Oxygen Utilization and Organic Carbon Remineralization in the Upper Water Column of the Pacific Ocean

Richard A. Feely[1]*, Christopher L. Sabine[1], Reiner Schlitzer[2], John L. Bullister[1], Sabine Mecking[3] and Dana Greeley[1]

[1]*NOAA/Pacific Marine Environmental Laboratory, Sand Point Way N.E., Seattle, WA 98115, U.S.A.*
[2]*Alfred Wegener Institute for Polar and Marine Research, Columbusstrasse, Bremerhaven, Germany*
[3]*Woods Hole Oceanographic Institution, Woods Hole, MA 02543, U.S.A.*

As a part of the JGOFS synthesis and modeling project, researchers have been working to synthesize the WOCE/JGOFS/DOE/NOAA global CO$_2$ survey data to better understand carbon cycling processes in the oceans. Working with international investigators we have compiled a Pacific Ocean data set with over 35,000 unique samples analyzed for at least two carbon species, oxygen, nutrients, chlorofluorocarbon (CFC) tracers, and hydrographic parameters. We use these data here to estimate in-situ oxygen utilization rates (OUR) and organic carbon remineralization rates within the upper water column of the Pacific Ocean. OURs are derived from the observed apparent oxygen utilization (AOU) and the water age estimates based on CFCs in the upper water and natural radiocarbon in deep waters. The rates are generally highest just below the euphotic zone and decrease with depth to values that are much lower and nearly constant in water deeper than 1200 m. OURs ranged from about 0.02–10 μmol kg^{-1}yr^{-1} in the upper water masses from about 100–1000 m, and averaged = 0.10 μmol kg^{-1}yr^{-1} in deep waters below 1200 m. The OUR data can be used to directly estimate organic carbon remineralization rates using the C:O Redfield ratio given in Anderson and Sarmiento (1994). When these rates are integrated we obtain an estimate of 5.3 $\pm$ 1 Pg C yr^{-1} for the remineralization of organic carbon in the upper water column of the Pacific Ocean.

1. Introduction

Recent estimates of the global export flux of carbon in the world's oceans range from approximately 6–16 Pg C yr^{-1} (Martin *et al.*, 1987; Falkowski *et al.*, 1998, 2000; Lee, 2001). This flux is thought to play a significant role in controlling the atmospheric concentration of CO$_2$ and global climate on millennial time scales (Falkowski *et al.*, 2000). Consequently, concern about the long-term fate of CO$_2$ in the oceans has prompted oceanographers to re-examine the fundamental processes controlling the distributions and fate of dissolved organic carbon (DOC) and particulate organic carbon (POC) in the oceans. Below the topmost euphotic layer of the oceans, the oxidation of DOC and POC results in a net consumption of oxygen. The remineralization of DOC and POC can be represented by the equation:

$$C_\beta H_\xi O_\varphi N_\alpha P + \gamma O_2 = H_3PO_4 + \alpha HNO_3 + \beta CO_2 + \gamma H_2O \tag{1}$$
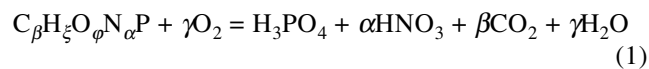
where the balance coefficients α, β, and γ are know as the "Redfield Ratios." Typical Redfield ratios for C:O are 117:170 (Anderson and Sarmiento, 1994), which is within the range of values (0.53–0.85) found by Li and Peng (2002) for the Pacific. The Redfield ratios are useful for estimating the biogeochemical utilization of one element from another. For example, the carbon:oxygen ratio (117/170) times the apparent oxygen utilization (AOU) gives a measure of the organic carbon remineralized (Anderson and Sarmiento, 1994). This change in oxygen concentration times the C:O "Redfield Ratio" provides a direct measure of the organic carbon remineralization in the so-called "Twilight Zone" of the oceans. If the time elapsed since the water mass last outcropped at the surface, i.e., their "ages" are determined, then the vertical integration of the resulting organic carbon remineralization rates provides an estimate of the export flux of carbon. In this pa-

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per, we use a subset of the WOCE/JGOFS/DOE/NOAA global CO_2 survey data from the Pacific Ocean to provide new estimates of organic carbon remineralization rates in the water column of the Pacific Ocean based on changes in AOU and mixing-corrected CFC-12 and natural radiocarbon ages of the water masses.

2. The WOCE/JGOFS/DOE/NOAA Data

Between 1991 and 1996, carbon, oxygen and hydrographic measurements were made on twenty-four cruises in the Pacific Ocean. This research was a collaborative effort between 15 laboratories and 4 countries. At least two carbon parameters, along with oxygen and chlorofluorocarbons (CFCs) and ^{14}C were measured on almost all cruises, but the choice of which carbon pairs were measured varied between cruises. The final data set contained about 35,000 samples. Oxygen measurements were made using the whole bottle technique of Carpenter (1965) employing an automated analyzer described by Friederich *et al*. (1991). The estimated relative accuracy is 0.2% and the precision is 0.3 μmol kg^{-1} (Lamb *et al*., 1997). The corresponding CFC-12 samples were collected from the PVC bottles before any other samples were taken and analyzed by the purge and trap, gas chromatograph/electron capture detector technique described in Bullister and Weiss (1988). As part of the WOCE data quality evaluation process, the entire Pacific CFC data set was carefully examined and individual quality flags assigned to each sample. The estimated overall accuracy of the Pacific WOCE CFC data set is about 1% or about 0.005 picomoles kg^{-1}. All CFC data were converted to the SIO-98 calibration scale (Prinn *et al*., 2000).

3. Results

Our estimates of the oxygen utilization rates in the water column have two components: (1) changes in oxygen concentration due to remineralization of organic matter; and (2) age differences in water masses. Thus, for a given isopycnal surface, the in-situ oxygen utilization rate, OUR, is calculated according to the following equation:

$$OUR = AOU/AGE \qquad (2)$$

where OUR has the units of μmol kg^{-1}yr^{-1}, AOU is the apparent oxygen utilization in units of μmol kg^{-1}, and is calculated using the in-situ oxygen data and the solubility equations of Garcia and Gordon (1992), and the AGE is the water mass age based upon the apparent CFC-12 partial pressures (pCFC-12 age) or the ^{14}C measurements. Similarly, the in-situ organic carbon remineralization rate is calculated according to a similar equation:

$$OCRR = \Delta C_{org}/AGE = R_{C:O}*AOU/AGE \qquad (3)$$

where OCRR is the organic carbon remineralization rate in units of μmol kg^{-1}yr^{-1}, and ΔC_{org}, the change in organic carbon, is the product of $R_{C:O}$ the C:O Redfield ratio for the Pacific (i.e., 0.688) times the AOU.

Figure 1 shows a meridional section of ΔC_{org} along 152°W (a) and two west-east sections along 30°N (b) and 32°S (c). Positive concentrations of ΔC_{org} are found everywhere in the North and South Pacific below the euphotic zone. Strong gradients are observed in the upper 1500 m, where concentrations of ΔC_{org} increase rapidly from <10 to as much as 240 μmol kg^{-1} (Fig. 1(a)). This large increase is primarily located in the thermocline and intermediate water masses of the North and South Pacific. For example, in the North Pacific north of 30°N, ΔC_{org} increases from ~10 μmol kg^{-1} at about 200–400 m to values >200 μmol kg^{-1} at about 900 m. In the South Pacific, where outcropping isopycnals extend to greater depths within the "bowl" of the subtropical gyre, the largest concentration increases occur between 200 m to 1500 m between 30°S and 50°S. Farther south, the increase in ΔC_{org} begins at much shallower depths, consistent with the shoaling of the isopycnal surfaces. The east-west sections (Figs. 1(b) and (c)) show the largest gradients in the far eastern Pacific near the North- and South American coasts due to strong upwelling and higher productivity along the coasts than in the open-ocean regions.

Several investigators have utilized the observed CFC partial pressure (pCFC) in water samples to calculate the "age" of the water, where the age is defined as the average time since the water parcel was in contact with the atmosphere (Doney and Bullister, 1992; Warner *et al*., 1996; Sonnerup *et al*., 1999). This method compares the observed pCFC with a reconstruction of the history of CFCs in the atmosphere to obtain an apparent age. However because of the non-linear atmospheric CFC history, mixing of waters with different pCFCs can introduce significant biases in the pCFC ages compared to the true age of the water. These biases evolve over time, and due to the curvature in the atmospheric concentration histories, pCFC ages are progressively becoming older (e.g., Mecking, 2001). Age biasing due to mixing has been examined using observational data and simple models (Doney *et al*., 1997; Sonnerup, 2001). Mecking (2001) estimated the potential biases between pCFC ages and true ages in a 2-D isopycnal advection-diffusion model of the North Pacific thermocline based on data-derived velocity fields and runs for 19 different isopycnals. In that study, it was estimated that at the time of the North Pacific WOCE cruises (~1993), pCFC-12 ages <=10 years were close to the true age of the water while pCFC-12 ages >10 years were biased increasingly young reaching an age bias of ~8 years for pCFC-12 ages of 25 years. In the present study, OUR (and consecutively OCRR) are determined by plotting AOU versus pCFC-12 ages cor-

Table 1. Oxygen utilization and organic carbon remineralization rates in the Pacific Ocean.

Water mass	Oxygen utilization rate (μmol kg^{-1}yr^{-1})	Organic carbon remineralization rate (μmol kg^{-1}yr^{-1})	Integrated carbon flux (PgC yr^{-1})
Pacific Subarctic Upper Water (200–900 m)	Range: 0.02–9.3 Average*: 4.1	Range: 0.01–6.4 Average*: 2.8	0.28 (0.46)**
Eastern North Pacific Transition Water (200–800 m)	Range: 0.03–5.2 Average*: 3.4	Range: 0.02–3.6 Average*: 2.3	0.10 (0.26)**
Western North Pacific Central Water (200–1100 m)	Range: 0.07–5.8 Average*: 4.5	Range: 0.05–4.0 Average*: 3.1	1.32 (0.49)**
Eastern North Pacific Central Water (200–700 m)	Range: 0.06–5.8 Average*: 3.7	Range: 0.04–4.0 Average*: 2.6	0.45 (0.62)**
Pacific Equatorial Water (200–700 m)	Range: 0.12–9.3 Average*: 4.6	Range: 0.08–6.4 Average*: 3.1	0.27 (0.34)**
Eastern South Pacific Transition Water (200–900 m)	Range: 0.13–6.5 Average*: 4.3	Range: 0.09–4.5 Average*: 3.0	0.08 (0.40)**
Western South Pacific Central Water (200–1000 m)	Range: 0.07–4.8 Average*: 2.1	Range: 0.05–3.3 Average*: 1.4	0.30 (0.50)**
Eastern South Pacific Central Water (200–1100 m)	Range: 0.2–6.7 Average*: 3.5	Range: 0.13–4.6 Average*: 2.4	1.16 (0.94)**
Subantarctic Surface Water (200–1400 m)	Range: 0.12–4.9 Average*: 2.1	Range: 0.08–3.4 Average*: 1.5	0.56 (0.72)**
Antarctic Surface Water (200–1800 m)	Range: 0.04–3.9 Average*: 3.0	Range: 0.03–2.7 Average*: 2.0	0.73 (0.28)**
		Total	**5.25** **(5.01)***

*Volume weighted average.
**Estimates based on the inverse modeling approach of Schlitzer (2000) are given in parentheses.

rected for these mixing biases using the results from the advection-diffusion model by Mecking (2001) along isopycnal surfaces, using the WOCE global survey data. The slope of a linear regression to these data gives the OUR (Table 1). The results were separated into 10 regions (Fig. 2) based on the upper water distributions outlined in Emery and Meincke (1986). The OUR values calculated in this manner were binned by depth increments and the mean and standard deviation were computed. Note that the age corrections, though derived from a model for the North Pacific, were applied to all the data. While the mixing in the equatorial Pacific and in the South Pacific may be different from the North Pacific, this is justified by the fact that the age biases depend to a large extent on the curvature of the atmospheric CFC histories and less on the strength of the mixing (Mecking, 2001). Also because of uncertainties in the open boundary condition of the model, the pCFC-12 age biases derived from the model provide minimum estimates for the age corrections. True ages could be older, and thus OURs estimated here are upper bounds (Mecking et al., 2003).

The volume weighted average OUR estimates for each region are presented in Table 1. Note that the depth ranges for each region are given in parentheses. The OUR

estimates reach a maximum of up to as much as approximately 10 μmol kg^{-1}yr^{-1} in the thermocline waters just below the euphotic zone and then decrease rapidly thereafter. The highest average OUR rates are observed in the highly productive equatorial Pacific and the western North Pacific; whereas the lowest average rates are in the oligotrophic waters of the western subtropical South Pacific. These results suggest that the OUR estimates are, to some degree, directly related to the productivity and export flux of carbon from the overlying water mass Lutz et al. (2002).

Since the pCFC-age method is limited to water mass ages from about 5–35 years (Sonnerup, 2001), we used natural ^{14}C ages to calculate OUR in deeper waters. The ^{14}C decay rate of 1% every 83 years makes this isotope a useful age tracer for processes in the deep sea (Rubin and Key, 2002). The bomb C-14 contamination was determined by first calculating the bomb C-14 distribution using the latest method of Rubin and Key (2002), removing this signal from the total C-14 signal to get the natural radiocarbon. We then used the change in natural radiocarbon distributions along isopycnal surfaces within each region to obtain the water mass age differences. This process limits mixing effect to only those within the re-
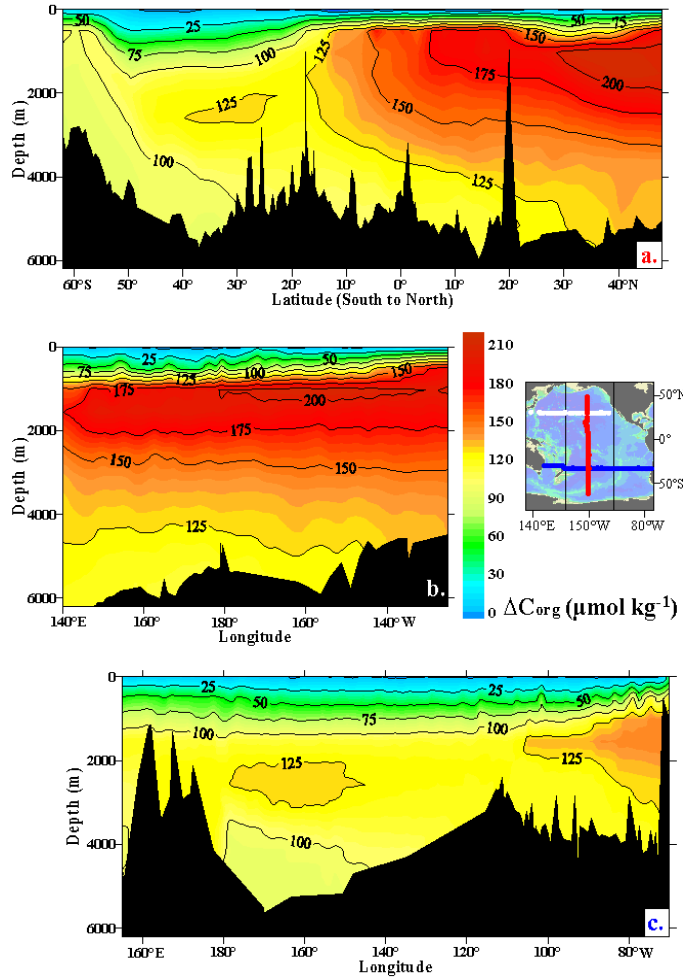
Fig. 1. Distribution of remineralized organic carbon in μmol kg^{-1} for: (a) P16 along 152°W; (b) P2 along 30°N; and (c) P6 along 30°S.
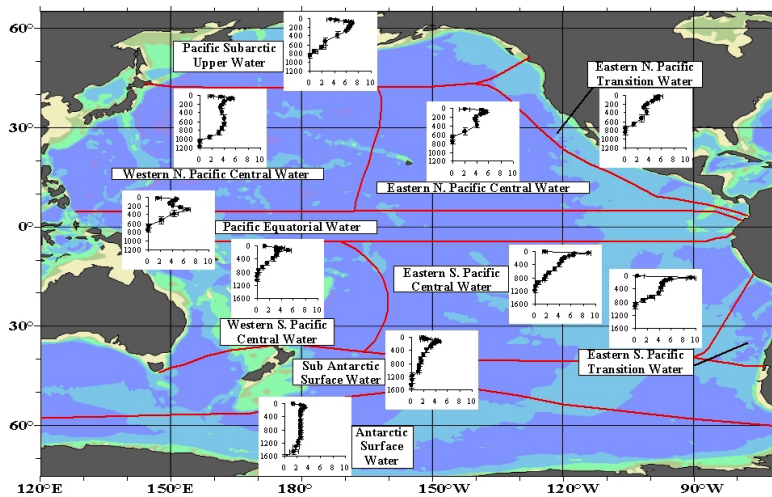

Fig. 2. Vertical Profiles of organic carbon remineralization rates in units of μmoles kg^{-1}yr^{-1} for the Pacific Ocean. The water masses shown in this figure are based on the upper water distributions outlined in Emery and Meincke (1986) and correspond to the water masses described in Table 1.

gion. The choice of where the natural radiocarbon distributions were used depended on the CFC distributions and varied from region to region, ranging from as shallow as 600–1100 m in the eastern North Pacific region to 1500–1800 m in the Antarctic region. The depth limit for each region was picked based on where the detection limit of ~0.05 μmol kg^{-1}yr^{-1} was reached. For waters >900 m the average OUR in the Pacific Deep Water was determined to be 0.10 μmol kg^{-1}yr^{-1}, which is within the range of values (0.03–0.19 μmol kg^{-1}yr^{-1}) reported by previous investigators (see Chen, 1990; and references therein). Higher OUR values were observed in the North Pacific relative to the South Pacific. The average OUR in the shallow waters immediately below the euphotic zone of the North Pacific is more than 10 times faster than the deep-water remineralization rates.

Comparison of these new results for OUR from the WOCE/JGOFS/DOE/NOAA global CO$_2$ survey data are in good agreement with the OUR values from the earlier analysis of the North Pacific pre-WOCE cruise data (9 and 15 μmol kg^{-1}yr^{-1}) given in Warner et $al.$ (1996) for shallow depths. They are also consistent with the more recent data (8.6 and 4.9 μmol kg^{-1}yr^{-1}) as reported in Mecking (2001) for the 25.6 and 26.0 potential density surfaces, respectively. While there have been changes in AOU in some parts of the eastern subtropical North Pacific over the past two decades, it is not clear whether these are due to changes in circulation or due to changes in new production (see Emerson et $al.$, 2001; Mecking, 2001). Hence, further studies and data collection will be necessary to put the OUR estimates reported here into a time-varying context.

4. Discussion

The new results from the WOCE/JGOFS/DOE/NOAA global CO$_2$ survey in the Pacific Ocean indicate that water column remineralization of organic carbon in shallow waters <1500 m accounts most of the total organic carbon remineralization in the water column, consistent with the decreases in the sediment trap POC flux data for the Pacific Ocean reported by Berelson (2001) and Lutz et $al.$ (2002). These results imply that the resupply of dissolved inorganic carbon to the surface waters via shallow water remineralization processes may vary regionally. The average OCRR estimates presented in Table 1 indicate more than a 2-fold difference between the OCRRs in highly productive regions as compared with regions of low productivity (Fig. 2). This is consistent with the general depth-dependent model of particulate carbon fluxes observed at the VERTEX stations seaward of the North American coast (Martin et $al.$, 1987; Lutz et $al.$, 2002), suggesting that remineralization of organic matter is primarily controlled by the downward flux and reminearalization of particulate organic matter. Recently,
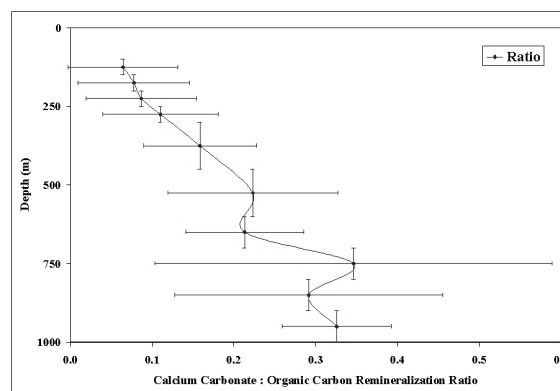

Fig. 3. Vertical Profile of the mean CaCO$_3$:C$_{org}$ remineralization ratios for the Pacific Ocean. The CaCO$_3$ data are from Feely et $al.$ (2002).

Armstrong et $al.$ (2002) have suggested that the formation and dissolution of ballast minerals (e.g., CaCO$_3$ and biogenic silica particles) play a major role in the controlling the remineralization process. Their model results suggest that regions of high CaCO$_3$ and biogenic silica flux and dissolution will contribute to higher rates of organic carbon remineralization by providing a substrate for the downward flux of organic matter. Our results qualitatively show that the highest organic carbon remineralization rates are also in the same regions where CaCO$_3$ and biogenic silica fluxes (i.e., Subarctic Pacific, Equatorial Pacific, etc.) are also high (Balch and Kilpatrick, 1996; Berelson et $al.$, 1997; Takahashi et $al.$, 2000).

The OCRR estimates in Table 1 have been integrated within each region to provide an estimate of the amount of organic carbon that is annually remineralized in the water column over the depth range indicated in column 1. The integrated remineralization estimates are shown in the last column of Table 1. The total amount of organic carbon that is remineralized in the upper water column of the Pacific is estimated to be 5.3 $\pm$ 1 Pg C yr^{-1} using the mixing corrected apparent CFC-12 ages and 6.9 $\pm$ 1 Pg C yr^{-1} using the apparent CFC-12 ages uncorrected for mixing (not shown). These estimates are uncertain at about the 20% level. They agree well with independent estimates based on the inverse modeling approach of Schlitzer (2000, 2002) as shown in parentheses in column 4 of Table 1. The modeled fluxes show less region-to-region variability than the mixing-corrected estimates, but the water column integrated fluxes for the Pacific Ocean are nearly the same. The mixing-corrected estimates are also consistent with the recent estimates of the export flux of carbon based on the data-based estimates of Lee (2001) and well within the ranges given in the earlier studies (Falkowski et $al.$, 1998, 2000).

The method described above is sensitive to the choice of C:O ratio used. For example, if the mean C:O ratio of 0.765 for the North Pacific, derived from a subset of the WOCE and GEOSECS data sets (Li and Peng, 2002), is used instead of the ratio proposed by Anderson and Sarmiento (1994) the annual organic carbon remineralized in the water column would be about 11 percent higher than the one presented here. However, as also discussed in Li and Peng (2002), the North Pacific mean ratio of 0.765 is significantly higher than the mean C:O ratio for the South Pacific, 0.573. This result is probably due to higher oxygen consumption in some regions of the North Pacific as a result of conversion of organic nitrogen into N_2 and N_2O via nitrification/denitrification processes. Thus, there appears to be regional differences in C:O ratios, depending on which biogeochemical processes predominate (Peng and Broecker, 1987; Li et al., 2000). The C:O ratio used for the estimates presented here is very close to the mean of all the Pacific data given in Li and Peng (2002). Clearly, additional studies are needed to provide more detailed determinations of the temporal and spatial variability of the C:O ratios in the Pacific Ocean.

4.1 $CaCO_3$:C_{Org} remineralization ratios

Over the past decade, there has been considerable discussion in the oceanographic literature concerning the export ratio of $CaCO_3$:C_{Org} from the euphotic zone (Bacastow and Maier-Reimer, 1990; Maier-Reimer, 1993; Yamanaka and Tajika, 1996; Aumont, 1998; Naijar and Orr, 1998; Archer et al., 2000; Matsumoto et al., 2002; Sarmiento et al., 2002). The motivation for this research is that over time scales of centuries to millennia significant changes in this ratio averaged over the global oceans will act to modify the distribution of CO_2 between the oceans and atmosphere. For example, Archer et al. (2000) showed that a decline of this ratio could have contributed to the decrease in atmospheric CO_2 during the last ice age. Previous estimates of $CaCO_3$:C_{Org} export ratios, based on the analysis of observational data and model results, range from 0.05 to 0.33 (see table 1 in Sarmiento et al., 2002). Recently, using a biogeochemical-transport box model and the WOCE bottle data, Sarmiento et al. (2002) determined a global average ratio of 0.06 ± 0.03 for the export ratio of $CaCO_3$:C_{Org} from of the euphotic zone which was consistent with other recent models (Yamanaka and Tajika, 1996; Naijar and Orr, 1998). If most of the organic matter is recycled in the upper 1000 m of the water column and most of the $CaCO_3$ dissolution occurs in the upper 1300 m of the water column (Feely et al., 2002), then the ratio of the integrated remineralization rates should give a reasonable lower limit of the $CaCO_3$:C_{Org} export ratio. Figure 3 shows a plot of the mean profile of the $CaCO_3$:C_{Org}, based on the remineralization rates for the North- and South Pacific

given in Table 1 and the $CaCO_3$ dissolution rates given in Feely et al. (2002). The low remineralization ratio, 0.06 ± 0.07, just below the euphotic zone is in reasonable agreement with the recent findings of Sarmiento et al. (2002). However, the remineralization ratios increase with depth to values averaging about 0.33 ± 0.15 at depths near 1000 m. These results may explain some of the earlier higher data-based estimates since they were based on changes in the concentrations of dissolved inorganic carbon and total alkalinity in the water column (Li et al., 1969; Tsunogai, 1972; Broecker and Peng, 1982; Chen, 1990). The overall average for the water column to a depth of 1000 m is 0.17 ± 0.14. This value is close to the mean of the previous estimates (see table 1 in Sarmiento et al., 2002). Thus, the variability of the different estimates of the export ratio for $CaCO_3$:C_{Org} are, to a large extent, a function of the variability associated with the different methods used to estimate them. However, if the value of the total amount of remineralized $CaCO_3$ in the Pacific (i.e., 0.31 Pg C yr^{-1}; from Feely et al., 2002) is divided by the total amount of remineralized organic carbon in the Pacific (i.e., 5.3 Pg C yr^{-1} from Table 1) a ratio of 0.06 is obtained which is in excellent agreement with the model-based estimate of Sarmiento et al. (2002). Our estimate should be considered to be a lower limit because a larger fraction of the $CaCO_3$-carbon reaches the seafloor than the organic carbon in the Pacific Ocean (Berelson et al., 1997).

5. Conclusions

The extensive amount of data collected as part of the WOCE/JGOFS global CO_2 survey has provided us with the first detailed view of remineralization processes in the Pacific Ocean. By coupling the carbon system parameters with the tracer data we have been able to determine regional differences in organic carbon remineralization rates, indicating more than a 2-fold drop in the rates from the highly productive regions to the regions of low productivity. The total amount of organic carbon that is remineralized in the upper water column is estimated to be approximately 5.3 ± 1 Pg C yr^{-1}, which is consistent with the recent estimates of the export flux of carbon in the Pacific.

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