from mesoscale eddies and frontal



**Figure 1.** Sections of DIC along 152°W measured in (A) 2005/2006, (B) 1991/1992, and (C) the difference between the two cruises. The black dots in Figures 1A and 1B show the measurement locations. Insert map shows the cruise track through the Pacific Ocean.

oscillations that displace isopycnal surfaces [Haine and Gray, 2001; Peacock *et al.*, 2005] as well as variations in water mass properties associated with climate modes such as the El Niño, Southern Oscillation [Feely *et al.*, 2002] can be significant when comparing two cruises separated in time.

[11] The multiple linear regression (MLR) approach was introduced by Wallace [1995] based on observations by Brewer *et al.* [1995] that carbon exhibited strong correlations with other oceanographic parameters over large areas of the ocean. First, the observed DIC on one cruise (usually the older cruise) is fit as a function of physical (e.g., potential density ( $\sigma_\theta$ ), potential temperature ( $\theta$ ), salinity (S)) and biological (e.g., silicate (Si), phosphate (P)) parameters that are not expected to be impacted by increasing atmospheric CO<sub>2</sub>. For example:

$$C_{\text{meas},t1} = F_1(\sigma_\theta, \theta, S, Si, P), \quad (1)$$

$$= a_1 \cdot \sigma_\theta + b_1 \cdot \theta + c_1 \cdot S + d_1 \cdot Si + e_1 \cdot P + k_1. \quad (2)$$

where,  $C_{\text{meas},t1}$  measured DIC and  $F_1$  is a function derived from the measured parameters on the first (older) cruise with the functional form shown in (2). The terms  $a_1$ ,  $b_1$ ,  $c_1$ ,  $d_1$ ,  $e_1$ , and  $k_1$  are coefficients of the multiple linear regression against measured DIC on the first cruise. The number and choice of the parameters varies depending on who is performing the calculation and the availability of high quality parameters. Using the coefficients from this fit together with the measured physical and biological parameters from the second cruise (indicated with primes), the DIC distribution is predicted for the second cruise ( $C_{\text{pred},t2}$ ):

$$C_{\text{pred},t2} = a_1 \cdot \sigma'_\theta + b_1 \cdot \theta' + c_1 \cdot S' + d_1 \cdot Si' + e_1 \cdot P' + k_1. \quad (3)$$

The difference between the DIC measured on the second cruise ( $C_{\text{meas},t2}$ ) and the predicted DIC is presumed to be the anthropogenic CO<sub>2</sub> increase between the two cruises:

$$\Delta C_{t2-t1} = C_{\text{meas},t2} - C_{\text{pred},t2}. \quad (4)$$

In theory the MLR approach should remove the majority of the DIC variability resulting from displaced isopycnal surfaces or fronts. Several studies have been published using this basic approach, e.g., *Sabine et al.* [1999, 2004b] and *Wanninkhof et al.* [2006].

[12] *Friis et al.* [2005] proposed a revision to the standard MLR approach that statistically reduces the measurement errors that the independent parameters introduce to the final calculation. In the so-called extended MLR (eMLR) multiple linear regression fits are determined for both cruises using the same set of independent parameters. Thus if the first cruise fit is represented by (1) and (2), then the second cruise is fit with a function of the same form:

$$C_{meas,t2} = F_2(\sigma'_\theta, \theta', S', S'_i, P'), \quad (5)$$

$$= a_2 \cdot \sigma'_\theta + b_2 \cdot \theta' + c_2 \cdot S' + d_2 \cdot S'_i + e_2 \cdot P' + k_2. \quad (6)$$

The coefficients of these two fits are subtracted and then applied to the measured parameter from the second cruise such that the resulting equation directly determines the net DIC change between the cruises:

$$\Delta C_{t2-t1} = (a_2 - a_1) \cdot \sigma'_\theta + (b_2 - b_1) \cdot \theta' + (c_2 - c_1) \cdot S' + (d_2 - d_1) \cdot S'_i + (e_2 - e_1) \cdot P' + k_2 - k_1 \quad (7)$$

With this approach, random variability in the independent parameters is minimized for both cruises and any propagated errors from an inability of a particular parameter to completely describe the dependent parameter is partially canceled out when the coefficients are subtracted [*Friis et al.*, 2005]. Qualitatively the eMLR does result in smaller variability in the deep water  $\Delta$ DIC estimates where no change is expected.

[13] Neither the MLR nor the eMLR approaches work in mixed layer waters that are strongly affected by seasonal physical and biological processes, and the relatively slow exchange rate between seawater and atmospheric  $\text{CO}_2$ , approximately 1 year. The MLR residuals show a pronounced increase in scatter with waters shallower than 150 m along the Pacific sections. Although some of the previous studies chose to include these shallow fits, we did not want to bias the water column results with shallow temporal trends. Instead, the near surface water DIC values were assumed to increase in proportion to the atmospheric  $\text{CO}_2$  increase during the time between the cruises. Measurements over the last several decades have shown that Pacific surface water  $\text{pCO}_2$  values have been increasing at approximately the same rate as the atmosphere [*Feely et al.*, 2006; *Takahashi et al.*, 2006]. Surface water alkalinity, on the other hand, does not appear to be increasing with time [*Keeling et al.*, 2004]. Thus increases in surface water DIC have been driven primarily by the rise in surface water  $\text{pCO}_2$  [*Keeling et al.*, 2004].

[14] The calculated increase in surface water DIC between the cruises for this study was determined by taking the DIC and alkalinity values measured in the top 50 m on the CLIVAR/ $\text{CO}_2$  cruises and calculating the  $\text{CO}_2$  fugacity ( $f\text{CO}_2$ ) using the  $\text{CO}_2\text{SYS}$  computer program [*Lewis and*

*Wallace*, 1998]. The change in global annual mean atmospheric  $\text{CO}_2$  between cruises (e.g., 26 ppm for the 2006 and 1991 P16 cruises) was then subtracted from the calculated  $f\text{CO}_2$  values. Since TA is not affected by gas exchange, the measured TA together with the revised  $f\text{CO}_2$  values was used to calculate the WOCE/JGOFS DIC distributions. The difference between the measured and calculated DIC values is the  $\Delta C_{t2-t1}$  estimate for the near surface waters. This approach is qualitatively consistent with a direct comparison of measured values on both cruises but removes a significant amount of the mesoscale variability in the measurements (Figure 1).

[15] *Friis et al.* [2005] note that the choice of which measurements to use for the independent parameters is somewhat subjective and has varied significantly in various published MLR estimates. Typically the choice of parameters is based on assessments of the statistical fits, data availability, and data quality. Almost all studies to date have used salinity (S) and potential temperature ( $\theta$ ) to characterize the conservative characteristics. Most of the differences have been in the use of nutrients, oxygen, or TA to characterize the biological characteristics. For this study all cruises were fit as a function of:  $\sigma_\theta$ , S,  $\theta$ , P, and Si. Oxygen or apparent oxygen utilization (AOU) were specifically not used for the fit in anticipation of using AOU to characterize the circulation changes as discussed below.

[16] Both the P16 and P02 lines were subdivided into three segments, and fit with three independent MLR functions. The divisions were chosen by first fitting all of the data along a section with a single equation and plotting the residuals as a function of latitude (P16) or longitude (P02). By examining the patterns of the residuals, one can see where the empirical relationships begin to break down. The data set is then divided into smaller subsets and fit again. If there is a significant improvement in the fits then the subsets are maintained. If the fits do not improve then the subsets are modified. We also look for subsets that make oceanographic sense (e.g., dividing the data at fronts). This process is repeated iteratively to achieve the best fits while maintaining the fewest number of subsets.

[17] The data from P16 were divided into the southern subtropics (south of  $15^\circ\text{S}$ ), the tropics ( $15^\circ\text{S}$ – $15^\circ\text{N}$ ), and the northern subtropics (north of  $15^\circ\text{N}$ ). The data from P02 were divided into the western boundary current region (west of  $145^\circ\text{E}$ ), the subtropical gyre region ( $145^\circ\text{E}$ – $140^\circ\text{W}$ ) and an eastern boundary region (east of  $140^\circ\text{W}$ ). A standard error for each of the fits is presented along with the coefficients in Table 1. The standard errors ranged from 2.2 to  $6.6 \mu\text{mol kg}^{-1}$ . The errors were generally larger for the older data sets, but also varied by geographical region. To obtain the eMLR coefficients one must subtract the WOCE/JGOFS coefficients from the Repeat Hydrography coefficients for each geographical region.

[18] Uncertainties in the eMLR approach come from three different sources. First, this approach assumes that the parameters can be adequately described with the empirical fits. This uncertainty can be assessed using the standard error of the fits. Since the standard errors were comparable to the measurement precision, this assumption is presumed to be valid. Second, uncertainties in all of the fit parameters can contribute to the overall uncertainty. Although the

**Table 1.** Coefficients for MLR DIC Fits

| Cruise     | Region      | Intercept | $\sigma_\theta$ | $\theta$ | S      | Si   | P     | Std. Error |
|------------|-------------|-----------|-----------------|----------|--------|------|-------|------------|
| P02 1994   | 135°E–145°E | 863.81    | 63.55           | 10.16    | −19.39 | 0.97 | 84.32 | 3.6        |
| P02 1994   | 145°E–140°W | 1527.32   | 43.19           | 6.57     | −21.71 | 1.06 | 76.98 | 5.8        |
| P02 1994   | 140°W–125°W | 884.32    | 7.35            | 0.99     | 26.25  | 0.82 | 85.22 | 5.8        |
| P02 2004   | 135°E–145°E | 2871.04   | 71.04           | 16.13    | −84.73 | 1.59 | 62.74 | 2.2        |
| P02 2004   | 145°E–140°W | 1585.99   | 76.63           | 14.94    | −51.30 | 1.19 | 76.60 | 2.5        |
| P02 2004   | 140°W–125°W | 956.48    | 5.74            | 0.70     | 26.10  | 0.70 | 81.52 | 2.5        |
| P16 1991/2 | 72°S–15°S   | −22.05    | 54.90           | 7.33     | 14.57  | 0.61 | 79.83 | 5.3        |
| P16 1991/2 | 15°S–15°N   | 522.58    | 73.08           | 14.55    | −17.27 | 1.05 | 78.48 | 6.6        |
| P16 1991/2 | 15°N–57°N   | 1023.41   | 44.15           | 9.92     | −8.73  | 1.07 | 84.85 | 3.8        |
| P16 2005/6 | 72°S–15°S   | −736.94   | 54.63           | 4.54     | 36.72  | 0.36 | 73.11 | 4.7        |
| P16 2005/6 | 15°S–15°N   | 954.02    | 65.22           | 14.88    | −23.70 | 1.07 | 81.03 | 4.4        |
| P16 2005/6 | 15°N–57°N   | 1417.27   | 19.38           | 5.47     | 0.51   | 0.90 | 86.90 | 4.1        |

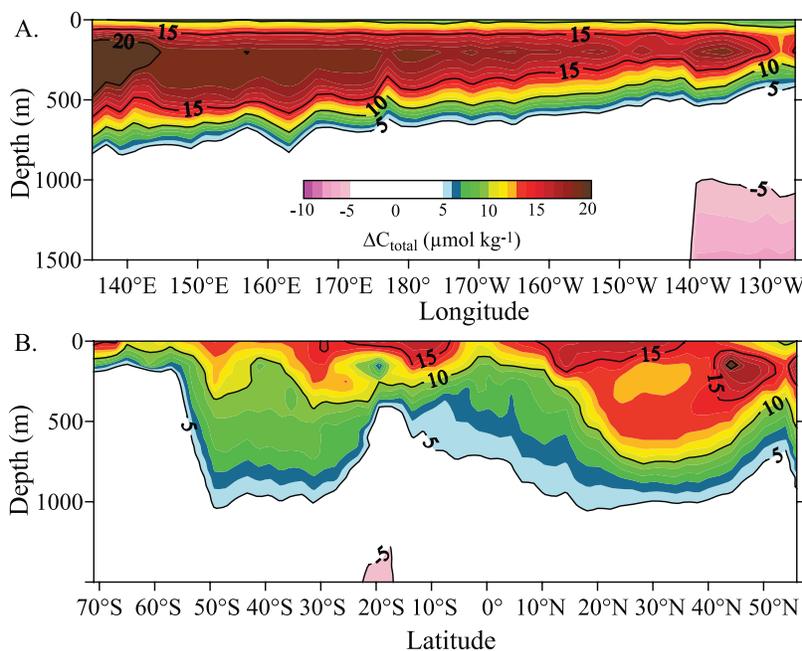
accuracies of the carbon measurements are generally well constrained with certified reference materials, some of the other parameters used in the fits, like the nutrients, can still have biases between cruises. These uncertainties can be addressed again with the standard error of the fits and with a careful evaluation of all the parameters in deep waters where decadal changes are assumed to be minimal. Finally, changes in the relative proportions of the fit parameters can introduce uncertainty. This is the basic premise that is exploited to determine the anthropogenic change in carbon, but if there were relative changes in the other parameters it would introduce uncertainty in the final result. Unfortunately, this uncertainty is very difficult to evaluate. However, we can again use the fact that the deep waters of the Pacific are not expected to have significant decadal changes to get a sense of this potential error. If the relative proportions of the fit parameters were to change, then an offset would be observed in the deep water values as well as the shallow values. Since the eMLR functions do not show any changes

in the deep waters, we have some confidence that the functions are behaving as expected.

## 4. Results and Interpretations

### 4.1. Observed DIC Changes

[19] Figure 2 shows the estimated total change in DIC for the P02 (10-year difference) and P16 (14/15-year difference) sections. The changes in both sections are restricted to the upper 1000 m. The P02 section shows larger and deeper changes in the western Pacific than in the east, consistent with a deepening of the isopycnal surfaces in the west. Maximum changes along P02 are 15–20  $\mu\text{mol kg}^{-1}$ , somewhat higher than the  $\sim 1 \mu\text{mol kg}^{-1} \text{a}^{-1}$  increase in DIC expected based on the rate of atmospheric  $\text{CO}_2$  increase. The maximum changes of 15–20  $\mu\text{mol kg}^{-1}$  along the P16 line fit with the anticipated magnitude of the anthropogenic signal, but the pattern of the observed changes is unusual. The distribution of chlorofluorocarbons



**Figure 2.** Sections of the total DIC change along (A) P02 (10-year difference) and (B) P16 (14/15 year difference) derived using eMLR.

**Table 2.** Coefficients for MLR AOU Fits

| Cruise     | Region      | Intercept | $\sigma_\theta$ | $\theta$ | S       | Si   | P      | Std. Error |
|------------|-------------|-----------|-----------------|----------|---------|------|--------|------------|
| P02 1994   | 135°E–145°E | −232.49   | −5.24           | 2.90     | 8.61    | 0.31 | 101.62 | 5.9        |
| P02 1994   | 145°E–140°W | 1334.39   | 54.09           | 23.49    | −89.97  | 1.15 | 110.59 | 9.4        |
| P02 1994   | 140°W–125°W | 40.09     | 31.97           | 17.05    | −33.13  | 0.53 | 125.49 | 8.0        |
| P02 2004   | 135°E–145°E | 1989.33   | 105.14          | 37.09    | −152.40 | 1.74 | 99.71  | 3.7        |
| P02 2004   | 145°E–140°W | 1390.56   | 102.08          | 37.19    | −133.11 | 1.38 | 122.96 | 4.0        |
| P02 2004   | 140°W–125°W | −10.45    | −25.56          | 6.03     | 15.94   | 0.30 | 122.76 | 4.0        |
| P16 1991/2 | 72°S–15°S   | −3076.69  | 126.27          | 27.845   | −18.92  | 0.62 | 121.11 | 7.4        |
| P16 1991/2 | 15°S–15°N   | −1094.01  | 151.90          | 36.50    | −97.89  | 0.70 | 115.88 | 6.4        |
| P16 1991/2 | 15°N–57°N   | 865.90    | 123.18          | 41.83    | −135.66 | 1.35 | 133.06 | 6.3        |
| P16 2005/6 | 72°S–15°S   | −4545.03  | 122.01          | 22.30    | 28.53   | 0.20 | 118.49 | 6.2        |
| P16 2005/6 | 15°S–15°N   | 640.09    | 104.97          | 33.00    | −111.33 | 0.74 | 128.31 | 6.0        |
| P16 2005/6 | 15°N–57°N   | 1239.3    | 94.37           | 36.08    | −122.49 | 1.16 | 137.42 | 6.1        |

(CFCs) along the P16 section suggests a shallower penetration of anthropogenic tracer in the North Pacific compared to the South Pacific (J. Bullister, personal communication, 2007). *Sabine et al.* [2002] also found a shallower penetration of anthropogenic CO<sub>2</sub> in the North Pacific, but the carbon distributions shown in Figure 2 show changes much deeper than expected for the North Pacific.

[20] The uptake of anthropogenic CO<sub>2</sub> is not the only process that can account for ocean carbon changes. While the eMLR technique accounts for physical displacements of water masses at interannual and shorter timescales, it does not fully correct for apparent changes in the recycling rate of organic and inorganic carbon. A number of recent papers have noted extensive changes in the apparent oxygen utilization (AOU) rates of North Pacific thermocline waters [*Emerson et al.*, 2001; *Ono et al.*, 2001; *Watanabe et al.*, 2001; *Andreev and Watanabe*, 2002]. Changes of a comparable magnitude were observed with this study. This AOU signal reflects either a change in the rate of oxygen consumption induced by changes in the export of organic matter from the near-surface waters, i.e., changes in the biological pump, or a change in the large-scale circulation which determines the time that subsurface waters have to accumulate an AOU signal at any given location. Whichever process is responsible for this signal, these AOU changes not only affect the dissolved oxygen budget of the North Pacific, but also the carbon distributions and must be accounted for in a proper assessment of Pacific carbon decadal changes.

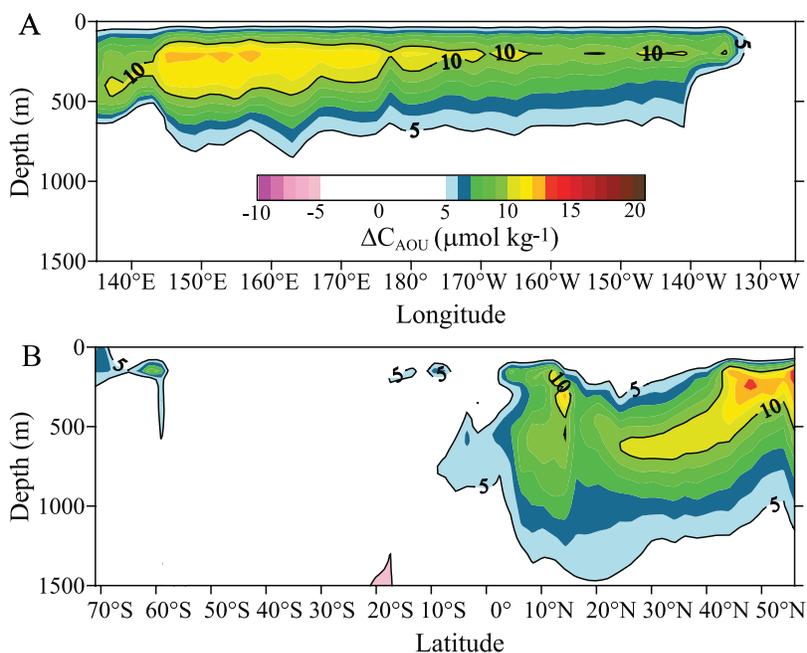
#### 4.2. Estimating Physical and Biological Changes

[21] Decadal variations in North Pacific dissolved oxygen, including changes along the P02 and P16N lines, have been examined by *Mecking et al.* [2008]. They find, as previous studies have shown, that the largest changes in AOU occur along the potential density ( $\sigma_\theta$ ) surface 26.6 kg m<sup>−3</sup>. This surface is the densest isopycnal to outcrop in the open North Pacific based on climatological data. *Mecking et al.* [2008] conclude that the AOU changes are consistent with a pattern of decadal-scale ventilation anomalies reflected in variations of the 26.6  $\sigma_\theta$  outcrop, including years when this surface did not outcrop. The idea of a physical control on AOU anomalies is further supported by *Deutsch et al.* [2006], who conducted a series of sensitivity runs using a coupled North Pacific general circulation model with a simple biogeochemical/ecological model. This model was

forced with “observed” winds and freshwater- and heat fluxes taken from an atmospheric reanalysis and indicated that changes in North Pacific circulation were sufficient to account for the observed pattern of large-scale decreases in O<sub>2</sub> in subpolar regions and simultaneous increases in the subtropics between the 1980s and 1990s. These findings are also consistent with circulation changes estimated from physical oceanographic observations [*McPhaden and Zhang*, 2002]. In the absence of consistent compelling evidence of large-scale changes in export production, we interpret the observed AOU changes to reflect North Pacific circulation changes during this time period.

[22] To estimate the potential impact these changes may have on the carbon distributions, AOU was fit using the same eMLR approach that was applied to the carbon data. The principal of the AOU eMLR is the same as for carbon. Using other tracer observations, the AOU of the water column can be fit with a multiple linear regression that characterizes the AOU patterns over large areas of the ocean [*Feely et al.*, 2004]. The parameters and regional subsets used for this study were the same as those determined for DIC. The coefficients and standard errors for the AOU fits given in Table 2 show that AOU can be fit to between 4 and 9  $\mu\text{mol kg}^{-1}$  of oxygen. Following through with equation (7), now used for AOU instead of DIC, gives the change in AOU between the cruises. As with the DIC eMLR, the AOU fits do not work in the mixed layer because of air-sea interactions. As with DIC, the eMLR was not used in the upper 150 m. The change in surface AOU between cruises was assumed to be zero and linearly interpolated between the surface and 150 m.

[23] After losing contact with the atmosphere, the primary mechanism for changing AOU is the decomposition of organic matter. The apparent rate of decomposition is a function of the biological production of particles that fall into the waters being examined and the length of time that the waters are being altered by the decomposing particles. If circulation slows, but the input of decomposing particle remains unchanged, then at any given location, AOU will be larger. Likewise, if circulation is unaltered, but the number of decomposing particles falling into the water column increases due to increases in export production, then the AOU will become larger. Either way, there will be a commensurate change in carbon inventory associated with the change in AOU because the decomposition of organic matter releases carbon as it consumes oxygen.



**Figure 3.** Sections of the carbon change estimated from AOU along (A) P02 (10-year difference) and (B) P16 (14/15-year difference) derived using eMLR.

[24] To convert the change in AOU between cruises as determined by the eMLR to carbon units, a carbon-to-oxygen stoichiometric ratio of 117/170 was used [Anderson and Sarmiento, 1994]. The changes in carbon distributions corresponding to the observed AOU changes over the last decade are shown in Figure 3. This estimate clearly shows a measurable signal in the North Pacific with very little change in the South Pacific. The tongue of carbon change associated with AOU seen in Figure 3b is centered on the  $26.6 \sigma_\theta$  surface, consistent with previous studies. Figure 3a shows that this signal extends essentially all the way across the North Pacific, but is stronger in the west than in the east. The details of the AOU patterns seen in Figure 3b are somewhat different from Mecking *et al.* [2008] because the eMLR approach adjusts for mesoscale variations of water properties (e.g., zonal changes in salinity). The magnitude of the core signal,  $>10 \mu\text{mol C kg}^{-1}$ , suggests that the processes affecting AOU can also explain a significant fraction of the total carbon change in the North Pacific.

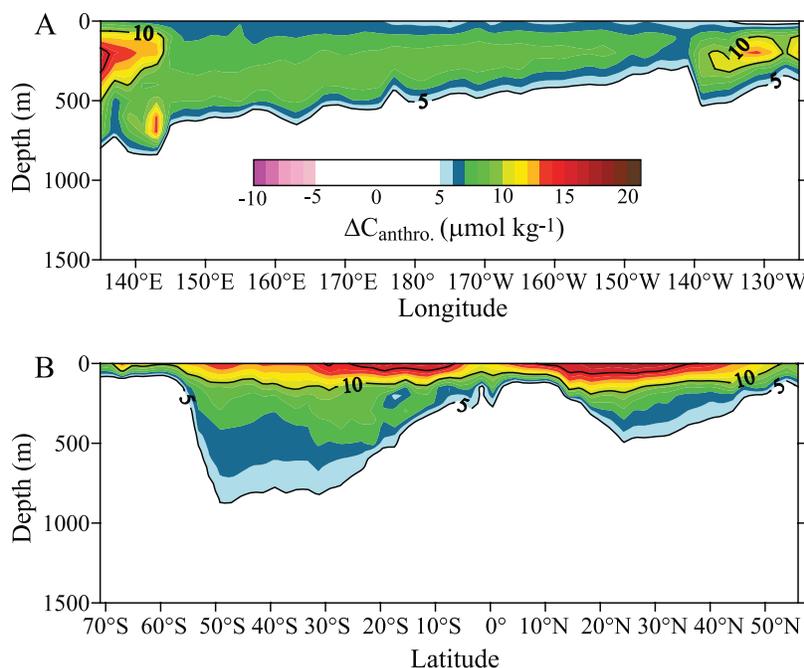
[25] Another possible mechanism for changing the DIC distributions is through a change in the production or remineralization of inorganic carbon in the water column. If significant changes in carbonate cycling in the North Pacific were present over the time frame of the cruises, a change in the total alkalinity distribution would be observed. A simple gridding and differencing of the alkalinity fields on the two lines (not shown) gives a complex pattern of positive and negative changes similar to the patterns observed in DIC. Some of these changes are large enough to have a measurable impact on the saturation state of the North Pacific waters as described by Feely *et al.* (R. A. Feely, *et al.*, Decadal changes in the oceanic carbonate system of the Pacific Ocean, submitted to *Global Biogeochemical Cycles*, 2008). Using the MLR approach to correct for the mesoscale variations, however, indicates that there is

no observable change in the alkalinity budget of the North Pacific that would impact the distributions of DIC.

#### 4.3. Anthropogenic DIC Changes

[26] To determine the change in inorganic carbon directly resulting from the uptake of  $\text{CO}_2$  from the atmosphere, the AOU-based DIC changes are subtracted from the total DIC change. The zonal North Pacific distribution of anthropogenic  $\text{CO}_2$  inventory changes over the last 10 years can be seen with the P02 section (Figure 4a). The P02 distributions show a very similar pattern as the total DIC changes, but the overall signal is much smaller and more consistent with anticipated changes for the 10-year time frame. There is a deeper penetration of anthropogenic  $\text{CO}_2$  in the western Pacific generally following the incline of the isopycnals, but the highest values appear to be concentrated in the far western Pacific with the Kuroshio Current. There is also a suggestion of higher anthropogenic  $\text{CO}_2$  associated with the California Current on the eastern edge of the basin. These patterns may reflect the importance of anthropogenic  $\text{CO}_2$  that is transported into the North Pacific Subtropical Gyre relative to local uptake.

[27] The P16 line shows the meridional pattern of anthropogenic  $\text{CO}_2$  accumulation over the last 14 years in the eastern central Pacific (Figure 4b). This section presents a very different pattern than the total DIC change section (Figure 2b). The North Pacific has a much smaller and shallower signal than the total change, while the South Pacific signal remained essentially the same. The anthropogenic  $\text{CO}_2$  P16 distribution is much more consistent with the CFC tracer distribution as well as the pattern of total anthropogenic  $\text{CO}_2$  inventory than the total change section [Sabine *et al.*, 2004a]. As in Sabine *et al.*, there is very little penetration of anthropogenic  $\text{CO}_2$  into the high-latitude Southern Ocean. The deepest penetration is associated with intermediate and mode waters of the southern hemisphere.



**Figure 4.** Sections of the change in anthropogenic CO<sub>2</sub> along (A) P02 (10-year difference) and (B) P16 (14/15-year difference) derived using eMLR.

Upwelling compresses the anthropogenic signal in the Equatorial Pacific and the North Pacific Subtropical waters have a lower inventory than the southern subtropical waters.

[28] Since the P02 and P16 lines cross, the resulting change in anthropogenic CO<sub>2</sub> profiles can be directly compared at the crossover point. Figure 5 shows the profiles of anthropogenic CO<sub>2</sub> at the crossover (152°W, 32°N) for the two cruises. The P16 values shown have been normalized to a 10-year time difference to be consistent with the time frame for the P02 difference. The error bars are based on propagating the standard errors from the fits of the two cruises as listed in Table 1. The differences in the two profiles are well within the uncertainties of the estimates.

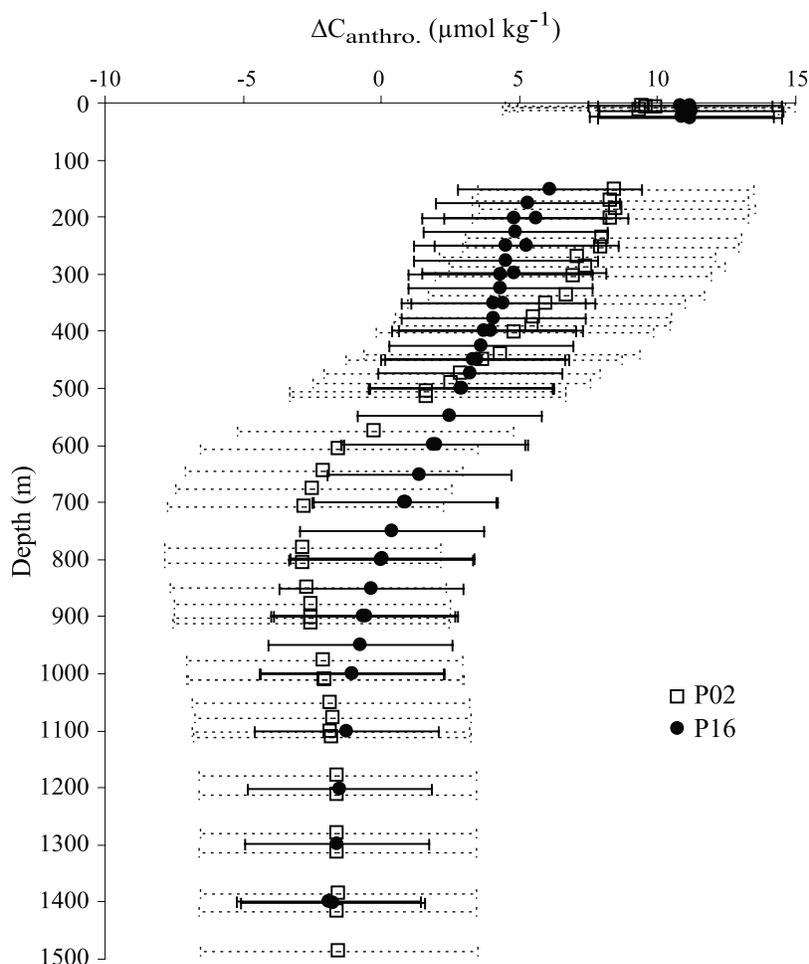
## 5. Discussion

[29] Since the P02 and P16 sections cover different time frames, it is difficult to compare the inventory sections. To allow a direct comparison of the results, the column inventory of anthropogenic CO<sub>2</sub> has been determined along each section and divided by the number of years between cruises (Figure 6). Values range from a maximum of approximately 0.9 to less than 0.1 mol C m<sup>-2</sup> a<sup>-1</sup>. If one takes an average anthropogenic CO<sub>2</sub> uptake of 2.2 PgC a<sup>-1</sup> [Denman *et al.*, 2007] and distributes it evenly over the global ocean area (335.2 × 10<sup>9</sup> km<sup>2</sup>), the expected column inventory change would be ~0.55 mol m<sup>-2</sup> a<sup>-1</sup>. Clearly the anthropogenic CO<sub>2</sub> is not distributed evenly and the uptake over the last two decades has not been a constant of 2.2 PgC a<sup>-1</sup>, but at this point we are unable to accurately resolve the anthropogenic CO<sub>2</sub> uptake at less than decadal intervals and this provides a reference for comparing the changes observed here to the average from the global carbon budgets. The deviations from this average trend illustrate the importance of assessing the inventory changes

in all of the ocean regions. Inventories in the southern subtropics are higher than the global mean value and inventories in the central and eastern North Pacific are lower. The average anthropogenic CO<sub>2</sub> inventory increase along 30°N between 1994 and 2004 was 0.43 mol m<sup>-2</sup> a<sup>-1</sup>, with much higher inventories in the western Pacific. Along P16, the average Northern Hemisphere increase was 0.25 mol m<sup>-2</sup> a<sup>-1</sup> between 1991/1992 and 2006 compared to an average Southern Hemisphere anthropogenic CO<sub>2</sub> inventory increase between 1991 and 2005 of 0.41 mol m<sup>-2</sup> a<sup>-1</sup>.

[30] Murata *et al.* [2007] recently examined the decadal (2003–1992) changes in CO<sub>2</sub> on a zonal section along 32°S (P06) using an isopycnal analysis approach. Despite the differences in approach, the estimated column inventory of anthropogenic CO<sub>2</sub> at the intersection of P16S and P06 were nearly identical (0.65 mol m<sup>-2</sup> a<sup>-1</sup>). In contrast to the zonal distribution observed in our study of the North Pacific, Murata *et al.* [2007] found that anthropogenic CO<sub>2</sub> concentrations were generally higher in the east than in the west. They attribute the observed South Pacific pattern to the more recent ventilation of intermediate and mode waters in the eastern South Pacific. Ventilation of mode and intermediate waters in the North Pacific, however, are predominantly in the Northwestern Pacific. This, together with the interactions between the North Pacific subpolar gyre and the subtropical gyre, could explain the different patterns of change between the North Pacific and the South Pacific.

[31] Although the anthropogenic CO<sub>2</sub> changes are relatively small in the eastern North Pacific, the total carbon change is quite large. Figure 7 shows the change in the column inventories of anthropogenic CO<sub>2</sub> (ΔC<sub>anthro</sub>), the carbon change determined from AOU changes (ΔC<sub>AOU</sub>), and the total carbon change (ΔC<sub>total</sub>) along the meridional P16 section. The anthropogenic CO<sub>2</sub> changes only account



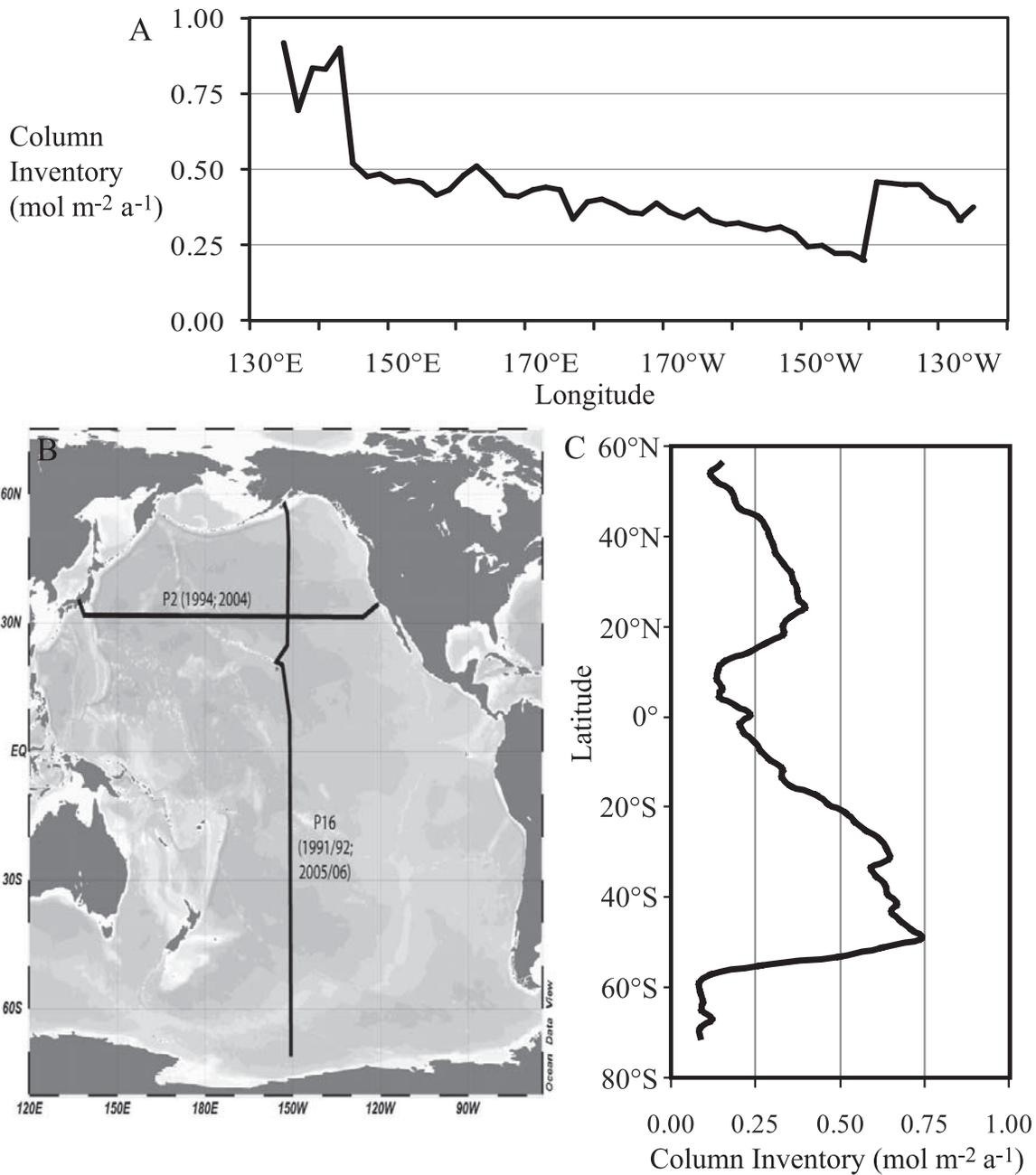
**Figure 5.** Profiles of the decadal change in anthropogenic  $\text{CO}_2$  at the crossover point between P02 and P16 ( $152^\circ\text{W}$ ,  $32^\circ\text{N}$ ). Note that the P16 cruises were 15 years apart so the values were normalized to 10 years to be consistent with the P02 time difference. The error bars are based on propagating the standard errors from the fits of the two cruises as listed in Table 1.

for  $\sim 17\%$  of the total inorganic carbon change north of the equator along this line. The inferred impact of a slower meridional circulation in the Pacific gives a mean  $\Delta C_{\text{AOU}}$  increase of about  $10 \text{ mol m}^{-2} \text{ a}^{-1}$  in the North Pacific Subtropical Gyre and about  $2 \text{ mol m}^{-2} \text{ a}^{-1}$  in the South Pacific Subtropical Gyre. Peaks in the circulation signal appear to be associated with the Subtropical Convergence Zone at about  $55\text{--}60^\circ\text{S}$  and the Intertropical Convergence Zone at  $2\text{--}10^\circ\text{N}$ . These peaks likely reflect differences in the location of the associated ocean fronts between the time the cruises were first run and when they were reoccupied. The Equatorial Pacific appears to be a transition zone from the southern anthropogenic  $\text{CO}_2$  dominated region to the northern circulation dominated region.

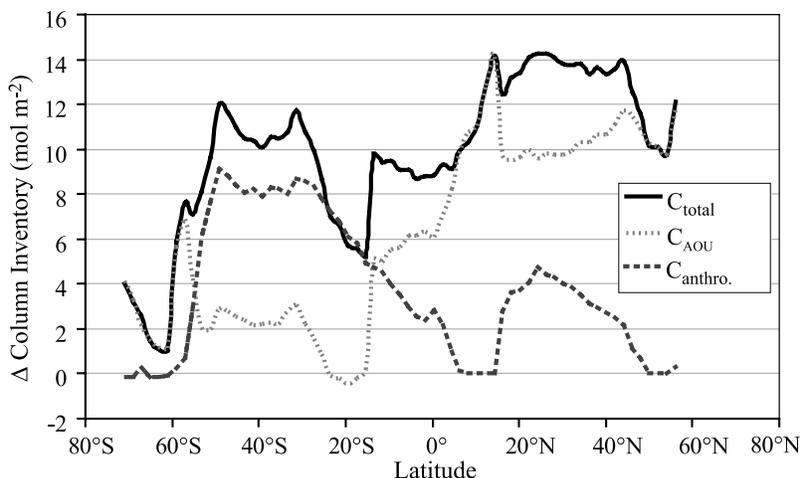
## 6. Conclusions

[32] The results presented here demonstrate that factors beyond anthropogenic  $\text{CO}_2$  uptake can have a measurable effect on ocean carbon distributions on decadal timescales. Many previous papers have either ignored the potential contribution of circulation changes or have assumed that

these changes would be implicitly corrected by the empirical MLR fits. Indeed, we found that an eMLR calculation along P16 which included AOU as a fit parameter (not shown), qualitatively looked similar to the final anthropogenic section shown in Figure 4b. However, the approach presented here provides additional information on the magnitude of the processes acting to change ocean carbon and a better understanding of the patterns of variability that are observed. While the anthropogenic signal is expected to continue to increase as atmospheric  $\text{CO}_2$  increases in the future, the circulation effects could very well reverse as the modes of climate variability change. Indeed, Mecking *et al.* [2008] suggest that the AOU anomalies may already be beginning to reverse in the North Pacific. An increase in ocean circulation could work to mask the anthropogenic signal rather than enhancing the total carbon change as observed in this study. The repeat hydrography sections will continue to provide valuable data for evaluating ocean carbon system changes in the different ocean basins over time. These data together with improving techniques for isolating the processes acting on the ocean carbon will allow



**Figure 6.** Estimates of annual anthropogenic CO<sub>2</sub> column inventory changes along (A) P02 and (C) P16. The map (B) shows the locations of the two lines being discussed.



**Figure 7.** Column inventories of the change in anthropogenic CO<sub>2</sub> (solid black line), circulation carbon changes estimated from AOU (light gray dotted line), and the total DIC change (dark gray dashed line) along the P16 section.

us to monitor the changing response of the oceans to global climate change.

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A. G. Dickson, Marine Physical Laboratory, Scripps Institution of Oceanography, University of California, 9500 Gilman Drive, La Jolla, CA 92093–0244, USA.

R. A. Feely, D. Greeley, and C. L. Sabine, Pacific Marine Environmental Laboratory, NOAA, 7600 Sand Point Way NE, Seattle, WA 98115, USA. (chris.sabine@noaa.gov)

C. Langdon and F. J. Millero, Rosenstiel School of Marine and Atmospheric Sciences, University of Miami, 4600 Rickenbacker Causeway, Miami, FL 33149–1098, USA.

S. Mecking, Applied Physics Laboratory, University of Washington, Seattle, WA 98105, USA.