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Edited by Yukihiro Nojiri

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National Institute for Environmental Studies**

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Preface

The first International Symposium, 'CO₂ in the Oceans' was held in Puerto Rico with the 6th CO₂ advisory panel meeting (UNESCO/IOC and SCOR/JGOFS) in January 1996. The symposium was hosted by University of Puerto Rico and organized by Prof. Franco Millero of University of Miami. It was sponsored by DOE (Department of Energy) and NOAA (National Oceanographic and Atmospheric Administration) of US government. It is needless to say the importance of ocean in the global circulation of carbon, or the fate of anthropogenically emitted carbon, to predict the effect of global warming for the near future global environmental change. It is one of the issues for the scientific aspect of Intergovernmental Panel on Climate Change. The Puerto Rico meeting was the first opportunity gathering the oceanic CO₂ researchers in the different fields of science in a same room for discussion. It was a very successful meeting to make global view about the oceanic role of the CO₂ circulation in the earth surface.

For the contribution to concentrate the recent advances of the oceanic CO₂ researches in the world, the National Institute for Environmental Studies made an arrangement for the 2nd international symposium in Tsukuba, Japan. The 8th CO₂ advisory panel meeting was also held conjunction with the Tsukuba symposium. The symposium was very successfully held with a large number of participants from the world and as well as in Japan. Encouraged by the success of the 2nd symposium, the CO₂ advisory panel is preparing the 3rd symposium in 2002.

The Center for Global Environmental Research (CGER) was established within the National Institute for Environmental Studies (NIES) in 1990. Our missions are to coordinate and promote research, information exchange, and database construction to address global environmental issues. CGER is also managing the long-term monitoring program about global environment, including greenhouse gases such as CO₂. The atmospheric CO₂ is monitored by ground-based stations, aircrafts, and commercial ships in the Pacific area. The atmosphere-ocean CO₂ exchange monitoring is continued by a Japan-Canada/US cargo ship. The recent results from these monitoring programs were presented in this symposium. The symposium brought great benefit to the communication of world's scientists in this field, and the Japanese activities of oceanic CO₂ studies were introduced.

The information presented in the papers in this volume will be of great benefit to the researchers on global CO₂ cycles. CGER will continue the effort to contribute the progress in the CO₂ studies relating to the global change.



Gen Inoue, D. Sc.

Director,

Center for Global Environmental Research,

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Editorial

At the symposium, many of participants had already submitted the extended abstracts and were printed as they submitted. Distribution of the extended abstract book at the symposium was very successful for the understanding of the works supplementing the short time of the presentation allocated.

In this volume, the revisions were accepted and style of the papers was uniformed for the ease of reading. Especially, this includes color figures of contoured map or informative graphs. I hope it is useful book for the oceanic CO₂ researchers to know the present status of the science in this field.

I wish to acknowledge the vital effort of Ms. Yuko Okabe, she was the symposium secretariat and also the management editor of this proceeding volume.

Yukihiro Nojiri, D. Sc.

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September 1999

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Opening Session

Welcome address

By Dr. Gen Ohi

Director General, the National Institute for Environmental Studies

Good morning. On behalf of the Environment Agency and the National Institute for Environment Studies (NIES), I would like to extend a welcome to all the participants of the 2nd International Symposium on CO₂ in the Oceans.

The Environment Agency was founded in 1972 when a societal concern peaked over the problems caused by classic environmental pollutants such as lead and mercury.

The NIES was established in the same year as its scientific research branch and its endeavor was directed primarily to the study of pollution problems originating in the 1960's and 70's, the time of high economic growth, as well as more basic researches. At that time, many of the research institutes in various scientific fields were relocated from Tokyo to Tsukuba. The NIES was one of the first institutes to be established in this area.

We recall that Tsukuba was then a quiet rural region encircling Mt. Tsukuba and featuring dark pine forests and patchy farms because the communal infrastructure was not developed yet. So the scientists who moved earlier in this region had the experiences only the first settlers in a new frontier can share with.

At this time of the year, you can enjoy from Mt. Tsukuba the panoramic view of the Kanto Plain, Mt. Fuji and the Nikko Mountain Range covered with incandescent snow. In early spring, plum blossoms are very beautiful. You see from there Mt. Fuji beyond the Tokyo Metropolitan area. Twenty years ago when air pollution was heavy, you could not see the mountain because of a gigantic dome of smog covering Tokyo. Now we see it with relative ease. It is particularly beautiful in winter since the skies are clearer.

It was in the 70's and early 80's when they started building research facilities here. That was toward the end of Japan's high economic growth and also the time when two oil crises hit Japan. Japan was alternately jolted by environmental problems caused by economic growth and by the demand for energy supply in quick succession. As a consequence, efforts for energy conservation were set up and serious health problems caused by environment pollution diminished. Because energy conservation was promoted by both industries and government, Japan's CO₂ emission per Gross Domestic Product (GDP) is one of the lowest in developed countries.

However, during our rapid economic growth consuming enormous, and discharging equally enormous amount of CO₂ and other global warming gases, relatively few Japanese realized the implication of those activities in relation to a global environment. Lately economists single-mindedly clamor of salvaging Japan from economic stagnation but eccentric environmentalists may be seeing brighter aspects of compromised human activities.

At the time of COP3, the Kyoto conference held in December 1997, Japan pledged to reduce CO₂ emission by 6% by 2010. It will be a painful task to achieve this goal and at the same time it is imperative for major industrialized countries to fulfill each of their promises. If our common goal of preventing irreversible man-made destruction of the global climate system, or more fundamentally, if those industrialized countries wish to gain moral ground

and to take leadership in this global enterprise in the relatively small space with limited resources, (i.e. the closed system like our earth) this is the time to learn the lesson that the earth can contain a maximum number of humans only under peaceful and symbiotic conditions. A system, which permits unrestrained greed to operate, creating only a few winners and many losers, will turn out unbearably destructive in the end.

The Environment Agency is responsible for both regional and global issues concerning human health and environment. Due to the increasing awareness of the importance of global issues, it was reorganized in 1990, installing the global environment department and its counterpart in the NIES. Accordingly, our research activities have expanded to encompass a variety of fields such as observation of the atmosphere and ocean, biodiversity, global environmental policy formulation, satellite monitoring for the stratosphere and land biosphere to name a few.

The Environment Agency has supported the activities of the International Geosphere and Biosphere Program (IGBP). The Agency hosts two symposiums in 1999: one is this CO₂ in the Oceans Symposium representing one of the IGBP co-projects, that is the Joint Global Ocean Flux Study (JGOFS), and the other is the IGBP congress which will be held in Kanagawa this May. The role of IGBP is to describe and understand the interactive physical, chemical and biological processes that regulate the total earth system, the changes that are occurring in this system, and the manner in which they are influenced by human activities.

Global warming is now emerging as one of the most serious environmental issues in the next century. To understand the role of the ocean in global CO₂ cycling is essential for the prediction of future climate changes because CO₂ is by far the major greenhouse gas. The CO₂ panel together with IGBP/JGOFS and UNESCO/IOC (Intergovernmental Oceanographic Commission) played an important role in the international collaboration in this field. The 1st Symposium regarding CO₂ in the Oceans was held in Puerto Rico just three years ago. Thanks to the efforts of Professor Frank Millero and others as a part of the panel activities, I understand that it was a quite success in gathering and exchanging recent scientific knowledge. It is great honor for me to sponsor the 2nd Symposium in Tsukuba. I hope the result of this symposium will be received with an equal satisfaction.

The Environment Agency also supports cooperative activities of Intergovernmental Panel on Climate Change (IPCC). Therefore, it is timely to hold this symposium in the sense that the body of scientific knowledge obtained here will enrich the IPCC report to be issued in the year 2000. At this symposium, you will make a handy review on the researches being conducted at representative institutions in Japan. Japan has continued ocean researches in cooperation with other countries and has a special interest in the Pacific because of obvious reasons.

During the past several years, our institute has conducted joint research with the Institute of Ocean Sciences Canada and had novel findings of considerable significance.

It is my sincere hope that through the lively exchange of new ideas and study results at this symposium, we will be able to contribute to the future progress in the sciences clarifying the relationship between the ocean and CO₂.

Thank you.

Opening address

Prof. Shizuo Tsunogai
Graduate School of Environmental Earth Sciences, Hokkaido University
Chair, Scientific Organizing Committee
2nd International Symposium on CO₂ in the Oceans

Ladies and gentlemen, I would like to address some words on behalf of the scientific committee of this symposium.

This is the 2nd International Symposium on the CO₂ in the Oceans. Now about 220 scientists from various countries are registered and more than 110 papers will be presented at this symposium.

The 1st CO₂ in the Oceans Symposium was held in 1996 in Puerto Rico, which was extremely successful. The symposium was realized during the discussion at the CO₂ panel. The CO₂ panel was first established by the CCCO (Committee on Climate Changes and the Ocean) of IOC (Intergovernmental Oceanographic Commission) and SCOR (Scientific Committee on Oceanic Research), and is now of IOC and JGOFS (Joint Global Ocean Flux Study) of IGBP (International Geosphere-Biosphere Programme: A Study of Global Change). The CCCO initiated many oceanic climate research programs such as WOCE (World Ocean Circulation Experiment) and disbanded in 1994. Among its activities the CO₂ panel is still ongoing. The first chair of the panel was Dr. Peter Brewer followed by Dr. Liliane Merlivat, and now Dr. Andrew Watson is the chair.

We, members of the organizing committee, tried to keep the style of the 1st symposium, namely all oral presentations and no parallel presentations. Unfortunately, we are forced to change partly the style adding a poster session due to such a large number of presentations. For the poster presentations, however, you can also talk for 1 minute at the lecture room. For each oral presentation the time is only 15 minutes including discussion. Please keep the time.

In this symposium, we consider all the aspects of CO₂ in the oceans and the results presented at this symposium will be used for the further development of our study in the oceans. Now an international program of SOLAS (Surface Ocean and Lower Atmosphere Studies) is being considered, and an IOC's program GOOS (Global Ocean Observing System) is also planned. Our results will be used for the realization of those programs. I also would like to make effort for these as a vice president of the SCOR.

I feel as compared to other international symposia held in Japan, the proportion of the presentations by Japanese scientists is smaller. I think this is due to the fact that there is no department of oceanography in Japan, where physical oceanographers are educated in the department of geophysics and the biological oceanographers are produced from the department of biology or fisheries. On the other hand, there is no department of

geochemistry. This makes our chemical oceanographers are a minority now in Japan. However, my supervisor, Professor Miyake started CO₂ work in Japan in 1940's. A group of Meteorological Research Institute succeeded the Professor Miyake's work.

I began my research work in 1960 and studied on calcium and alkalinity in seawater, but I did not study CO₂ itself until Dr. Peter Brewer invited me to the CO₂ panel in 1988. Attending the panel I was greatly surprised knowing the world level in this field. Then I have decided to raise the Japanese level in this field and increase the number of Japanese carbonate chemists. Of course, I myself started the oceanic CO₂ study, but I did not have to buy a coulometer, because I could not wait days for funding. I bought the first one with my own pocket money. The price was 3 million yen. The apparatus helped me to obtain my first results on total CO₂ in the western North Pacific.

I am now very happy, because Japanese young scientists in this field are growing. I think in the near future, especially through this symposium, they will be splendid scientists. I also would like to assist them as a president of the Oceanographic Society of Japan. I, however, must say our Japanese community is still a minority in the world and the level is not high. I always say to my colleagues as follows. "We are now climbing a mountain, of which top is the same as that for the world scientists. They are going ahead, but we Japanese scientists should not always follow them because there are many ways to its top. We must find a shortcut or a bypass." Although international cooperative works are very important, a high language barrier makes often it difficult. I think this symposium will be greatly stimulating them in providing contact with European and American scientists attending this symposium. So I would like to say my hearty thanks to the participants from various countries and attending this symposium.

Finally, I would like to say please enjoy this symposium and Japanese life during the stay for the participants outside Japan.

Thank you.

**18 (Mon) Morning
Observation 1**

Chaired by R. Feely and A. Murata

Net sea-air CO₂ flux over the global oceans: An improved estimate based on the sea-air pCO₂ difference

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Introduction

The distribution of the oceanic sink and source areas for atmospheric CO₂ and the magnitude of the net CO₂ flux across the sea surface are important for understanding the global carbon cycle. Based on approximately 2.5 million measurements made for the pCO₂ in surface waters of the global ocean since 1960, the climatological distributions of monthly sea-air pCO₂ difference, ΔpCO₂, and the net sea-air flux have been estimated for a reference year of 1995. The method used and the results obtained are presented.

Method

For this study, only the ocean water pCO₂ values measured using direct gas-seawater equilibration methods are used. Observations made in the equatorial Pacific between 10°N and 10°S during El Niño events have been excluded from the data set. Thus, the results shown in this paper represent the climatological distributions under non-El Niño conditions. Since the measurements were made in different years, during which the atmospheric pCO₂ was increasing, they were corrected to a single reference year (arbitrary chosen to be 1995) on the basis of the following assumptions. Since surface waters in subtropical gyres mix vertically at slow rates with subsurface waters due to the presence of strong stratification at the base of the mixed layer and have a long time to take up atmospheric CO₂, their CO₂ chemistry tends to follow the atmospheric CO₂ increase. Therefore, the pCO₂ in these warm waters follows the increasing trend of atmospheric CO₂, and hence the sea-air pCO₂ difference tends to be independent of the year of measurements. On the other hand, since surface waters in high latitude regions are replaced by winter convection with upwelling deep waters yearly, their CO₂ properties tend to remain unchanged from year to year reflecting those of deep waters, in which the effect of increased atmospheric CO₂ is diluted to undetectable levels. Accordingly, the sea-air pCO₂ difference measured in a given year was corrected to the

reference year using the observed increase in the atmospheric CO₂ concentration. These adjusted values are binned into a total of 750,000 pixels (72 pixels along the longitude × 41 along the latitude × 365 days), which represent the global 4°×5° grid for each day in a single virtual calendar year.

Mean monthly global distributions of $\Delta p\text{CO}_2$ have been constructed using an interpolation method based on a lateral 2-dimensional advection-diffusion transport equation [Takahashi *et al.*, 1995; Takahashi *et al.*, 1997]. The equation yields $\Delta p\text{CO}_2$ values for 4°×5° pixels where no observations exist, while it satisfies the observed values explicitly. The effects of vertical mixing and sea-air CO₂ flux are considered inherently imbedded in the observed data. Therefore, the short-term behavior of surface water properties may be approximated using the lateral transport model without vertical mixing and gas exchange terms. For advective transport, the mean monthly surface flow field of *Bryan and Lewis* [1979] is used and, for diffusive transport, a constant value of 2000 m²/sec. The equation has been solved iteratively using a finite-difference algorithm, in which the computational domains are joined at December 31, 1995, with January 1, 1995, and along the prime meridian to ensure continuity in time and space. The ocean-land boundaries are assumed to be reflective boundaries. Singularities at the poles are avoided by the presence of the Antarctic continent in the south and the polar ice cap in the north, which was assumed to be a landmass. Typically, several thousand iterations are necessary before solutions are converged. Although the solutions give daily distributions, monthly mean distributions have been computed and used for flux calculations.

The net sea-air CO₂ flux in each pixel has been computed using the monthly mean $\Delta p\text{CO}_2$ values thus obtained, and the *Wanninkhof* [1992] formulation (Eq. (1)) for the effect of wind speed on the CO₂ gas transfer coefficient with the mean monthly wind speed of *Esbensen and Kushnir* [1981]. The combination of the Wanninkhof relation and *Esbensen & Kushnir* wind data yields a mean global gas transfer rate of 0.063 mole CO₂/m²/μatm/yr. This is consistent with 19 moles CO₂/m²/yr (= 0.064 mole CO₂/m²/μatm/yr) estimated on the basis of carbon-14 distribution in the atmosphere and oceans [Broecker *et al.*, 1986]. The distribution maps of $\Delta p\text{CO}_2$ during February and August, 1995, over the global oceans are shown in Figure 1a and b, and a map for the mean annual net CO₂ flux across the sea surface during 1995 is shown in Figure 2.

Results

The $\Delta p\text{CO}_2$ maps (Figure. 1) show that oceanic sources for atmospheric CO₂ (indicated by red and yellow colors) are located in the areas of deep water upwelling. The effect of winter upwelling is seen in the sub-arctic western Pacific (Figure 1a) and that of upwelling induced by the southwest monsoon during July-August is seen in the Persian Gulf (Figure 1b). The strong CO₂ source zone located along the Pacific equatorial belt is supported by the coastal upwelling along South America as well as by the upward entrainment of the equatorial undercurrent water. The source intensity is reduced toward the western Pacific due mainly to CO₂ losses to photosynthesis and to the atmosphere. Strong CO₂ sinks (blue and purple areas) are seen along the pole-ward edges of subtropical gyres, where major warm currents are located. The Gulf Stream in the North Atlantic and the Kuroshio in the North Pacific are both major CO₂ sinks (see Figure 2) due primarily to cooling as they flow from

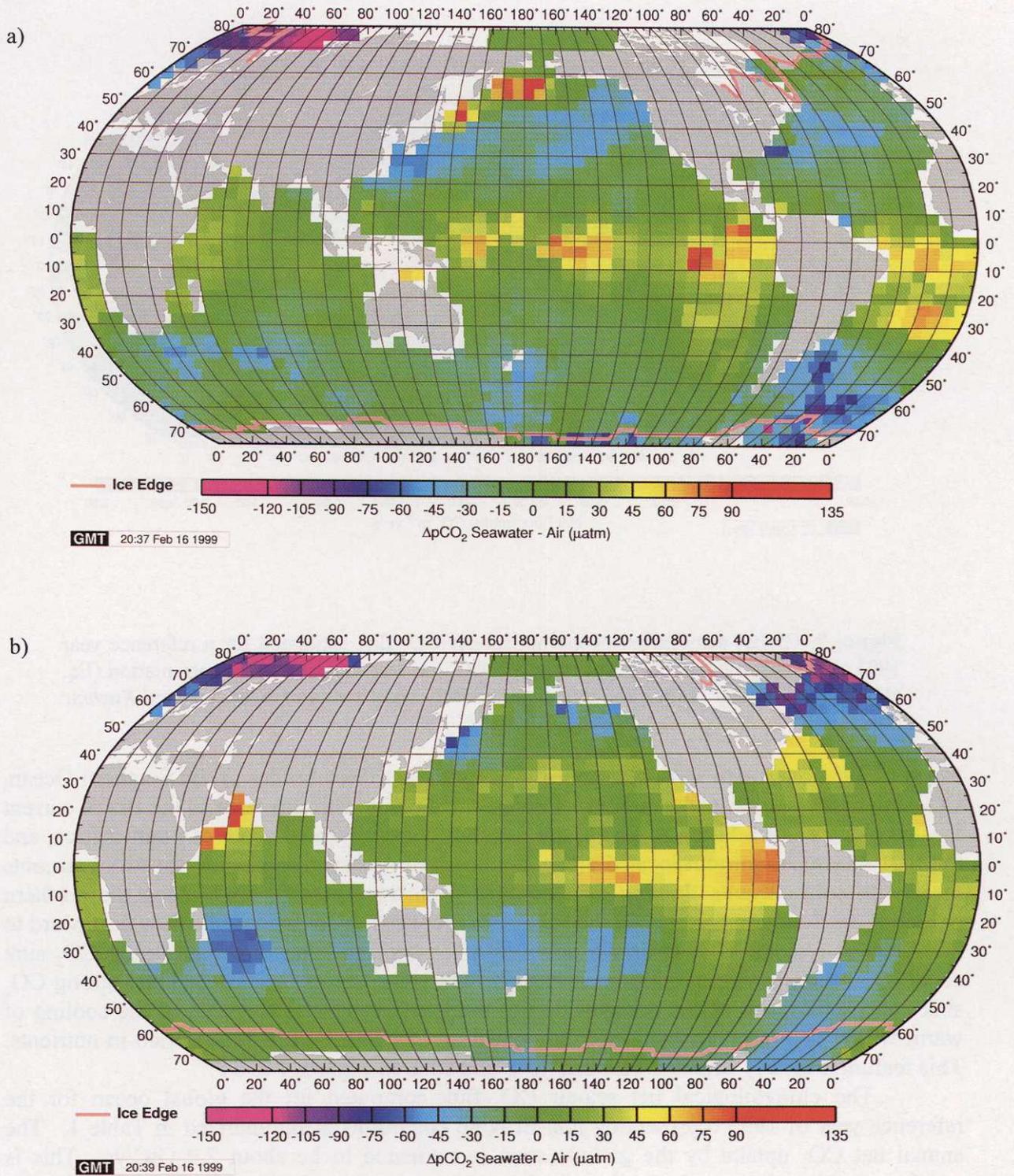


Figure 1. Global distribution of the sea-air pCO₂ difference estimated for: a) February, 1995 and b) August, 1995. Pink lines indicate the edge of ice fields.

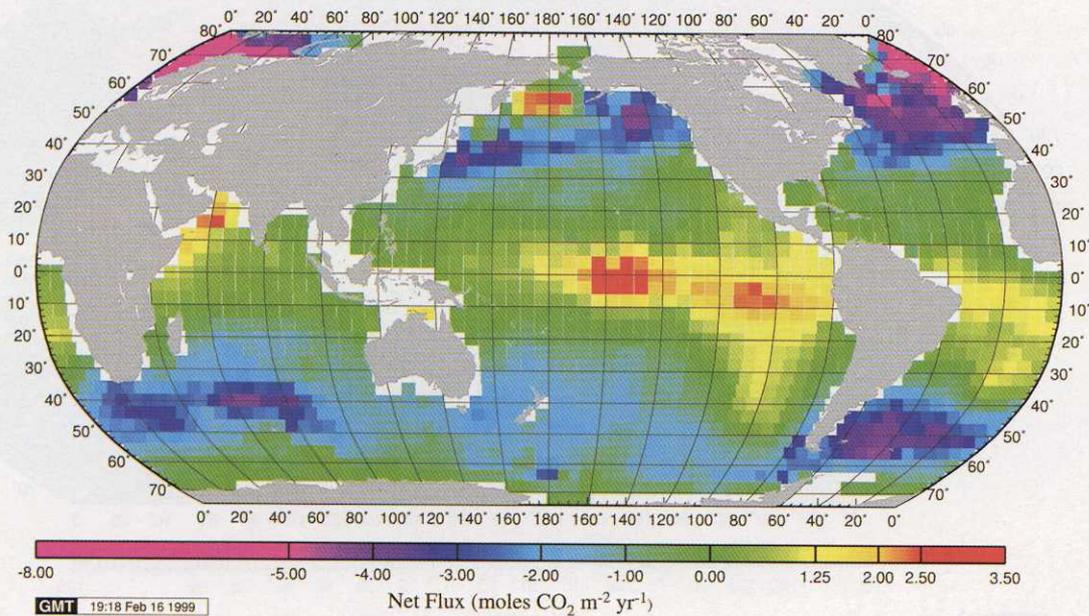


Figure 2. Global distribution of the net sea-air CO₂ flux estimated for a reference year 1995 using the effect of wind speed on the CO₂ gas transfer coefficient formulation (Eq. (1)) by Wanninkhof [1992] and the mean monthly wind speed of Esbensen and Kushnir [1981].

warm tropical oceans to sub-polar zones. Along the northern border of the Southern Ocean, CO₂ sink areas caused by the cooling of pole-ward flowing currents such as the Brazil current located along eastern South America, the Agulhus current located south of South Africa, and the East Australian current located along southeastern Australia. These warm water currents meet with cold currents flowing equator-ward from the Antarctic zone along the northern border of the Southern Ocean. As the sub-Antarctic waters rich in nutrients flow northward to more sun-lit regions, CO₂ is drawn down by photosynthesis thus creating strong CO₂ sink conditions. Confluence of subtropical waters with polar waters forms broad and strong CO₂ sink zones as a result of the juxtaposition of the lowering effects on pCO₂ of the cooling of warm waters and the photosynthetic drawdown of CO₂ in sub-polar waters rich in nutrients. This feature is clearly depicted between 40°S and 55°S in Figures 1 and 2.

The climatological net sea-air CO₂ flux computed for the global ocean for the reference year of 1995 representing non-El Niño conditions is summarized in Table 1. The annual net CO₂ uptake by the global ocean is estimated to be about 2.2 GtC/yr. This is consistent with estimates obtained on the basis of ocean-atmosphere models (calibrated using carbon-14 distributions) with constant biology. If the effects of wind speed on the gas transfer coefficient of Liss and Merlivat [1986] is used instead, the global uptake flux of 1.1 GtC/yr is obtained.

Table 1. The net sea-air flux of CO₂ estimated for a reference year of 1995 using the effect of wind speed on CO₂ gas transfer coefficient (Eq. (1)) of *Wanninkhof* [1992] and the monthly wind field of *Esbensen and Kushnir* [1981]. The 1990 fluxes [*Takahashi et al.*, 1997] have been corrected to the same gas transfer coefficient and wind field and compared with the 1995 values. The positive values indicate sea-to-air fluxes, and the negative values, the air-to-sea fluxes. (95) and (90) indicate the flux values estimated for the reference years 1995 and 1990 respectively.

Latitudes		Pacific Ocean	Atlantic Ocean	Indian Ocean	Southern Ocean	Global Oceans
		Sea-air Flux in 10 ¹⁵ grams Carbon / year				
N. of 50°N	(95)	-0.03	-0.45	-	-	-0.48
	(90)	+0.00	-0.45	-	-	-0.45
50°N-14°N	(95)	-0.48	-0.29	+0.03	-	-0.74
	(90)	-0.39	-0.32	+0.02	-	-0.69
14°N-14°S	(95)	+0.62	+0.12	+0.09	-	+0.83
	(90)	+0.68	+0.10	+0.12	-	+0.90
14°S-50°S	(95)	-0.34	-0.22	-0.60	-	-1.17
	(90)	-0.32	-0.20	-0.38	-	-0.90
S. of 50°S	(95)	-	-	-	-0.62	-0.62
	(90)	-	-	-	-0.31	-0.31
TOTAL	(95)	-0.23	-0.84	-0.48	-0.62	-2.17
	(90)	-0.04	-0.87	-0.23	-0.31	-1.45
% UPTAKE	(95)	11%	39%	22%	29%	100%
	(90)	2%	60%	16%	22%	100%
AREA (10 ⁶ km ²)		151.6	72.7	53.2	31.68	309.12
AREA (%)		49.0%	23.5%	17.2%	10.2%	100%

The 1995 estimate for the global ocean uptake flux is about 0.7 GtC/yr greater than the 1990 estimate. This difference is partly the result of improvements in observational database and partly the effect of an increase in the atmospheric pCO₂ of about 7 μatm that occurred from 1990 to 1995. An increase of about 0.5 GtC/yr uptake by the Southern Ocean and the South Indian Ocean may be attributed to the improved database and to a -7 μatm change in ΔpCO₂ over the southern high latitude oceans caused by the increase in atmospheric pCO₂. If a mean ΔpCO₂ over the global high latitude oceans pole-ward of 50° latitude were lowered by 7 μatm (i.e. ocean became a stronger sink), an additional uptake of 0.3 GtC/yr would be expected.

The uptake flux for the northern hemisphere oceans (north of 14°N) is 1.22 GtC/yr, whereas that for the southern hemisphere oceans (south of 14°S) is 1.79 GtC/yr. Thus, the southern hemisphere oceans are a stronger CO₂ sink by about 0.5 GtC/yr. This is due partially to the much greater oceanic areas in the southern hemisphere and partially to that the

Southern Ocean south of 50°S is an efficient CO₂ sink while it has 10% of the global ocean area, it takes up about 29% of the global ocean CO₂ uptake. Cold temperature and moderate photosynthesis are both responsible for the large uptake by the Southern Ocean.

The 1995 flux values listed in Table 1 shows that the Atlantic Ocean is the largest net sink for atmospheric CO₂ (39%); the Southern Ocean (22%) and the Indian Ocean (22%) the next; and the Pacific (11%) the smallest. The large sink flux of the northern oceanic areas is attributed to the intense biological drawdown of CO₂ in the high latitude areas of the North Atlantic and arctic seas during the summer months. This is also due to low CO₂ concentrations in upwelling deep waters, which are, in turn, caused primarily by the short residence time of the North Atlantic Deep Waters. The small uptake flux of the Pacific can be attributed to the combined sink flux of the northern and southern subtropical gyres, and is roughly balanced by the source flux from the equatorial Pacific. The equatorial Pacific CO₂ source flux may be totally or partly eliminated during El Niño events. This effect alone could increase the global ocean uptake flux up to 0.6 GtC/yr during an El Niño year.

Error estimates

The sea-air CO₂ flux values reported in this paper are subject to two sources of errors: 1) biases in $\Delta p\text{CO}_2$ values interpolated from relatively sparse observations, and 2) errors due to uncertainties in the gas transfer coefficient estimated on the basis of the wind speed dependence.

Possible biases in $\Delta p\text{CO}_2$ differences have been estimated by *Takahashi et al.* [1997] using sea surface water temperatures (SST) as a proxy. The monthly SST in each pixel was computed using the SST values measured concurrently with $p\text{CO}_2$ and the same interpolation scheme used for obtaining the global distribution of $\Delta p\text{CO}_2$. This was compared with the climatological SST value obtained by *Shae et al.* [1992], and the global mean of the differences was computed. The global mean SST estimated on the basis of our data and method has been found to be about 0.4°C greater than the climatological mean estimated by *Shae et al.* [see *Takahashi et al.*, 1997]. Using the mean effect of SST on seawater $p\text{CO}_2$ observed over the global ocean (3.5% per°C), we estimate that the SST difference corresponds to about 5 μatm or 50% error in $\Delta p\text{CO}_2$. Therefore, the estimated global sea-air CO₂ flux is subject to a systematic error of up to 50%.

The reliability of the wind speed dependence on the CO₂ gas transfer coefficient has been significantly improved as a result of the recent GASEX98 study reported by W. McGillis, J. Edson and R. Wanninkhof during this symposium. Eq. (1) of Wanninkhof used in the present study is consistent within about $\pm 20\%$ with their new eddy correlation flux measurements conducted over the North Atlantic. Hence, the estimated fluxes are subject to this level of uncertainty.

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