Anthropogenic CO₂ Distribution in the North Pacific Ocean

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The depth of penetration of anthropogenic CO₂ in the North Pacific Ocean based on carbonate data in the literature is discussed. The results indicate that the deepest penetration (over 2000 m) is found in the northwest North Pacific. The shallowest penetration (to less than 400 m) is found in the eastern equatorial Pacific. Depth of penetration of anthropogenic CO₂ appears to have been controlled by such factors as deep water formation in the Northwest Pacific; upwelling in the equatorial Pacific and; vertical mixing in the western boundary areas. These results compare well with results implied from tritium, C-14, and freons distributions. The total inventory of excess carbon in the North Pacific was $14.7 \pm 4 \times 10^{15}$ g around 1980.

1. Introduction

It has been known for decades that the concentration of carbon dioxide in the atmosphere has been increasing due to burning of fossil fuels and clearing of forests over the last 140 years. The matter is of great concern because the so called “excess” CO₂ in the atmosphere enhances the greenhouse effect which may significantly change the climate on earth. The rate of the climate change depends on the rate of CO₂ increase in the atmosphere. How much CO₂ is left in the atmosphere is in turn controlled by the amount taken up by the oceans (Chen and Drake, 1986).

The estimates of oceanic uptake of CO₂ come primarily from models with varying complexity, ranging from the simple one-dimensional box-diffusion and outcrop-diffusion models to the sophisticated 3-dimensional ocean general circulation models (Maier-Reimer and Hasselmann, 1987; Sarmiento et al., 1992). Recently it has been shown that the oceanic penetration of excess CO₂ can be calculated directly using carbonate data (Brewer, 1978; Chen and Millero, 1978; Chen and Pytkowicz, 1979; Kanamori and Ikegami, 1982; Chen, 1982a, b, 1987; Cline et al., 1985; Chen et al., 1986; Poisson and Chen, 1987; Krumgalz et al., 1990; Anderson and Jones, 1991; Ono et al., 1992). Because the method is subject to large uncertainties, the accuracy of the results is not known (Chen and Millero, 1979; Shiller, 1981; Chen et al., 1982; Broecker et al., 1985). However, the precision of the method is adequate to show the excess CO₂ signal. The abundant carbonate data in the literature can thus be used to supplement the tracer data in showing oceanic mixing features for waters formed in the last 140 years.

2. Method of Calculation

The method of computation and its limitations have been described in detail elsewhere (Chen and Millero, 1979; Chen and Pytkowicz, 1979; Chen et al., 1982, 1990; Chen, 1985). The following equations summarize the method.

\[ \Delta NTCO_2 ° = NTCO_2 ° (\text{old}) - NTCO_2 ° (\text{present}) \]
\[ = NTCO_2 (\text{measured}) - 0.5 NTA (\text{measured}) - 0.78 \text{AOU} \]
\[ +0.5 NTA° (\text{present}) - NTCO_2 ° (\text{present}) \] (1)
where $\Delta NT_{CO_2}^\circ$ is the difference of the preformed total CO$_2$ values for a water formed some time ago and for a water formed recently. NT$_{CO_2}$ and NTA are the normalized total CO$_2$ and total alkalinity respectively ($NT_{CO_2} = \text{total CO}_2 \times 35/S$; $NT_A = \text{alkalinity} \times 35/S$), node indicates preformed values, AOU is the apparent oxygen utilization.

The above equation is rearranged:

$$
\left[ NT_{CO_2} \, \text{(measured)} - NT_{CO_2}^\circ \, \text{(present)} \right] - 0.5 \left[ NT_A \, \text{(measured)} - NT_A^\circ \, \text{(present)} \right] = 0.78 \, \text{AOU} + \Delta NT_{CO_2}^\circ
$$

$$
\Delta NT_{CO_2} - 0.5 \, \Delta NT_A = 0.78 \, \text{AOU} + \Delta NT_{CO_2}^\circ.
$$

Hence, if ($\Delta NT_{CO_2} - 0.5 \, \Delta NT_A$) is plotted against AOU a slope of 0.78 should be obtained and the intercept is $\Delta NT_{CO_2}^\circ$ observed in old waters formed prior to industrialization (Chen and Pytkowicz, 1979). Results based on selected GEOSECS data below the AOU maximum layer is shown in Fig. 1. The slope is $0.78 \pm 0.05$ and the intercept is $-40 \, \mu$mol/kg. This indicates that old seawaters contain 40 $\mu$mol/kg less total CO$_2$ than present day seawaters. We now add 40 $\mu$mol/kg to the calculated $\Delta NT_{CO_2}^\circ$, we obtain the excess CO$_2$ signal. For surface waters, $\Delta NT_{CO_2}^\circ$ approaches zero and the excess CO$_2$ concentration is thus 40 $\mu$mol/kg. For deep waters formed prior to industrialization $\Delta NT_{CO_2}^\circ \approx -40$ and thus the excess CO$_2$ concentration approaches zero. Different interception is obtained for different data sets depending on the year samples were collected.

3. Sources of Data

YALOC 69 (Yaloc, 1969) 31 stations (Wyatt et al., 1970);
KH-71-5 (Hakuho Maru, 1971) 13 stations (Tsubota, 1973);
GEOSECS (Melville, 1973) 72 stations (Takahashi et al., 1980);
KH-75-4 (Hakuho Maru 1975) 12 stations (Hattori, 1977);
INDOPAC (Melville, 1976) 36 station (INDOPAC, 1978);  
KH-78-3 (Hakuho Maru, 1978) 7 stations (Hattori, 1979);  
KH-80-2 (Hakuho Maru, 1980) 3 stations (Horibe, 1983);  
CO₂ Dynamics Cruise ENP (Miller Freeman, 1981) 10 stations (Chen et al., 1986);  
CO₂ Dynamics Cruise WNP (Discoverer, 1982) 10 stations (Chen et al., 1986);  
Outer Continental Shelf Environmental Assessment Program (Polar Sea, 1983) 2 stations (Chen et al., 1985);  

4. Penetration Depth along 150°W and 165°E Cross-Sections

The lower boundary of the excess CO₂ penetration is defined as the depth at which the excess CO₂ concentration is less than 5 μmol/kg, approximately 12% of the magnitude of the signal. Because of the uncertainty of the method and the large vertical spacing of the samples, the lower boundary may be up to 150 m too deep or up to 300 m too shallow.

Figure 2(a) shows the results of fossil-fuel CO₂ penetration along the 150°W cross-section (CO₂ Dynamics, Eastern North Pacific; ENP). The waters underlying the 0 μmol/kg excess CO₂ contour are free from fossil fuel CO₂. The increasing excess CO₂ from the deep to the surface layer indicates that the fossil-fuel CO₂ content is greater in the shallow water than in the deep water. Chen (1982a) has discussed the penetration of excess CO₂ along the ENP section. He proposed that the deepening of penetration depth at 30°N (Fig. 2(a)) is not a local phenomenon but is an advected feature derived from the northwest North Pacific. Local convergence in the winter only brings surface water to approximately 300 m (Niiler and Reynolds, 1984). Tsunogai et al. (1992) reported that the Intermediate Waters in the North Pacific are an important excess CO₂ sink.

The values of excess CO₂ plotted vs σθ in Fig. 3 cannot be contoured in detail but nevertheless provide supporting information (Chen et al., 1986). Chen (1982a) gives detailed discussion of the excess CO₂ along this section. The isopleths less than 10 μmol/kg are relatively flat and the 0 μmol/kg contour which is indicative of old water is at or below ~27.2 σθ throughout the section. At values greater than 10 the distribution is very non-isopycnal, with a value greater than 20 centered in the S-min at the intermediate depth, indicating that younger water is associated with these strata. Since this water originates in the northwest Pacific as a result of near surface processes, this higher excess CO₂ indicates more recent surface origin.

The shallow excess CO₂ values (i.e. at values greater than 10) are more closely aligned with the AOU and salinity isopleths than the other parameters (Chen et al., 1986). In the vicinity of the subarctic front near 45°N the strongest north-south gradient exists and extends to depths of about 600 m and σθ of 27.00. Between 50°N and 40°N the 20 μmol/kg isopleth shallows from the 26.8 σθ isopleth to the 25.5 σθ isopleth, suggesting that vertical processes are able to rejuvenate the water north of the subarctic front more effectively than south of this front.

The 26.8 σθ density surface was chosen by Fine et al. (1981, 1983) as the lower boundary of the intermediate layer containing the most tritium in the North Pacific. Evidently excess CO₂ has penetrated much deeper than this level. The 26.8 σθ density surface is just below the maximum outcrop density for the winter North Pacific and thus diffusion downward from the mixed layer and lateral ventilation along outcrops provided a direct atmospheric link. The depth of this surface was from 800 m at 30°N in the western North Pacific to 150 m further north and to 300 m in the equatorial regions. For the 150°W section presented here the depth of this isopycnal is generally near 500 m and somewhat deeper near 30°N as part of the geostrophic
Fig. 2. Cross-section of excess CO$_2$ along 150°W from (a) carbonate data (taken from Chen (1982a)) and from (b) a 3-D model (taken from Sarmiento et al. (1992)).
response to the subtropical gyre. There is also some indication that the center of the gyre shifts northward due to the northward progression of deepest penetration for each isopycnal. It is important to note that significant tritium is found below this surface despite no direct atmospheric links having been established (Van Scoy et al., 1991). Van Scoy et al. also reported that tritium measured during the Long Lines cruise (1983–1985) penetrated 100–200 m deeper compared with the GEOSECS data (1973–74), particularly in the subtropical gyre and in the tropics.

The excess CO$_2$ vs salinity distribution (Fig. 4) for the 150°W section shows a scatter which changes its slope in the vicinity of $S = 34.5$. Less excess CO$_2$ is associated with waters with a salinity between 34.5 and 34.6. The shallow waters at salinities higher than approximately 34.6, or lower than approximately 34.4 are younger. These waters are believed to be coming from the Northwest Pacific where intermediate water formation occurs and where vertical mixing, mode water formation and thermocline ventilation appear to be most enhanced (Chen et al., 1986). The salinity where the deep water $\theta/S$ curves converge is near or less than 34.6. Deep waters with salinity higher than 34.6 do not contain excess CO$_2$.

The CO$_2$ Dynamics: Western North Pacific (WNP) cross-section (Fig. 5(a)) indicates that the excess CO$_2$ has penetrated to the deepest depth of approximately 1500 m ($\sigma_0 = 27.5$) at 25–30°N where the contours of the other physical and chemical properties also exhibit a downward dip (Chen et al., 1986).
Figure 5(a) shows a similar form as the ENP section but with a generally deeper penetration depth. The shoaling of the penetration from west to east is also seen in tritium distributions. The east-west vertical section along 35°N of tritium distribution (Fine et al., 1981) indicates that the 0.5 TU contour shoals from 900 m at 165°E to 600 m at 150°W. If the zero TU contour is assumed to be 100 m deeper than the 0.5 TU contour, then the penetration depth from the tritium data is 1000 m at 165°E and 700 m at 150°W. This result is consistent with our results for excess CO₂ (Figs. 2(a) and 5(a)).

The vertical section of tritium distribution has been produced by Fine et al. (1981) from selected GEOSECS stations between 15°–55°N between 165°W and 170°E. This section is located between our two sections. Similar to our excess CO₂ pattern, the 0.2 TU contour shows an overall concave structure with the deepest penetration at 900 m or deeper at 38°N, and the shallowest penetration at 400 m at 10°N. It is significant to note that the tritium data are from the GEOSECS expedition which occurred about ten years earlier than our ENP and WNP expeditions. The penetration depth obtained from the tritium data, therefore, may be shallower than today’s depth of penetration, since vertical diffusion and continued advection are still bringing the fossil fuel CO₂ signal downward over the last decade. An increasing penetration depth of tritium has been observed in several regions of the world oceans (Van Scoy et al., 1991). Gamo et al. (1987) also measured carbon-14 between 12°N and 30°N along approximately 170°E. They found that carbon-14 values have increased above the \( \sigma_\theta = 27.5 \) layer between 1973 and 1982. This increase indicates a rapid penetration of surface water, and hence excess CO₂, to such depth.

Recently Sarmiento et al. (1992) simulated the uptake of excess CO₂ by the ocean using a perturbation approach in a 3-dimensional global general circulation model. Figures 2(b) and 5(b)
Fig. 5. Cross-section of excess CO$_2$ along 165°E from (a) carbonate data and from (b) a 3-D model (taken from Sarmiento et al. (1992)).
show the comparison of the 3-D model-predicted excess CO$_2$ with estimations based on carbonate data along our two north-south sections. The 3-D model results in a penetration pattern similar to that obtained by the direct approach. The structure, however, is somewhat different. The carbonate-derived results show a deepening of the penetration depth at 30°N while this feature is lacking in the 3-D model. These differences may reflect problems in the 3-D model’s thermocline ventilation (Sarmiento et al., 1992).

5. Lower Boundary of Excess CO$_2$ Penetration in the Bering Sea

Figure 6 shows the estimated penetration depth in the North Pacific Ocean using all available data sets.

On the Bering Sea shelf the excess CO$_2$ penetration is limited by the depth of the shelf. In the Aleutian Basin the penetration is the deepest, to approximately 1000 m, in the eastern and southern regions. The penetration is slightly shallower off the Kamchatka Peninsula (Fig. 6).

Overall the Bering Sea contains $0.19 \pm 0.05 \times 10^{15}$ g excess carbon which is a small amount compared with $149 \times 10^{15}$ g of biospheric carbon released between 1600 and 1975 or with the $135 \times 10^{15}$ g of carbon generated by fossil fuel (Stuiver et al., 1984). Because of the small volume of the seawater in the Bering Sea, its capacity as an excess-CO$_2$ sink is limited (Chen, 1993). The carbonate deposits on the vast Bering Sea shelf, however, could provide a major sink for excess CO$_2$ in the near future. This will be discussed in the later section.

Fig. 6. Penetration depth of fossil fuel CO$_2$ in the North Pacific Ocean.

6. Lower Boundary of Excess CO$_2$ Penetration in the North Pacific Ocean

The overall pattern in Fig. 6 shows a similarity with major physical oceanographic features. The penetration depth is generally deeper for areas of high advection and is shallower for more stagnant regions or upwelling areas.

Several prominant features can be found: (1) A general shoaling trend from west to east is observed (Chen, 1987; Chen et al., 1986); (2) a deepening of penetration depth between 30 and 40°N which is coincident with the subtropical convergence; (3) a deep pool (to below 2000 m) at the western North Pacific off Japan, is observed. This is the region off Japan near the area of circulation of the North Pacific variety of the Subtropical Mode Water. The deepening is probably related to the interaction of Kuroshio and Oyashio currents. The mixing of these
currents enhances the penetration of fossil-fuel CO₂ into the ocean; (4) the penetration depth in the equatorial upwelling region is the shallowest in the North Pacific Ocean. The penetration depth is less than 300 m, especially in the eastern equatorial area.

The major difference between the Pacific and the Atlantic Ocean (Chen and Millero, 1979; Chen 1982b, 1987) is that excess CO₂ does not penetrate below the thermocline everywhere in the Pacific Ocean because there is no bottom water formation in the North Pacific. The shallowest penetration outside of the Southern Ocean occurs in the eastern equatorial region where the excess CO₂ only penetrates to 300 m, or shallower. Because few data are available for the complex oceanic region in the western equatorial Pacific, results there are less reliable. The general trend, however, indicates a deeper penetration (800 m) in the western Pacific and in the South China Sea. Overall, the excess CO₂ penetrates to a shallower depth in the equatorial Pacific than in the Atlantic, perhaps reflecting the higher equatorial upwelling rate in the Pacific and less influence of the newly formed water advected from the north.

These results agree qualitatively with the recent freons, tritium and C-14 data. Freons are found as deep as 1800 m at the western boundary, rising to about 900 m at 160°E and continuing

Fig. 7. Comparison of the excess CO₂ profiles based on carbonate data (dots) and those modeled using freon-11 data (thick lines; taken from Cline et al. (1985)).
to rise to 650 m at the eastern boundary along 47°N. Along 24°N, the freons penetrate to 900 m throughout most of the section but deepen to 1200 m at the western boundary (Warner and Weiss, 1985).

Freons data have also been used to quantitatively model the excess CO₂ penetration in the North Pacific Gyre (Cline et al., 1985). The results (Fig. 7) show that excess CO₂ signal derived from the freon-11 distribution agrees reasonably well with the signal calculated from the carbonate data. Figure 8 is an isogram map of the depth where tritium equals to 0.1 TU. The similarity in distribution with the excess CO₂ results is striking.

The preindustrial ΔC-14 concentration for low and mid-latitude waters in the Pacific Ocean is probably about −50‰, similar to the value in the Atlantic Ocean (Toggweiler et al., 1989). Figure 9 shows an isogram map of the depth where the ΔC-14 concentration equals to this value. The agreement with the excess CO₂ results is rather good between Equator and 30°N. At higher latitudes the agreement is poor because the subsurface waters in the Subarctic and Antarctic regions have a pre-industrial ΔC-14 concentration below −110‰ (Toggweiler et al., 1989; Chen and Rodman, 1990). Figure 10 shows an isogram map of the depth at which ΔC-14 equals −110‰.

Fig. 8. Isogram of the lower boundary of tritium penetration in the Pacific Ocean. Data taken from Ostlund et al. (1979), Fine et al. (1981), and Van Scoy et al. (1991).

Fig. 9. Isogram of the depth in the Pacific Ocean where the ΔC-14 concentration equals −50‰. Data taken from Ostlund et al. (1979) and Ostlund and Stuiver (1980).
The results north of 30° agree better with the excess CO$_2$ results shown in Fig. 6, but the excess CO$_2$ still penetrates much deeper than the bomb-produced ΔC-14 except in the equatorial region.

Overall the North Pacific (including the Bering Sea) contains $14.7 \pm 4 \times 10^{15}$ g excess carbon around 1980, with approximately 75% of the total occurring in the equatorial and subtropical regions below 40°N and the remaining 25% occurring in the subarctic region above 40°N.

7. Discussion

The existence of data and their use are fundamental to future progress. Higher resolution oceanic models must be built upon extensive sets of consistent oceanographic data. Few high quality carbonate data, however, exist in the Sea of Okhotsk, the Japan Sea, the Yellow Sea, the East China Sea, the South China Sea, and the Philippine Sea. Countries adjacent to these seas must collaborate in order to study these regions cost-effectively.

Winter data in the western Bering Sea near the Kamchatka Peninsula and in the Sea of Okhotsk are particularly important. The highest effective thermocline diffusion coefficient found anywhere in the North Pacific is in this region (Hansen et al., 1984), where an oxygen undersaturation as much as 20% probably occurs in winter (Chen, 1988). Strong winds coupled with strong cooling increase the gas solubility (causing the high degree of undersaturation) and induce intensive vertical mixing in the winter in the Sea of Okhotsk to enhance the penetration of excess CO$_2$. Subsequently the winter-formed subsurface water contributes to the formation of the North Pacific Intermediate Water which spreads out into a large portion of the North Pacific (Reid, 1973; Watanabe et al., 1991). Winter data in the formation region of the Intermediate Water is needed in order to provide the “preformed” concentrations which are needed for obtaining complete information on the excess CO$_2$ penetration in the North Pacific (Chen, 1985).

The 100% aragonite saturation horizon in the North Pacific Ocean corresponds to roughly the $\sigma_t=26.7$ surface (Chen et al., 1986, 1988). Figures 2(a), 3, 5(a) and 6 indicate that excess CO$_2$ has penetrated deeper than the aragonite saturation horizon everywhere in the North Pacific (Chen et al., 1986). Except in the equatorial region, the excess CO$_2$ has also penetrated deeper than the calcite saturation horizon (Chen et al., 1986). Continued increase of excess CO$_2$ in the oceans will cause upward migration of the saturation horizons and induce calcite and aragonite dissolution at a shallower depth (Feely and Chen, 1982; Chen, 1985), providing a sink for excess
CO₂. In this connection data from the Yellow Sea, and the East and the South China Seas are also important because the shallow bottoms of these seas are covered by CaCO₃ sediments. Water chemistry and sediment data on the shelf are needed in order to assess the potential of the shelf carbonate as an excess CO₂ sink.

The estimates of in situ calcium carbonate dissolution need to be verified by sediment trap studies. Seasonal variations of calcium carbonate fluxes need to be determined for subarctic, subtropical and equatorial regions.

Carbonate data in the Kuroshio formation region in the Philippine Sea are also needed in order to understand the carbonate dynamics in the most important current system in the Pacific Ocean. Effects of El Niño on the air/sea oxygen and CO₂ exchanges in the western North Pacific also need to be studied.

8. Conclusion
The penetration depth of fossil-fuel CO₂ in the North Pacific Ocean was constructed. The penetration depth is influenced not only by vertical diffusion and advection but also by horizontal advection. Pycnoclines also affect the penetration depth in the region. The deepest penetration depth is greater than 2000 m, which is found in the western North Pacific Ocean. The shallowest penetration depth is less than 300 m, which is found in the eastern equatorial region.

The excess CO₂ signal agrees qualitatively with the carbon-14, tritium and freons data. Some preliminary investigations also indicate a quantitative agreement with the freons data for stations in the Subarctic Gyre. Overall the North Pacific Ocean contains 14.7 ± 4 × 10¹⁵ g excess carbon around 1980.

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