

Anthropogenic CO₂ accumulation in the North Pacific Ocean from changes in ¹³C/¹²C of dissolved inorganic carbon

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Running Header: CO₂ uptake in the North Pacific from changes in ¹³C/¹²C
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Abstract

An anthropogenic CO₂ accumulation rate was estimated for the Northeastern Pacific Ocean using $\delta^{13}\text{C}$ of dissolved inorganic carbon ($\delta^{13}\text{C}$ –DIC) measured during WOCE 1991 and CLIVAR P16 2006 cruises. A significant $\delta^{13}\text{C}$ decrease was observed in the subtropical North Pacific surface layer between the WOCE 1991 and both CLIVAR P16 and STUD 08 cruises. In the subarctic North Pacific (40°N to 50°N) there was a significant $\delta^{13}\text{C}$ decrease in the surface waters between 1991 and 2006, however there was little $\delta^{13}\text{C}$ decrease between 1991 and 2008, likely due to seasonality. The $\delta^{13}\text{C}$ depth profiles, collected at 47°N 152°W and 28°N 152°W, revealed a $\delta^{13}\text{C}$ decrease between 1991 and 2008 to a maximum depth of 1000 m with a greater $\delta^{13}\text{C}$ change seen in the subtropics due to general circulation patterns. A multiple linear regression approach was applied to determine the anthropogenic $\delta^{13}\text{C}$ decrease from 1991 to 2006, from which a depth-integrated $\delta^{13}\text{C}$ change of -11.30‰ m yr^{-1} was calculated. The anthropogenic CO₂ uptake rate of $0.53 \pm 0.17 \text{ mol C m}^{-2} \text{ yr}^{-1}$ was determined by conversion of the depth-integrated $\delta^{13}\text{C}$ change to an integrated DIC change using the $\delta^{13}\text{C}/\text{DIC}$ anthropogenic change ratio, $-0.022 \pm 0.004\text{‰ } \mu\text{mol}^{-1} \text{ kg}$. Our rate compares well with other anthropogenic CO₂ uptake rates estimated for this region, as well as the mean global ocean CO₂ uptake rate of $0.5 \text{ mol C m}^{-2} \text{ yr}^{-1}$.

Oceans play a key role in the uptake of anthropogenic CO₂ added to the atmosphere. About 60 percent of the CO₂ generated by the burning of fossil fuels is accumulating in the atmosphere. Oceanic CO₂ uptake, estimated at ~ 2 Pg C yr⁻¹ (IPCC 2007), represents about 40 percent of the total fossil fuel CO₂ emitted to the atmosphere and establishes the ocean as a dominant sink for anthropogenic CO₂. Several different methods have been used to estimate anthropogenic CO₂ uptake in the oceans. One method used measured changes in the dissolved inorganic carbon (DIC) concentrations (Sabine et al. 2002; Peng et al. 2003). Takahashi et al. (2002) used the difference between the partial pressure of CO₂ (*p*CO₂) in the surface ocean and the air. Quay et al. (2003, 2007) used measured changes in the ¹³C/¹²C of dissolved inorganic carbon (δ¹³C-DIC). (Here δ¹³C (‰) = [(¹³C/¹²C)_{meas}/(¹³C/¹²C)_{std} - 1] x 1000 and the standard is PDB).

The present study used measured δ¹³C-DIC changes in the Northeastern Pacific over the last decade to determine the uptake rate of anthropogenic CO₂ in this region and to compare the rate to previous estimates.

Background

CO₂ uptake in the North Pacific Ocean – Measured changes in the DIC concentration between GEOSECS (1973) and WOCE (1990s) in the Northeast Pacific yielded an anthropogenic CO₂ uptake rate of 1.3 ± 0.5 mol C m⁻² yr⁻¹, with much of the uncertainty attributed to the older, lower quality GEOSECS CO₂ data (Peng et al. 2003).

Measured differences between the *p*CO₂ in the surface ocean and air have been used to determine the sea-air CO₂ flux and yield a CO₂ uptake rate of 0.6 mol C m⁻² yr⁻¹ for the Northern Pacific (0° – 50°N) (Takahashi et al. 2002). Takahashi et al. (2002) estimated that

the annual air-sea CO₂ flux into the Pacific north of 14°N is almost equal to the observed flux of CO₂ out of the equatorial Pacific and that the strongest region of CO₂ uptake in the North Pacific is at the boundary between the subtropical and subarctic gyres, along ~ 40°N.

Takahashi et al. (2002) hypothesized that in this region, where the warm tropical waters traveling poleward are cooled and nutrient-rich subpolar waters meet, the cooling effect on warm waters and biological drawdown combine to decrease the *p*CO₂ in the surface waters (increasing the oceanic CO₂ uptake). Biological mechanisms tend to dominate sea-air CO₂ flux in the subarctic regions of the North Pacific, while temperature effects dominate in the subtropics (Takahashi et al. 2002). Since biological processes, circulation and mixing in the surface ocean will potentially be affected by climate change the question is, will global warming cause the North Pacific to become a stronger or weaker sink for CO₂?

δ¹³C as a tracer of anthropogenic CO₂ – Measuring the change in δ¹³C is a useful approach in estimating the anthropogenic CO₂ uptake due to the similar time histories of atmospheric CO₂ and δ¹³C (Fig. 1). Comparison of two snapshots, one of recent δ¹³C measurements and another with values from a decade or two earlier, allows for the estimation of temporal δ¹³C change in the ocean (e.g. Quay et al. 2003). However, the temporal δ¹³C change is comprised of both natural variability and anthropogenic change. A multiple linear regression (MLR) approach has been used to separate natural and anthropogenic δ¹³C changes (e.g., Sonnerup et al. 2000; Quay et al. 2007).

In this study, the anthropogenic CO₂ uptake rate was determined from the δ¹³C change in this region of the Northeast Pacific Ocean over the last decade using δ¹³C

measurements on the STUD 2008 cruise compared to $\delta^{13}\text{C}$ measurements on the WOCE 1991 cruise along the same transect (152°W).

Methods

Sampling – Seawater samples were collected between 48°N 152°W and 18°N 152°W during the STUD 2008 cruise (Aug/Sept 2008) aboard the R/V Thompson (Fig. 2). Surface seawater samples were collected from a depth of 5 m, at 20 stations, every 1° of latitude from 48°N to 40°N and every 2° of latitude from 40°N to 18°N. Seawater samples for depth profiles were collected at 2 stations, 47°N and 28°N, to depths of 4500 m and 1800 m respectively. Seawater samples (~250 mL) were collected from Niskin bottles, poisoned with mercuric chloride (HgCl_2) and stored in sealed glass bottles following Quay et al. (1992).

$\delta^{13}\text{C}$ measurements – At the University of Washington Stable Isotope Lab, CO_2 gas for $^{13}\text{C}/^{12}\text{C}$ analysis was extracted from ~100 mL of seawater following the procedure described by Quay et al. (1992). Briefly, ~100 mL of seawater was acidified with 50% H_3PO_4 then, CO_2 gas was stripped by He flow and then trapped cryogenically using liquid nitrogen. The $^{13}\text{C}/^{12}\text{C}$ of the CO_2 was analyzed using a Finnigan MAT 251 mass spectrometer. The precision of the $\delta^{13}\text{C}$ -DIC measurement procedure, at $\pm 0.03\text{‰}$, has been determined by analyzing replicate seawater samples collected from the same Niskin bottle (Quay et al. 2003).

Multiple Linear Regression – The basis of the multiple linear regression (MLR) approach is establishing the relationship between spatial variations in the ocean of $\delta^{13}\text{C}$ and other seawater properties. The MLR expression relating these properties was described as follows by Sonnerup et al. (2000),

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

where β is the intercept, m_1 through m_4 are the regression coefficients for each variable, the residual (R) represents the difference between predicted and observed $\delta^{13}\text{C}$ for each data point, θ is potential temperature, S is salinity, AOU is apparent oxygen utilization (AOU = O_2 saturation - O_2 measured) and NO_3 is nitrate. Quay et al. (2007) modified the Sonnerup et al. (2000) approach by applying the MLR along isopycnal intervals (isoMLR), which allowed for a more accurate estimate of predicted $\delta^{13}\text{C}$ compared to that determined by a single MLR applied to the entire density range. The MLR approach is used to account for $\delta^{13}\text{C}$ changes that resulted from natural variations in the seawater properties (e.g., T, S, AOU, NO_3) over the time interval between the two cruises. The assumption is made that the same relationship between $\delta^{13}\text{C}$ and the seawater properties holds over time, thus any deviations between measured $\delta^{13}\text{C}$ and MLR predicted $\delta^{13}\text{C}$ is assumed to result from anthropogenic CO_2 uptake.

For the present study, the 2006 CLIVAR P16 cruise from 51°N 152°W to 24°N 152°W was used to establish the correlation coefficients between $\delta^{13}\text{C}$ and other seawater properties. The isoMLR approach was used to determine the MLR coefficients for data sorted into 18 isopycnal intervals (range $23.8 < \sigma < 27.7$). The magnitudes of the residuals in the MLR, when applied to CLIVAR 2006, yield the goodness of fit. The difference between measured $\delta^{13}\text{C}$ from a previous cruise (WOCE 1991) and the MLR predicted $\delta^{13}\text{C}$ value for

this cruise, using the seawater properties (e.g., T, S, AOU, NO₃) measured during the cruise is assumed to be the anthropogenic $\delta^{13}\text{C}$ decrease over the time elapsed between the two cruises.

Results

Interlaboratory $\delta^{13}\text{C}$ offset – A comparison of $\delta^{13}\text{C}$ depth profiles collected in the North Pacific during CLIVAR P16 (2006) and at the same stations during STUD 08 (Fig. 3.), at depths >1000 m (n=30), revealed a $\delta^{13}\text{C}$ decrease between 2006 and 2008 confirming a laboratory offset of 0.20‰. The seawater samples collected during CLIVAR P16 were analyzed at Woods Hole Oceanographic Institute (WHOI) and those collected during STUD 08 were analyzed at the University of Washington Stable Isotope Lab (UW). A similar interlaboratory $\delta^{13}\text{C}$ offset has been observed between WHOI and both Japan Marine Science and Technology Center (JAMSTEC) and UW labs during other cruises (P. Quay, pers. comm.). A correction for the lab offset was applied to the $\delta^{13}\text{C}$ data collected during the CLIVAR P16 (2006) cruise, after which no apparent $\delta^{13}\text{C}$ decrease from 2006 to 2008 was observed in the $\delta^{13}\text{C}$ depth profiles (Fig. 4.).

Samples collected during the WOCE 1991 cruise were analyzed by WHOI however, it is unlikely the laboratory offset observed in the $\delta^{13}\text{C}$ measurements made in 2006 was of concern for the WOCE 1991 $\delta^{13}\text{C}$ measurements. Quay et al. (2002) compared $\delta^{13}\text{C}$ measurements made at both WHOI and UW laboratories from deep water samples (>1200 m) collected during repeat WOCE cruises in the Indian Ocean and found a mean interlaboratory offset of 0.02‰, which is within the measurements uncertainty. The possibility of an offset will be revisited as additional STUD 08 $\delta^{13}\text{C}$ measurements become available.

$\delta^{13}\text{C}$ changes – A measured $\delta^{13}\text{C}$ decrease was observed in the depth profiles collected during WOCE 1991 and STUD 2008 cruises (Fig. 5a.). A greater $\delta^{13}\text{C}$ decrease was observed in the subtropics (28°N 152°W) than in the subarctic (47°N 152°W). Similarly, $\delta^{13}\text{C}$ depth profiles collected during WOCE 1991 and CLIVAR P16 (2006) reveal a greater $\delta^{13}\text{C}$ decrease in the subtropical region than in the subpolar region (Fig. 5b.).

Multiple linear regression – When a single multiple linear regression (MLR) is applied to the entire CLIVAR P16 (2006) $\delta^{13}\text{C}$ data set (n=300), the mean residual is $\pm 0.09\text{‰}$ (Fig. 6.). Application of the isopycnal-based MLR (isoMLR) to the $\delta^{13}\text{C}$ data collected during the CLIVAR P16 (2006) cruise resulted in a lower mean residual of $\pm 0.04\text{‰}$ (Fig. 6.). A similar improvement in MLR fit, using the isopycnal MLR rather than the single MLR, was observed by Quay et al. (2007).

An MLR was applied to determine the anthropogenic $\delta^{13}\text{C}$ decrease between 1991 and 2006. The assumption that the relationship between $\delta^{13}\text{C}$ and seawater properties (T, S, AOU, NO_3) established by the MLR approach holds over time, assigns the difference between WOCE 1991 measured $\delta^{13}\text{C}$ and 1991 predicted $\delta^{13}\text{C}$ (for 2006) to anthropogenic change (Fig. 5b.). As found by Quay et al. (2007), the anthropogenic $\delta^{13}\text{C}$ change is greater than the change due to natural variation suggesting the anthropogenic signal dominates over natural variability.

Surface ocean $\delta^{13}\text{C}$ changes – The surface ocean $\delta^{13}\text{C}$ trend in the North Pacific Ocean between 1991 and 2008 (Fig. 7.) revealed a $\delta^{13}\text{C}$ decrease of a much greater magnitude in the

subtropics (~28°N to 37°N) than in the subarctic region (~38°N to 50°N). A similar surface layer decrease in $\delta^{13}\text{C}$ in the subtropics was observed between 1991 and 2006. However, in the higher northern latitudes (40°N to 50°N), there were noticeable decreases in the measured surface $\delta^{13}\text{C}$ between the CLIVAR P16 (2006) cruise in March and the STUD 08 cruise conducted late August through early September.

Discussion

Surface $\delta^{13}\text{C}$ trends in the North Pacific Ocean – The significant surface $\delta^{13}\text{C}$ decrease in the subtropical North Pacific surface layer between cruises in 1991 and both 2006 and 2008 (Fig. 7.) was expected based on previous $\delta^{13}\text{C}$ comparisons (Quay et al. 2003). In subtropical regions where surface waters are renewed slowly, the surface water $\delta^{13}\text{C}$ has a longer time to equilibrate with the $\delta^{13}\text{C}$ of atmospheric CO_2 as opposed to subarctic regions where surface water is renewed rapidly, not allowing enough time for the surface waters to isotopically exchange with the atmosphere (Quay et al. 2003). In the subarctic North Pacific (40°N to 50°N) there was little $\delta^{13}\text{C}$ decrease between 1991 and 2008, however, there was a significant decrease between 1991 and 2006. Seasonality likely contributes to the $\delta^{13}\text{C}$ change observed in the surface ocean in this region. The WOCE 1991 and CLIVAR P16 (2006) cruises were both conducted in March while STUD 08 was conducted late August through early September. In the early spring there is less biological production in subarctic regions due to light limitations, resulting in lower $\delta^{13}\text{C}$ in the surface ocean than later in the summer when biological production is high and $\delta^{13}\text{C}$ is enriched in the surface ocean. In the subtropical regions, in contrast, where there is less variability in biological production

throughout the year, there is little seasonal $\delta^{13}\text{C}$ change ($\sim 0.15\text{‰}$ at ALOHA by Quay et al. 2003) and thus the $\delta^{13}\text{C}$ observed in the subtropical surface layer is similar in 2006 and 2008.

$\delta^{13}\text{C}$ depth trends – A comparison of $\delta^{13}\text{C}$ depth profiles collected at two stations, one in the subarctic North Pacific (47°N 152°W) and one in the subtropical North Pacific (28°N 152°W), during WOCE 1991 and STUD 08 cruises confirmed a $\delta^{13}\text{C}$ decrease to a maximum depth of 1000 m (Fig. 5a.). Below 1000 m a $\delta^{13}\text{C}$ change was not expected due to the age of the water and the lack of exposure of these older water masses to anthropogenically produced CO_2 .

The $\delta^{13}\text{C}$ depth profiles were expected to reveal a greater $\delta^{13}\text{C}$ decrease from 1991 to 2008 in the subtropics than in the subarctic due to general circulation patterns. The subtropics are located in a zone of convergent surface Ekman flow, where residence times of surface waters are longer and isopycnal surfaces are deeper than in zones of divergent surface Ekman flow. This results in greater accumulation of anthropogenic tracers in zones of convergence as opposed to zones of divergence in subarctic regions (Quay et al. 2002).

Depth trend in $\delta^{13}\text{C}$ decrease – The WOCE 1991 $\delta^{13}\text{C}$ residuals (MLR predicted $\delta^{13}\text{C}$ for 2006 minus 1991 measured $\delta^{13}\text{C}$) decreased from a maximum of -0.58‰ at ~ 100 m, to being indistinguishable from 0‰ at 600 m (Fig. 8.). Similarly, in the Indian Ocean Sonnerup et al. (2000) found the $\delta^{13}\text{C}$ residuals decreased between 1978 and 1995 from a maximum in the surface ocean to 0‰ at 1000 m. Quay et al. (2007) found $\delta^{13}\text{C}$ residuals decreased from a maximum in the surface ocean of the North Atlantic, from 1993 to 2003, to being

indistinguishable from 0‰ at ~1200 m. The residual $\delta^{13}\text{C}$ decrease observed at deeper depths in the North Atlantic is likely due to NADW formation.

Anthropogenic CO₂ uptake rate – The depth-integrated $\delta^{13}\text{C}$ change, -11.30‰ m yr^{-1} , was calculated by depth integrating the difference between isoMLR predicted and measured $\delta^{13}\text{C}$ in 1991 from 100 m to 600 m (Fig. 8.). The depth-integrated $\delta^{13}\text{C}$ change was restricted between 100 and 600 m to exclude both the surface layer and below 600 m, where the residual lies within the uncertainty and cannot be distinguished from zero. The anthropogenic CO₂ uptake rate is then estimated by conversion of the depth-integrated $\delta^{13}\text{C}$ change to an integrated DIC change by use of the $\delta^{13}\text{C}/\text{DIC}$ anthropogenic change ratio. The $\delta^{13}\text{C}/\text{DIC}$ anthropogenic change ratio was determined by combining the $\delta^{13}\text{C}$ changes determined by Sonnerup et al. (2000) with the DIC changes determined by Peng et al. (2003) along the 26.4, 26.6 and 26.8 isopycnal surfaces. The mean $\delta^{13}\text{C}/\text{DIC}$ ratio, -0.022 ± 0.004 ‰ μmol^{-1} kg, used to convert from $\delta^{13}\text{C}$ change to DIC change compared well with the -0.024 ‰ μmol^{-1} kg determined by Körtzinger et al. (2003) for the North Atlantic. An anthropogenic CO₂ uptake of 0.53 ± 0.17 mol C m⁻² yr⁻¹ was estimated for the North Pacific Ocean.

Error analysis – Statistical analysis, using propagation of errors, takes into account the uncertainty in the MLR approach (0.07‰) and in the $\delta^{13}\text{C}/\text{DIC}$ ratio (± 0.004 ‰ μmol^{-1} kg), which allowed for the determination of the ± 0.17 mol C m⁻² yr⁻¹ uncertainty in the estimated anthropogenic CO₂ accumulation rate.

Previous anthropogenic CO₂ uptake rate estimates for the North Pacific – Several different techniques and tracers used to estimate anthropogenic CO₂ uptake rates with a range of magnitudes that are not in agreement. Sabine et al. (2004) summarized published CO₂ uptake rates ranging from 0.33 ± 0.05 to 1.3 ± 0.5 mol C m⁻² yr⁻¹ for the North Pacific Ocean (Table 1.). CO₂ uptake rates determined by CFC tracers are on the low end of these estimates, while MLR-based estimates using DIC as a tracer are the largest CO₂ uptake rates (Sabine et al. 2004). The anthropogenic CO₂ uptake rate of 0.53 ± 0.17 mol C m⁻² yr⁻¹ estimated for the North Pacific by this study falls within the range of previous estimates. It appears that different tracers and techniques used to measure carbon change in the ocean likely lead to the observed discrepancies in the estimates of CO₂ uptake rates, this remains to be resolved.

An MLR-approach was applied to DIC concentrations measured during the STUD 08 cruise and an anthropogenic CO₂ uptake rate of 0.67 mol C m⁻² yr⁻¹ was estimated for the North Pacific (K. Salemmé, pers. comm.). The CO₂ uptake rate determined using DIC as the tracer was comparable to both the rate determined using $\delta^{13}\text{C}$ as a tracer, and with other MLR-based CO₂ uptake rates determined using DIC concentrations (Sabine et al. 2004). In comparison, the global ocean CO₂ uptake rate over the last 20 years is ~ 0.5 mol C m⁻² yr⁻¹ (IPCC 2007).

Comparison to the North Atlantic Ocean – Using the same isoMLR $\delta^{13}\text{C}$ approach in the North Atlantic, Quay et al. (2007) determined an anthropogenic CO₂ uptake rate of 0.63 ± 0.16 mol C m⁻² yr⁻¹ in good agreement with the rate estimated by this study for the North Pacific. Quay et al. (2007) found in the North Atlantic Ocean about half of the

anthropogenic CO₂ inventory change was due to air-sea CO₂ exchange with the other half resulting from meridional circulation. Meridional circulation is absent in the North Pacific and the anthropogenic CO₂ accumulation rate should be largely due to air-sea CO₂ exchange (Quay et al. 2007). This further suggests the anthropogenic CO₂ uptake rate, 0.53 ± 0.17 mol C m⁻² yr⁻¹, determined here for the North Pacific is reasonable. If the CO₂ uptake rate for the North Pacific was ~ 1 mol C m⁻² yr⁻¹ this would imply an air-sea CO₂ flux in the North Pacific that is $\sim 4x$ greater than in the North Atlantic (Quay et al. 2007).

North Pacific Ocean as a sink for anthropogenic CO₂ – Recent studies have found on a global scale the ocean has weakened as an anthropogenic CO₂ sink, since the year 2000, due primarily to human-induced climate change. The Southern Ocean is responsible for approximately half of the decrease in anthropogenic CO₂ uptake however, it is still unknown what role the North Pacific region plays (Canadell et al. 2007). Continued measurements using anthropogenic tracers are necessary over long time periods to provide sufficient data to determine whether the North Pacific will become a stronger or weaker sink for anthropogenic CO₂ in response to climate change.

Future research – The difference in magnitudes of anthropogenic CO₂ uptake rates estimated for the North Pacific by different tracers may be attributed to characteristics of the particular tracer technique. Since each tracer approach has substantial uncertainty, a convergence of rates by multiple approaches would improve our confidence in the rate estimate. Future research would be necessary to resolve these observed differences in CO₂ uptake rates and define the North Pacific Oceans role in the global carbon cycle.

Conclusions

A decrease in $\delta^{13}\text{C}$ was observed over the last 15 years along 152°W in the eastern North Pacific. An anthropogenic CO_2 accumulation rate of $0.53 \pm 0.17 \text{ mol C m}^{-2} \text{ yr}^{-1}$ was estimated for the North Pacific by conversion of the depth-integrated $\delta^{13}\text{C}$ change, -11.30‰ m yr^{-1} , to an integrated DIC change by use of the $\delta^{13}\text{C}/\text{DIC}$ anthropogenic change ratio, $-0.022 \pm 0.004 \text{‰ } \mu\text{mol}^{-1} \text{ kg}$. This anthropogenic CO_2 uptake rate falls within the range of previous estimates in the North Pacific (Sabine et al. 2004) and is similar to both the rate determined for the North Atlantic, $0.63 \pm 0.16 \text{ mol C m}^{-2} \text{ yr}^{-1}$, using the same approach (Quay et al. 2007) and the global ocean mean anthropogenic CO_2 uptake rate of $0.5 \text{ mol C m}^{-2} \text{ yr}^{-1}$ (IPCC 2007).

References

- Canadell, J.G., C. Le Quéré, M.R. Raupach, C.B. Field, E.T. Buitenhuis, P. Ciais, T.J. Conway, N.P. Gillett, R.A. Houghton, and G. Marland. 2007. Contributions to accelerating atmospheric CO₂ growth from economic activity, carbon intensity, and efficiency of natural sinks. *PNAS*. doi:10.1073/pnas.0702737104.
- Feely, R.A., Y. Nojiri, A. Dickson, C.L. Sabine, M.F. Lamb, and T. Ono. 2003. CO₂ in the North Pacific Ocean, PICES working group 13 final report. PICES Scientific Report No. 24, 49pp.
- IPCC. 2007. Climate change 2007: synthesis report. Contribution of working groups I, II, and III to the fourth assessment report of the Intergovernmental Panel on Climate Change. [Core writing team, Pachauri, R.K., and A. Resinger. (eds.)]. IPCC, Geneva, Switzerland, 104 pp.
- Körtzinger, A., P.D. Quay, and R.E. Sonnerup. 2003. The relationship between anthropogenic CO₂ and the ¹³C Suess effect in the North Atlantic Ocean. *Global Biogeochem. Cycles*. **17**, GB1005, doi:10.1029/2001GB001427.
- Ortiz, J.D., A.C. Mix, P.A. Wheeler, and R.M. Key. 2000. Anthropogenic CO₂ invasion into the Northeast Pacific based on concurrent δ¹³C_{DIC} and nutrient profiles from the California Current. *Global Biogeochem. Cycles*. **14**: 917-929.
- Peng, T.H., R. Wanninkhof, R.A. Feely. 2003. Increase of anthropogenic CO₂ in the Pacific Ocean over the last two decades. *Deep-Sea Res. II*. **50**: 3065-3082.
- Quay, P., R. Sonnerup, T. Westby, J. Stutsman, and A. McNichol. 2003. Changes in the ¹³C/¹²C of dissolved inorganic carbon in the ocean as a tracer of anthropogenic CO₂ uptake. *Global Biogeochem. Cycles*. **17**, 1004, doi:10.1029/2001GB001817.
- Quay, P., R. Sonnerup, J. Stutsman, J. Maurer, A. Körtzinger, X. A. Padin, and C. Robinson. 2007. Anthropogenic CO₂ accumulation rates in the North Atlantic Ocean from changes in the ¹³C/¹²C of dissolved inorganic carbon. *Global Biogeochem. Cycles*. **21**, GB1009, doi:10.1029/2006GB002761.
- Sabine, C.L., R.A. Feely, R.M. Key, J.L. Bullister, F.J. Millero, K. Lee, T.-H. Peng, B. Tilbrook, T. Ono, and C.S. Wong. 2002. Distribution of anthropogenic CO₂ in the Pacific Ocean. *Global Biogeochem. Cycles*. **16**, GB1083, doi:10.1029/2001GB001639.
- Sabine, C. L., R. A. Feely, Y. W. Watanabe, and M. Lamb. 2004. Temporal evolution of the North Pacific CO₂ uptake rate. *J. Oceanogr.* **60**: 5-15.
- Sonnerup, R. E., P. D. Quay, and A. P. McNichol. 2000. The Indian Ocean ¹³C Suess effect. *Global Biogeochem. Cycles*. **14**: 903-916.

Takahashi, T., S. C. Sutherland, C. Sweeney, A. Poisson, N. Metzl, B. Tilbrook, N. Bates, R. Wanninkhof, R. A. Feely, C. Sabine, J. Olafsson, and Y. Nojiri. 2002. Global sea-air CO₂ flux based on climatological surface ocean pCO₂, and seasonal biological and temperate effects. *Deep-Sea Res. II.* **49**: 1601-1622.

Watanabe, Y.W., T. Ono, and A. Shimamoto. 2000. Increase in the uptake rate of oceanic anthropogenic carbon in the North Pacific determined by CFC ages. *Mar. Chem.* **72**: 297-315.

Table 1. Summary of published CO₂ uptake rates in the North Pacific Ocean.

Integrated uptake rate (mol C m ⁻² yr ⁻¹)	Approach	Area of study	Years	Reference
0.33 ± 0.05	CFC	North Pacific	1968-1978	Watanabe et al. (2000)
0.44 ± 0.07	CFC	North Pacific	1978-1988	Watanabe et al. (2000)
0.58 ± 0.09	CFC	North Pacific	1988-1998	Watanabe et al. (2000)
1.3 ± 0.5	MLR	North Pacific	1973-1991	Peng et al. (2003)
1.1 ± 0.4	MLR	Central N. Pacific	1973-1999	Feely et al. (2003)
Sabine et al. (2003)				

Fig. 1. Atmospheric time history of CO₂ and δ¹³C since preindustrial times. CO₂ from fossil fuels has a different isotopic composition from the CO₂ in the atmosphere because plants prefer the lighter isotope (¹²C) and therefore have lower ¹²C/¹³C ratios.

Fig. 2. STUD 08 cruise track with δ¹³C sampling stations (marked by x).

Fig. 3. Depth profiles of δ¹³C measured during the CLIVAR P16 cruise in 2006 (diamonds) and the STUD 08 cruise (squares) in the North Pacific Ocean.

Fig. 4. Depth profiles of δ¹³C measured during CLIVAR P16 in 2006 then corrected for lab offsets (dashed line), and δ¹³C measured during STUD 08 (solid line) in the North Pacific Ocean.

Fig. 5. a) Depth profiles of δ¹³C measured during the WOCE 1991 cruise (diamonds), the STUD 08 cruise (squares) and the MLR predicted profiles for 2008 (line) and b) depth profiles of δ¹³C measured during the WOCE 1991 cruise (diamonds), CLIVAR P16 (squares) and MLR predicted profiles for 1991 (line) in the North Pacific Ocean.

Fig. 6. The mean residuals from the isopycnal MLR (squares) compared to the mean residuals from the MLR applied to the entire CLIVAR P16 δ¹³C data set (diamonds). The error bars represent the standard deviation of the mean residuals.

Fig. 7. The δ¹³C measured in the surface layer during WOCE 1991 (diamonds), CLIVAR P16 (squares) and STUD 08 (triangles) along 152°W in the North Pacific Ocean.

Fig. 8. Mean residuals of δ¹³C for WOCE 1991 used to calculate the depth-integrated δ¹³C change, -11.30‰ m yr⁻¹, from 1991 to 2006. The error bars represent the standard deviation of the mean residuals.

Time History of Atmospheric CO₂ and δ¹³C

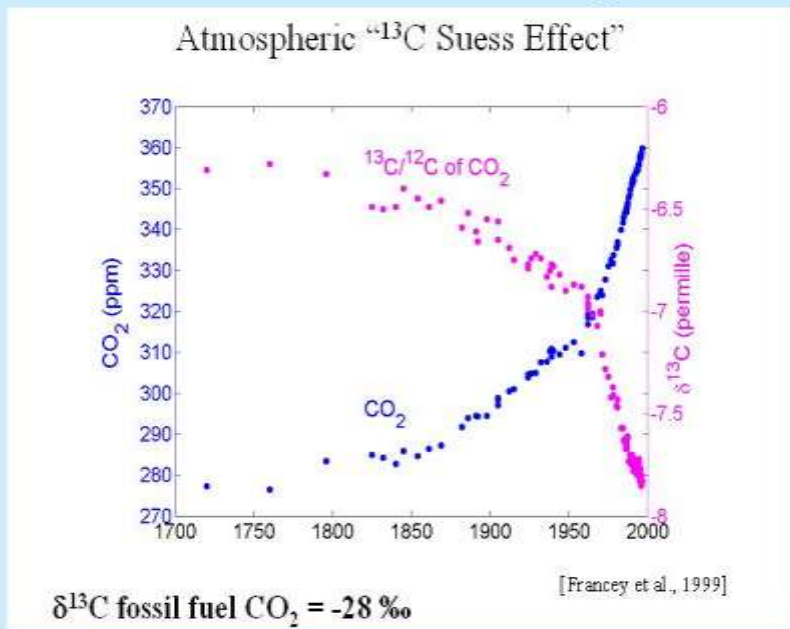


Fig. 1.

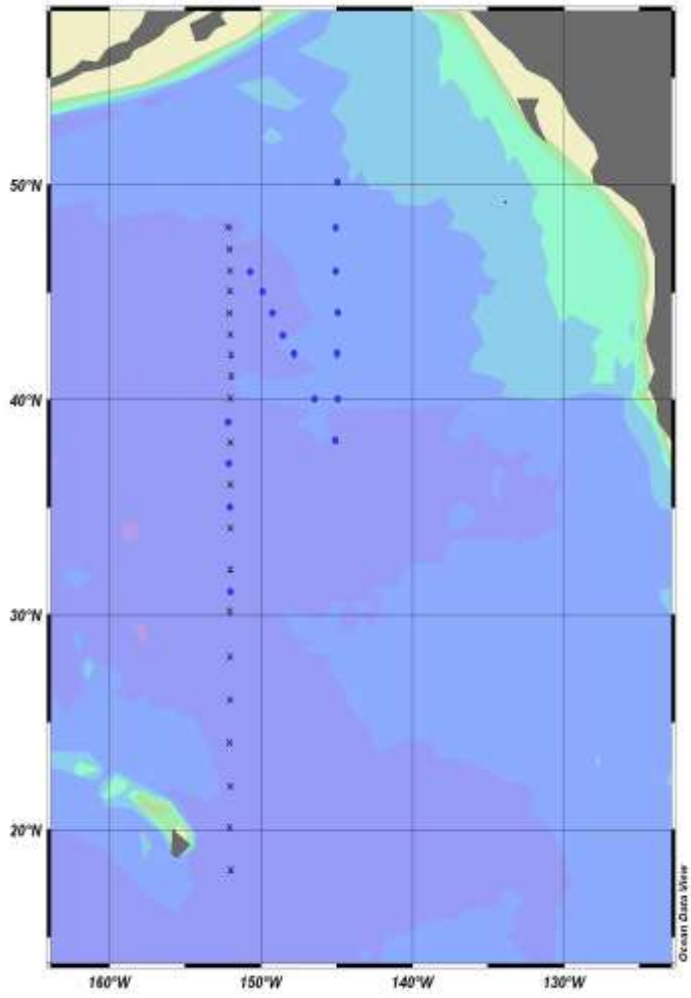


Fig. 2.

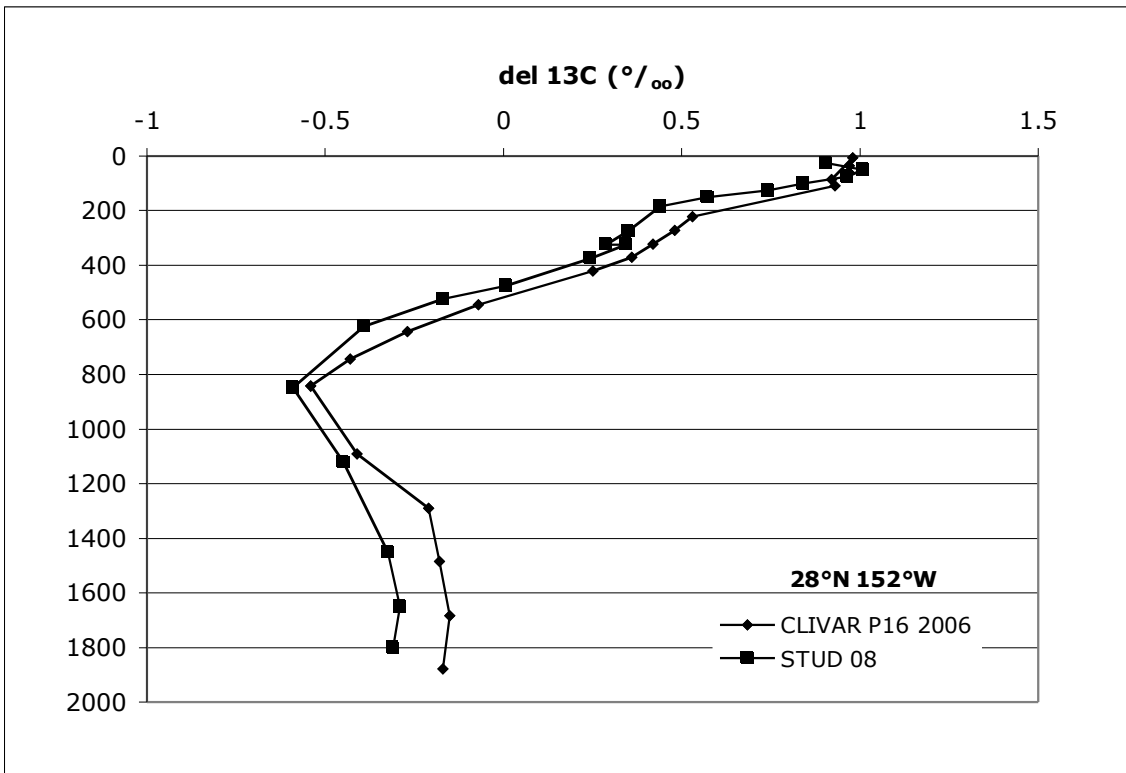
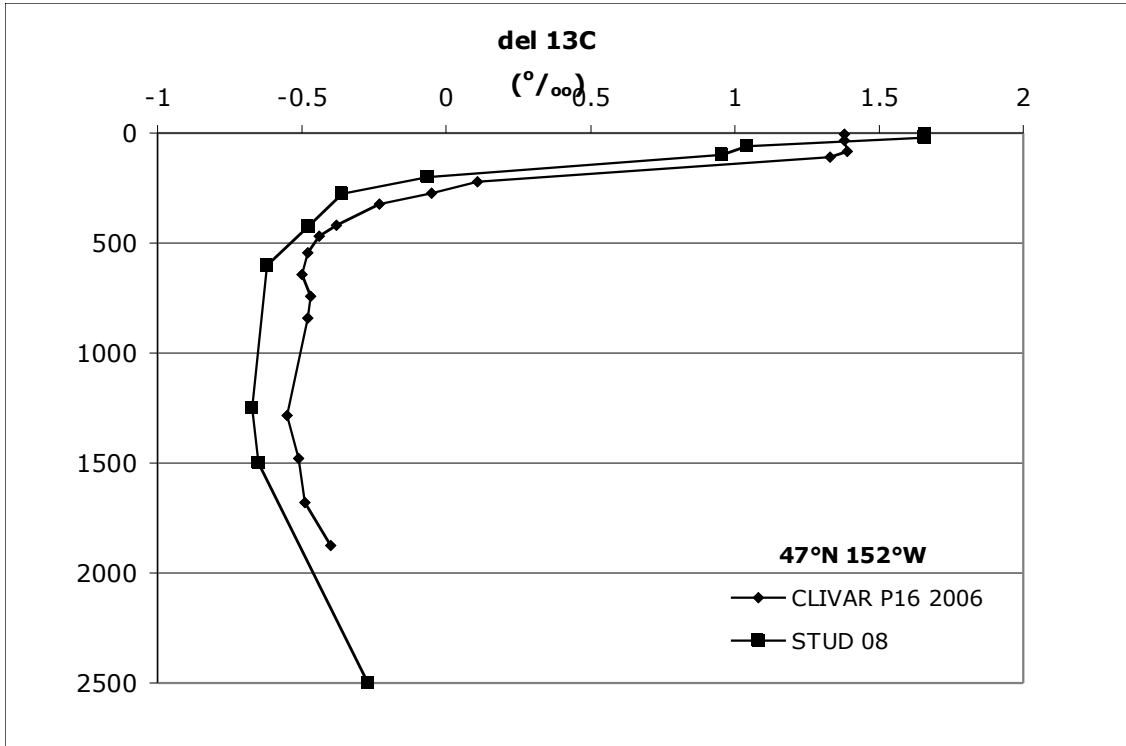


Fig. 3.

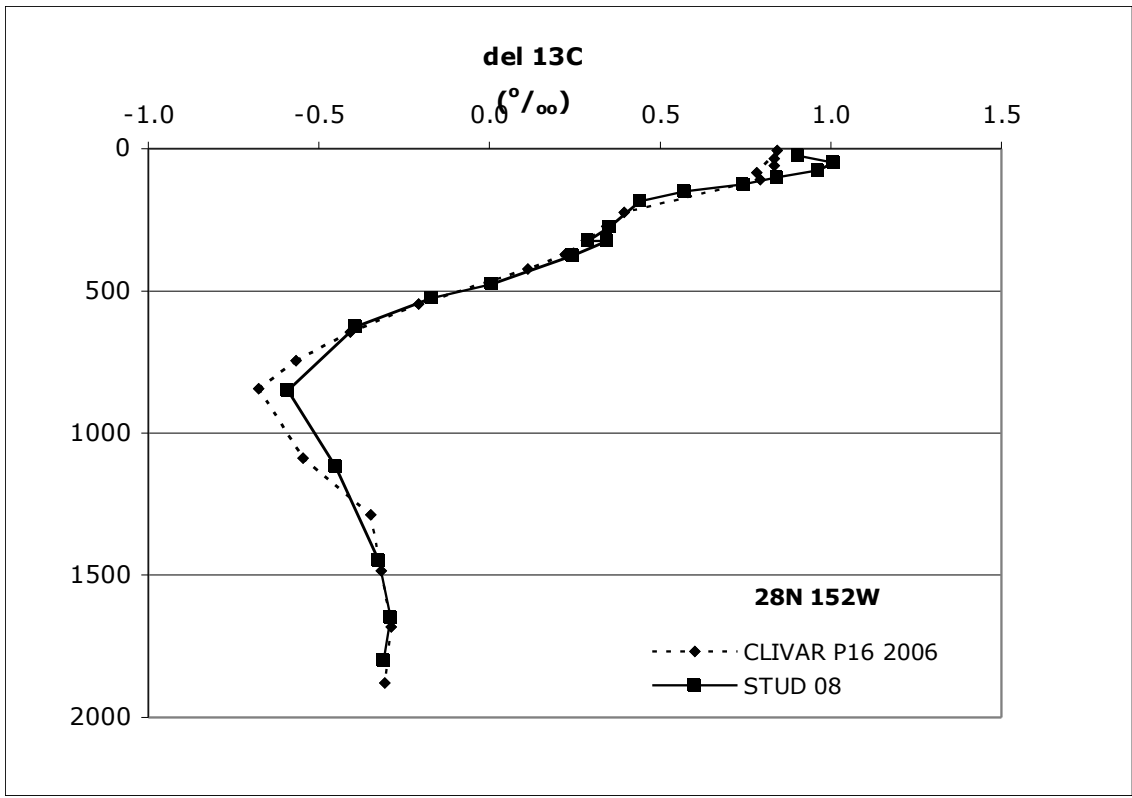
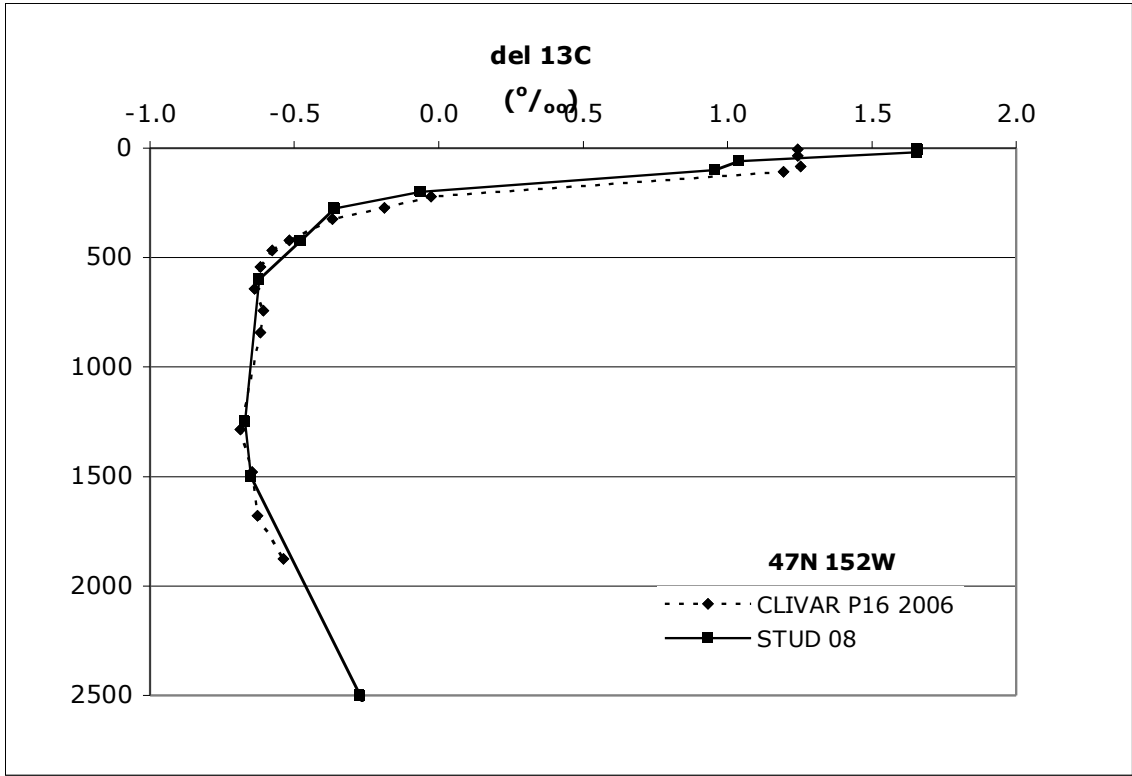


Fig. 4.

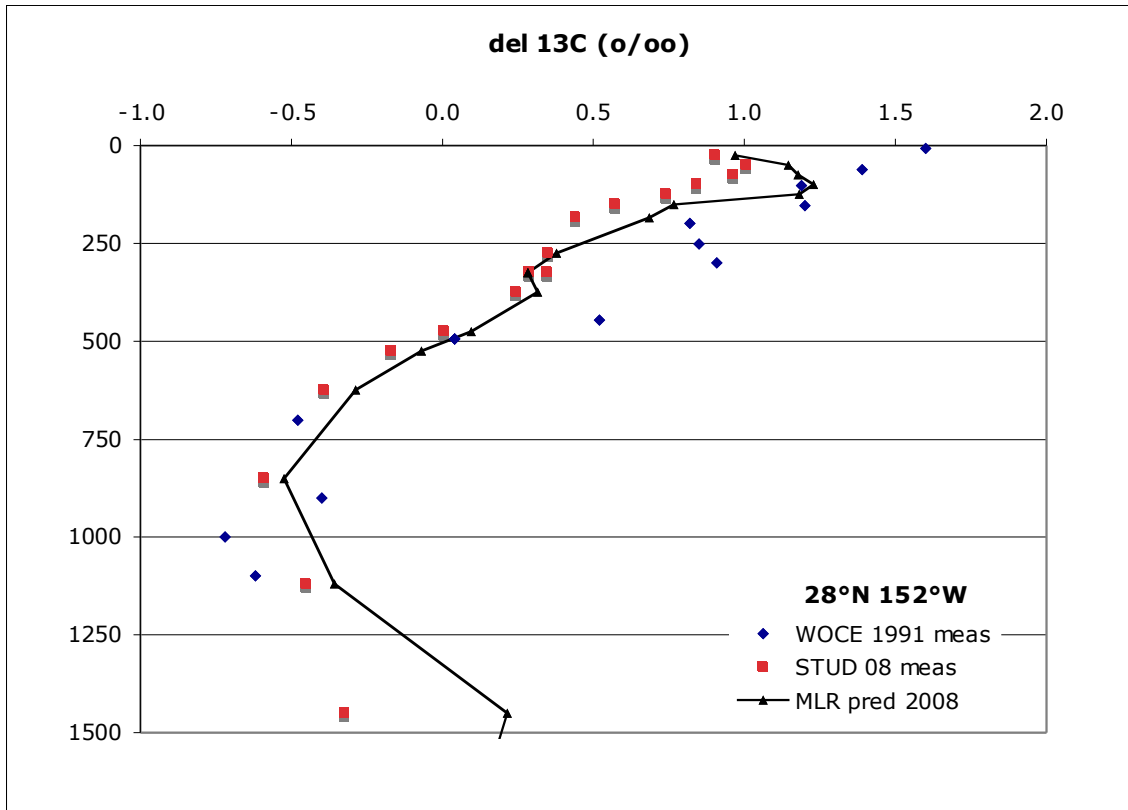
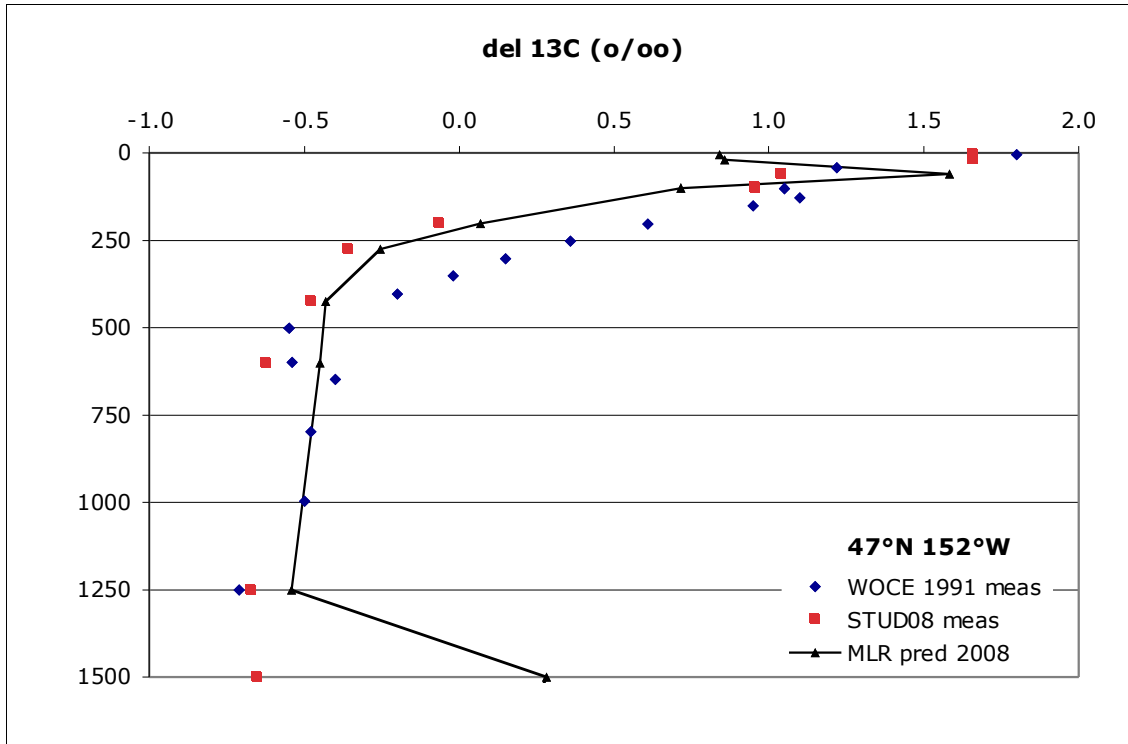


Fig. 5a.

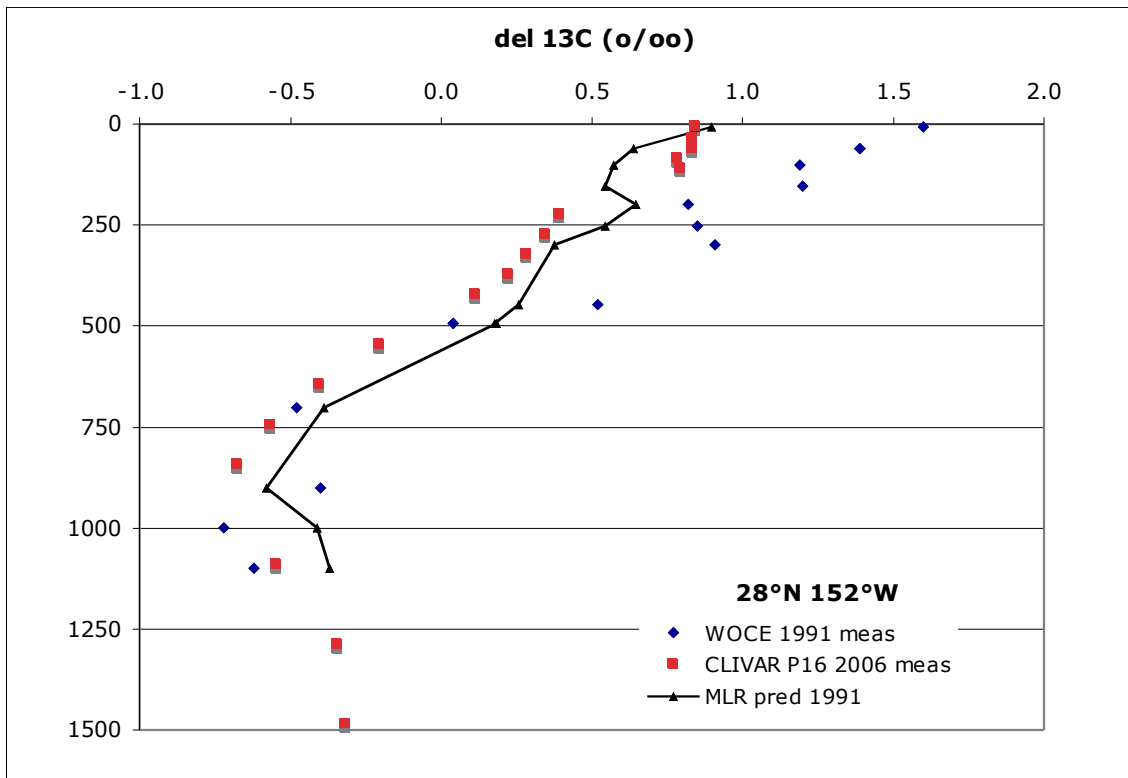
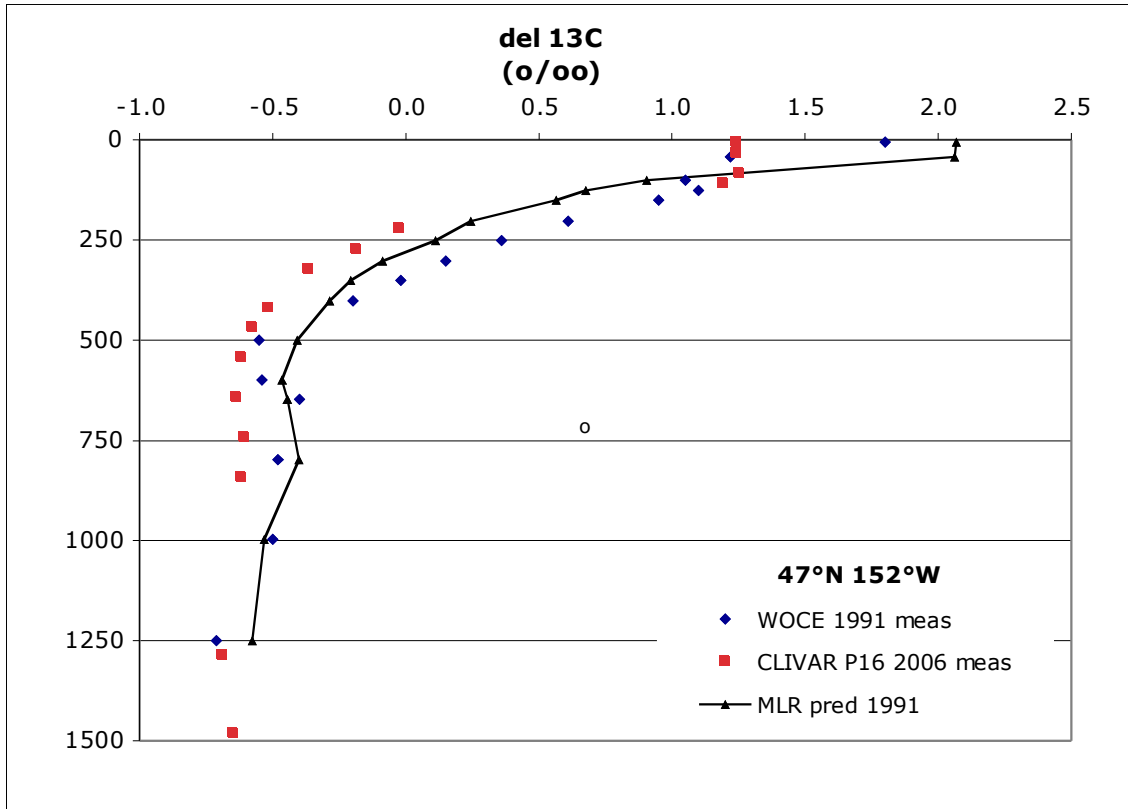


Fig. 5b.

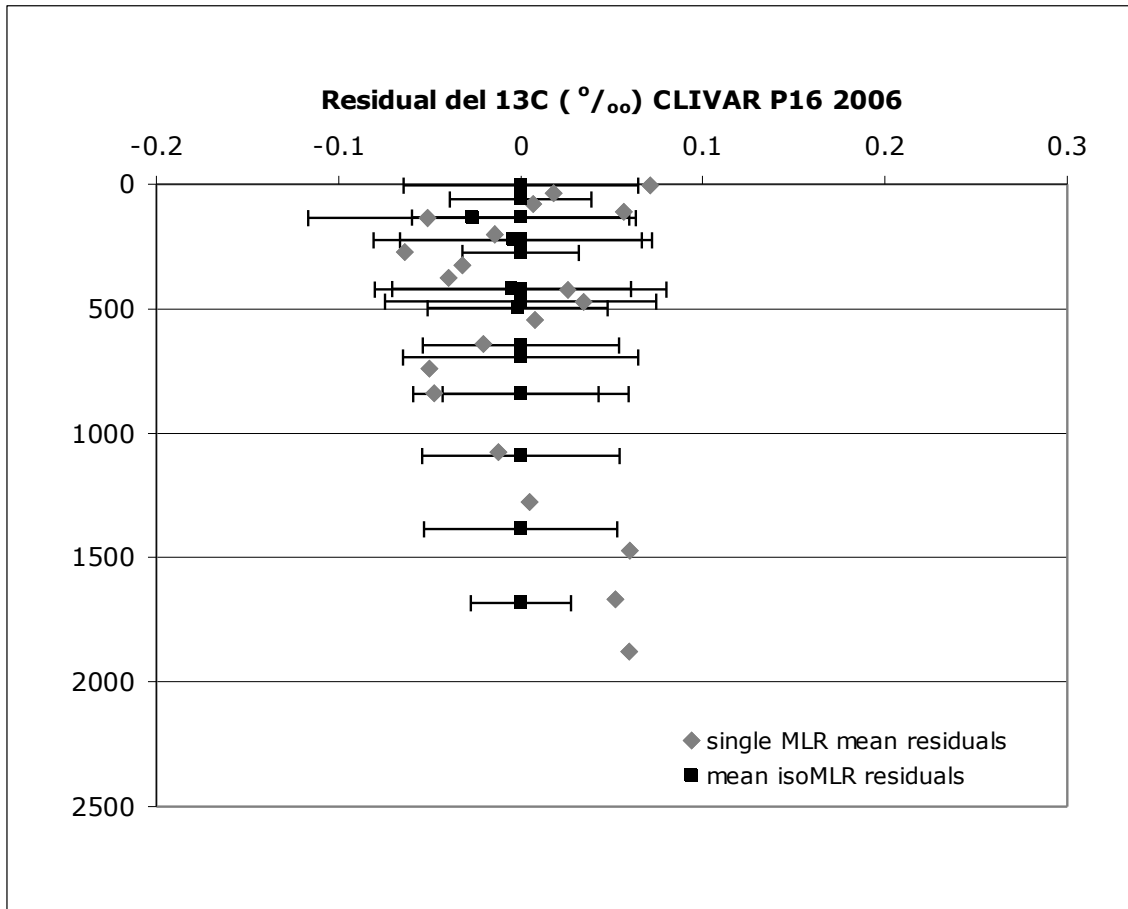


Fig. 6.

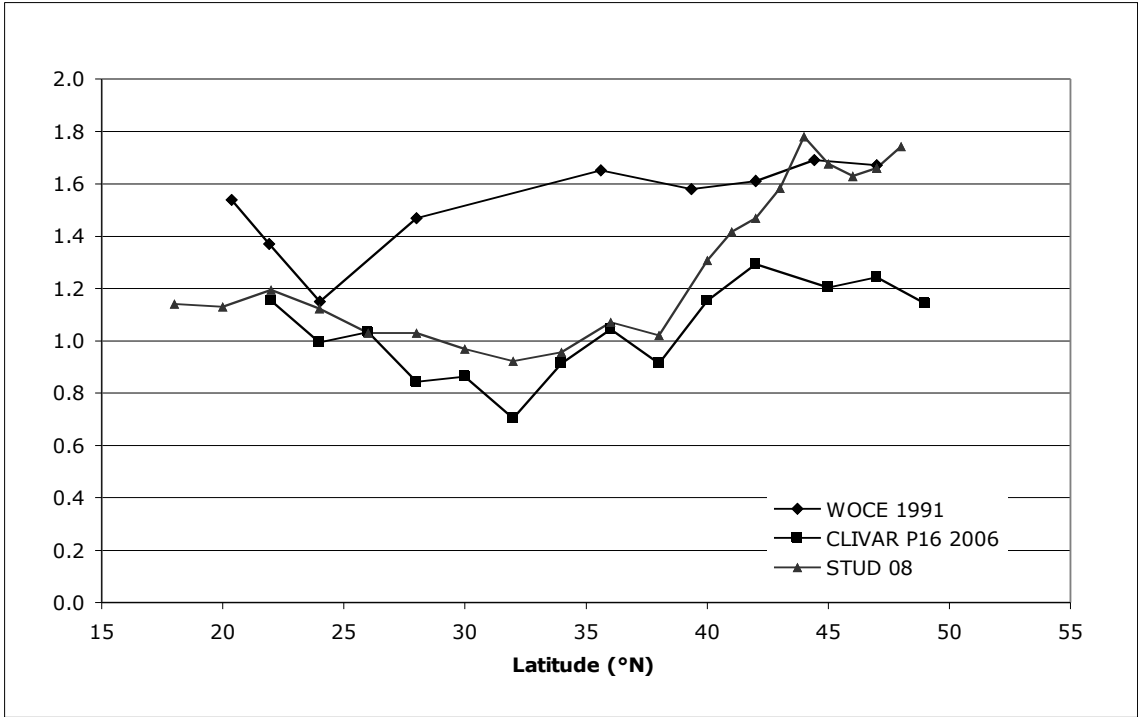


Fig. 7.

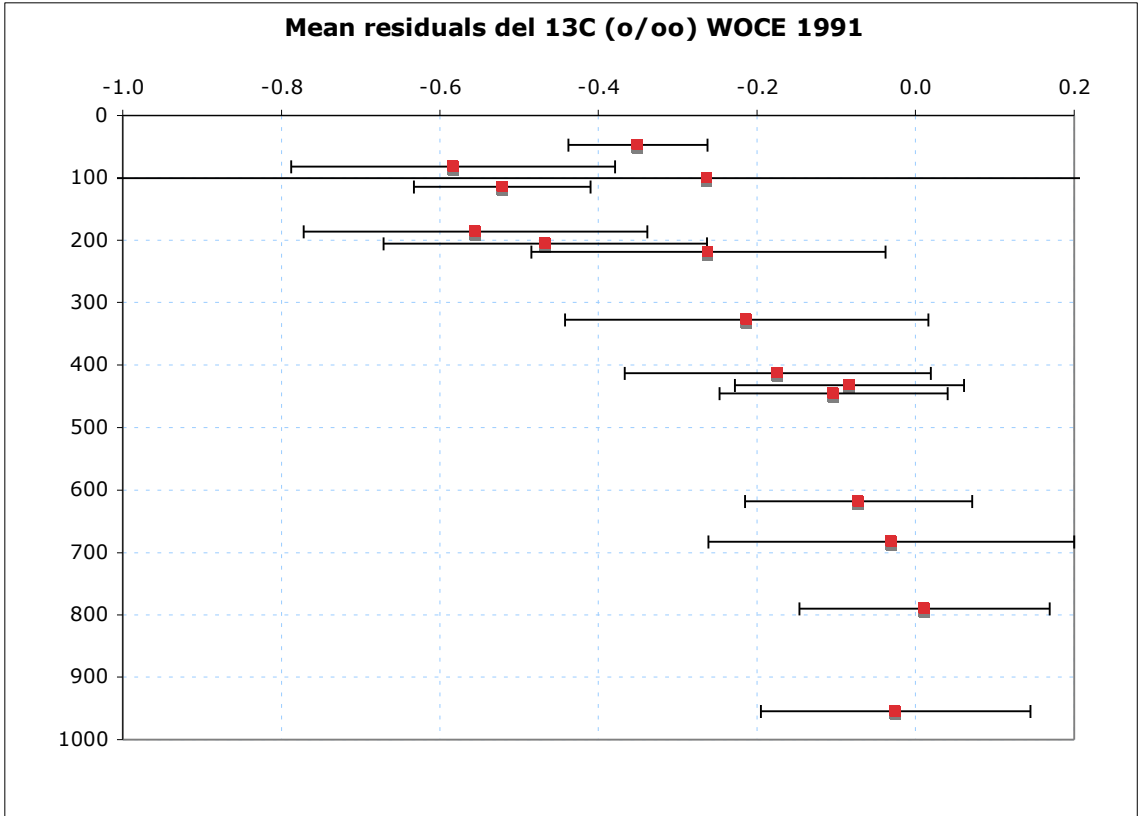


Fig. 8.

