

1995 ACTIVITIES

INTRODUCTION

Clarifying the Earth's chemical environment is an important objective of the Geochemical Research Department (GRD). GRD scientists have studied the geochemical cycle of chemical substances found in the terrestrial environment – the atmosphere, the hydrosphere, and the lithosphere – since the 1940s.

With increasing human activity and the introduction of anthropogenic materials, the terrestrial chemical environment has been changing rapidly. Given the scientific and public considerations involved, our recent studies have been focused on obtaining precise, accurate information on the following subjects: temporal and spatial variation of greenhouse gases such as CO₂ in the atmosphere, carbon and nitrogen cycles in the marine environment, the exchange rate of chemical substances between the ocean and atmosphere as well as the biosphere and atmosphere, radioactive contamination of the ocean and atmosphere, and the behavior of natural and artificial chemicals in the air and ocean.

We are involved in many scientific projects related to large national and international programs such as the Japanese Study on the Radioactivity in the Environment (JASRE), Japanese Study on the Behavior of Greenhouse Gases and Aerosols (JASBEGGA), World Climate Research Program (WCRP)/World Ocean Circulation Experiment (WOCE), International Geosphere-Biosphere Program (IGBP)/Joint Global Ocean Flux Study (JGOFS) and International Global Atmospheric Chemistry Program (IGBP/IGAC).

At yearend, the department had 10 scientific staffers.

International/National scientific programs and budgetary funds for our studies are shown in Table 1.

GREENHOUSE GAS (CO₂)

1. Exchange of Chemical Substances Between Atmosphere and Ocean

1.1 Long-term trends in carbon dioxide partial pressure (pCO₂) in western North Pacific surface waters

Inoue, Matsueda, Fushimi, Hirota, Asanuma, and Takasugi (1995)

Carbon dioxide in the atmosphere appears to play a major role in determining the earth's climate and habitability through its regulation of solar radiation balance. Atmospheric CO₂ levels have been increasing since the Industrial Revolution, mainly due to anthropogenic of CO₂ emissions.

Open oceans have been regarded as an important "sink" for processing anthropogenic CO₂, but few measurements have confirmed oceanic CO₂ uptake. Inoue *et al.* (1995) studied the long-term trends of carbon dioxide partial pressure (pCO₂) in surface waters of the western North Pacific, analyzing data in the western North Pacific and the overlying air measured regularly on board the Japan Meteorological Agency (JMA) research vessel "Ryofu Maru" (Fig. 95-1).

pCO₂ in surface seawater (pCO₂^{sea}) observed every boreal winter from 1984 to 1993 gives a growth rate of $1.8 \pm 0.6 \mu\text{atm yr}^{-1}$ north of 15°N and $0.5 \pm 0.7 \mu\text{atm yr}^{-1}$ south of 14°N, averaging $1.2 \pm 0.9 \mu\text{atm yr}^{-1}$. The rate

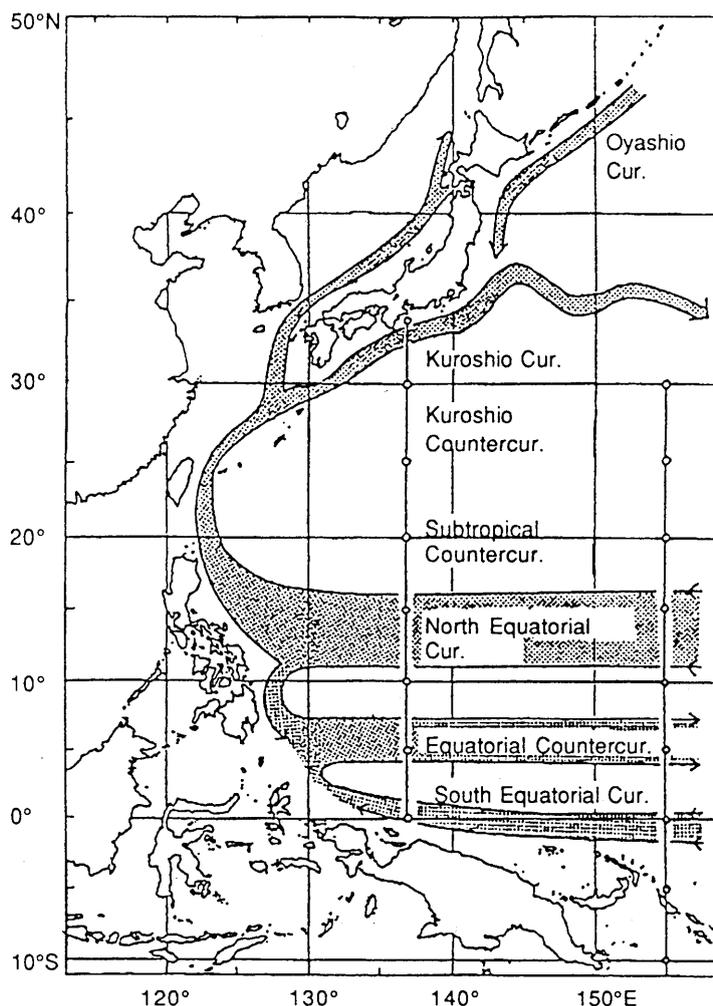


Fig. 95-1 Repeated transects where $p\text{CO}_2^{\text{sea}}$ measurements were made since 1981. Open circles: oceanographic stations, where seawater samples from surface to the bottom were taken by CTD casts for measurement of oxygen, TCO_2 , nutrients, salinity, etc. South of 32°N along 137°E , oceanographic stations were located every 1° and CTD casts to 1000 (or 2000) dbar were obtained. Typical major western North Pacific currents patterns are indicated.

of $p\text{CO}_2^{\text{sea}}$ increase north of 15°N equals that of atmospheric CO_2 ($1.8 \mu\text{atm yr}^{-1}$) during the same period although that south of 14°N is lower (Fig. 95-2).

They estimated the annual CO_2 flux between the sea and air based on monthly mean $\Delta p\text{CO}_2$ and the gas transfer coefficient calculated according to Liss and Merlivat (1986), Tans *et al.* (1990), and Wanninkhof (1992). Large CO_2 influx occurred off the coast of Japan ($-16 \text{ mmol m}^{-2}\text{day}^{-1}$, based on Tans *et al.*, 1990) because of the large negative $\Delta p\text{CO}_2$ ($-60 \mu\text{atm}$) and strong wind during winter. In the sink area, the annual mean air-sea CO_2 flux ranged from $-8 \text{ mmol m}^{-2}\text{day}^{-1}$ at 31°N to $-1 \text{ mmol m}^{-2}\text{day}^{-1}$ at 11°N .

The $p\text{CO}_2^{\text{sea}}$ increase rate is therefore high where the ocean acts as a strong sink for atmospheric CO_2 . South of 10°N , the ocean acts as a source ($0.2\text{--}0.7 \text{ mmol m}^{-2}\text{day}^{-1}$), but CO_2 evasion flux is considerably smaller than in the central and eastern equatorial Pacific.

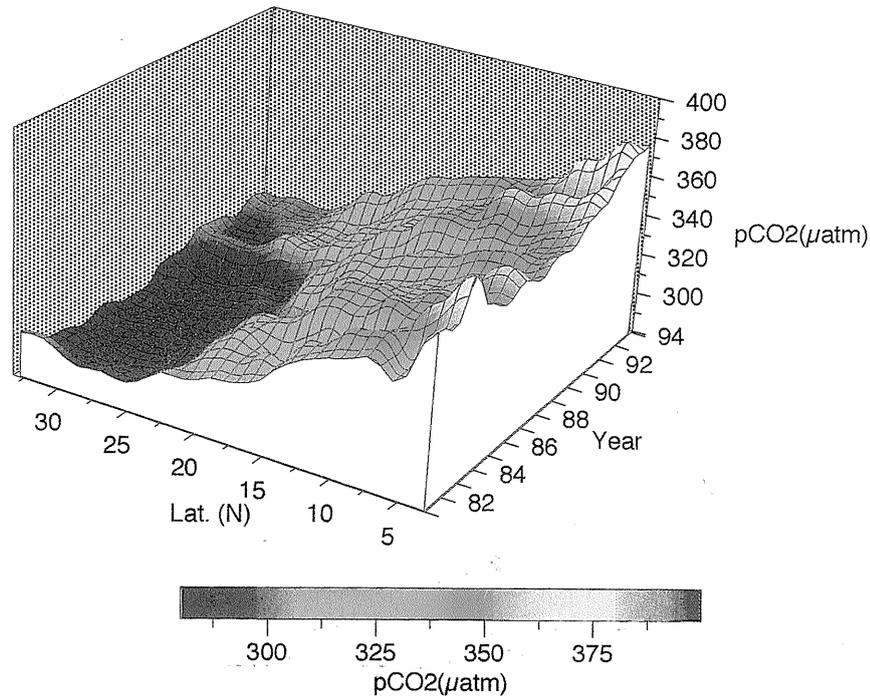


Fig. 95-2 Temporal variations in $p\text{CO}_2$ along 137°E observed in late January.

1.2 Air-sea CO_2 exchange in central and western equatorial Pacific

Ishii and Inoue (1995)

Ishii and Inoue (1995) studied the air-sea exchange of CO_2 in the central and western equatorial Pacific. They made measurements of CO_2 in marine boundary air and in surface seawater of the central and western Pacific west of 150°W from September to December 1990 (Fig. 95-3). They observed a steep decrease in $p\text{CO}_2^{\text{sea}}$ from $400 \mu\text{atm}$ to $350 \mu\text{atm}$ between 179°E and 170°E along with a decrease in salinity (Fig. 95-4). West of 170°E , where salinity is low due to heavy rainfall, $p\text{CO}_2^{\text{sea}}$ was nearly equal to $p\text{CO}_2^{\text{air}}$. They evaluated the net CO_2 flux from the sea to the atmosphere in the region (15°S - 10°N , 140°E - 150°W) from the $p\text{CO}_2$ distribution and several gas transfer coefficients reported so far (Table 95-1). It ranged from $0.13 \text{ GtC year}^{-1}$ to $0.29 \text{ GtC year}^{-1}$. This CO_2 evasion flux is thought to almost disappear during an El Niño event.

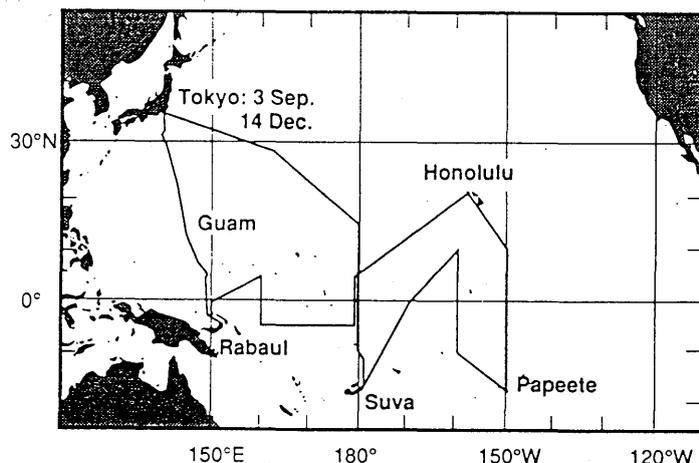


Fig. 95-3 Tracks of KH-90-2 and KH-90-3 of the R.V. *Hakuho Maru*. KH-90-2 is from Tokyo (September 3, 1990) to Honolulu (Hawaii) (October 25, 1990) via Suva (Fiji) and Papeete (Tahiti). KH-90-3 is from Honolulu (October 31, 1990) to Tokyo (December 14, 1990) via Rabaul (Papua New Guinea) and Guam.

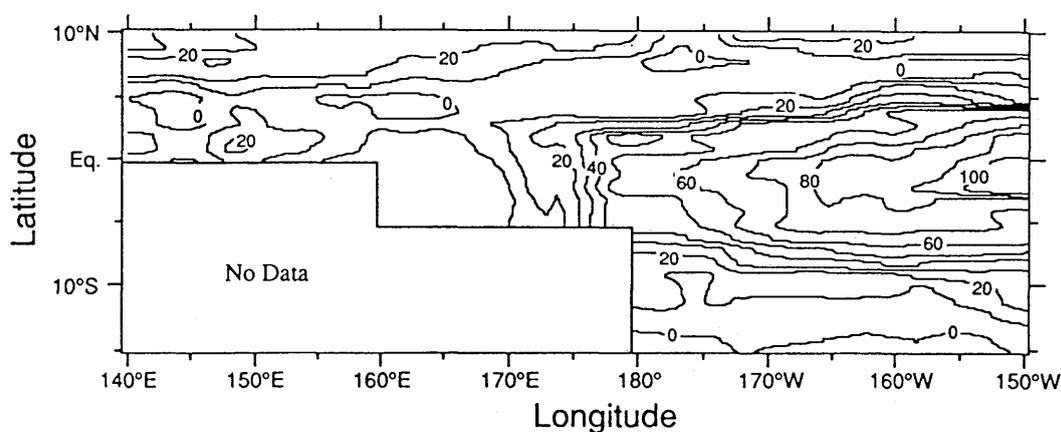


Fig. 95-4 Distribution of $\Delta p\text{CO}_2$ in central and western equatorial Pacific in September 1990.

Table 95-1 CO_2 flux (MtC) in the central and western equatorial Pacific in September-November 1990

Eq.	Flux (15°S-1°S) (MtC)	Flux (0°-10°N) (MtC)		Total flux (MtC)
<i>(Flux accumulated twice-daily flux over a month)</i>				
	GANAL	GANAL		GANAL
Liss and Merlivat	19.5	13.1		32.6
Wanninkhof (short)	30.2	21.1		51.3
Wanninkhof (short, chem)	36.6	26.8		63.4
Wanninkhof (long)	38.0	26.5		64.5
Tans et al.	42.3	29.0		71.3
<i>(Flux calculated by using monthly mean wind speed)</i>				
	GANAL	GANAL	WSCLI	GANAL
Liss and Merlivat	19.5	12.9	11.4	32.4
Wanninkhof (short)	28.9	19.8	18.5	48.7
Wanninkhof (short, chem)	35.4	25.6	24.4	61.0
Wanninkhof (long)	36.4	24.9	23.3	61.3
Tans et al.	42.2	29.0	25.7	71.2

Table 95-2 DIC ($\mu\text{mol} \cdot \text{dm}^{-3}$) of natural water standards prepared with lake or sea water as measured by both the TOC-5000 and coulometer. Lake standards 1 and 2 were prepared from surface water collected at station 9 in the middle of Lake Kasumigaura in November 1993 and April 1994, respectively. Sea water standard 1 was prepared from surface water collected from the Western North Pacific at 20°N, 130°E in February 1992. Sea water standard 2 was prepared from a mix of surface waters collected from the Western North Pacific at 20°N, 137°E in January 1993 and at 19°N, 137°E in January 1994.

standard	instrument	mean [DIC]	std. dev.	bottles	reps/bottle
Lake 1	TOC-5000	997.1	4.8	2	6 & 10
Lake 1	Coulometer	994.2	0.6	4	1
Lake 2	TOC-5000	1013.9	3.0	5	4 to 8
Lake 2	Coulometer	1011.2	0.7	3	1
Sea 1	TOC-5000	2028.3	2.2	2	10
Sea 1	Coulometer	2026.5	0.6	4	1
Sea 2	TOC-5000	2048.4	6.2	3	6 to 16
Sea 2	Coulometer	2053.1	2.0	2	1 or 2

1.3 Methods for dissolved inorganic carbon measurement in natural waters

Weisburd, Ishii, Fukushima, and Otsuki (1995)

Knowing the concentrations of dissolved inorganic carbon (DIC) in natural waters is important in studies of ecology and biogeochemistry. The available measurement methods remain relatively difficult and imprecise. Weisburd *et al.* (1995) demonstrated that a commercially available, nondispersive infrared (ND-IR) organic carbon analyzer could provide relatively accurate, precise ($\pm 2-3 \mu\text{mol dm}^{-3}$) DIC measurements (Table 95-2). This precision, though less than the $0.5-1 \mu\text{mol dm}^{-3}$ for the state-of-the-art instrument (CO_2 Coulometer), is adequate for many applications and is easier to use. For DIC transects and profiles in systems with moderate to large DIC variations or for discrete DIC productivity measurements, the TOC-5000's precision is generally sufficient. The precision of the coulometer remains necessary, however, for marine DIC surveys.

2. Chemical Substances Exchange Between Atmosphere and Biosphere

2.1 Atmospheric CO_2 variations at Meteorological Research Institute, Tsukuba, Japan

Inoue and Matsueda (1996)

The CO_2 exchange between the air and biosphere is an important process controlling the level of atmospheric CO_2 on a time scale of hours to decades. To get a better understanding of CO_2 exchange between the air and terrestrial biosphere, measurements of the CO_2 concentration in the surface air have been conducted at the Meteorological Research Institute (MRI, 36°04'N, 147°07'E, 25 m above sea level) in Tsukuba, 50 km northeast of Tokyo, Japan, since April 1986 (Fig.95-5).

Inoue and Matsueda (1996) began continuous measurements of CO_2 in the air 1.5 m above the earth's surface in April 1986 and studied variations in atmospheric CO_2 at the Meteorological Research Institute, Tsukuba, Japan (Fig. 95-6). At the end of February 1992, they added measurements of atmospheric CO_2 at 200 m on a meteorological tower, 213 m high, 250 m from the 1.5 m sampling site. They selected CO_2 data observed at MRI in daytime with hour-to-hour variation less than 1 ppmv and compared data with that of Ryori (39°02'N, 141°50'E), a continental station operated by the JMA. They showed that the selected CO_2 record provided a representative CO_2 level in surface air on spatial scales of at least a few hundred kilometers.

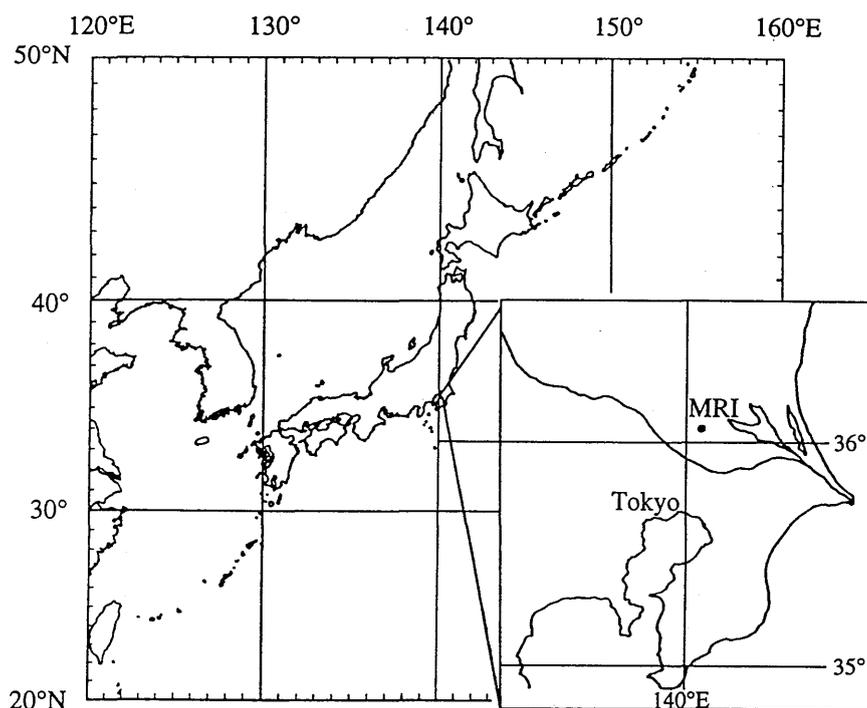


Fig. 95-5 MRI atmospheric CO₂ sampling site.

Reprinted from *J. Atmospher. Chem.*, **23** (1996), 137-161, Variations in atmospheric CO₂ at the Meteorological Research Institute, Tsukuba, Japan, Inoue and Matsueda, fig. 1 (©1996 Kluwer Academic Publishers. Printed in the Netherlands.) with kind permission from Kluwer Academic Publishers.

They also studied the interannual changes in photosynthesis/respiration against changes in climatological parameters. They found that respiration was sensitive to the temperature change within a small temperature anomaly (ca. 1°C), while photosynthesis was less sensitive. When the temperature anomaly is large, however, photosynthesis and respiration tend to be competitive. The decrease in respiration due to cooling (-0.7°C) could play an important role in determining the 1992 CO₂ level, which did not increase compared to that of 1991 in the Northern Hemisphere.

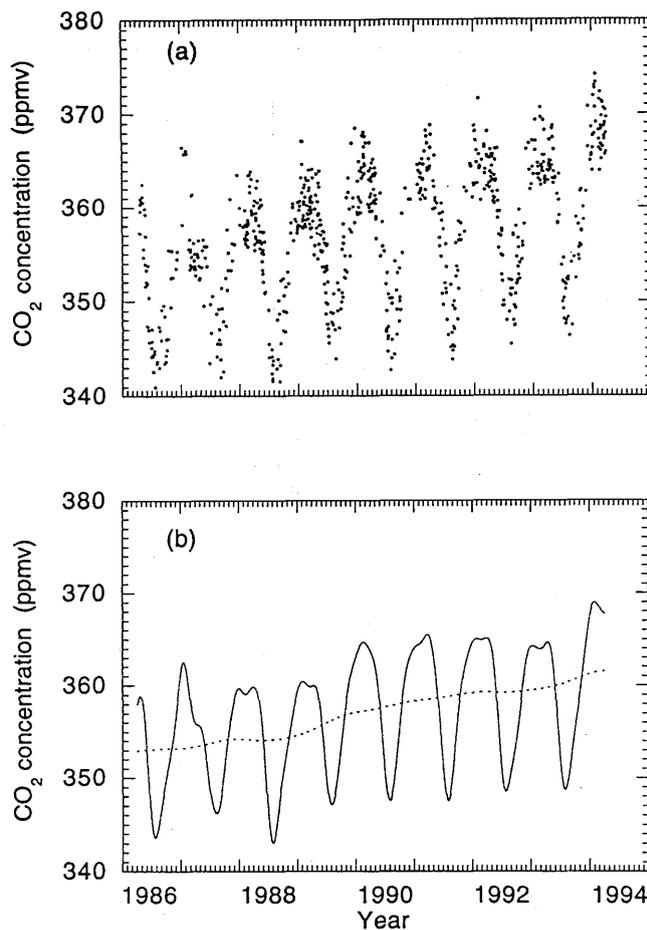


Fig. 95-6 Temporal variations in daily average when hour-to-hour changes are less than 1 ppmv (Cp) between 11:00 JST and 16:00 JST. (a) displays all raw data from April 13, 1986, to March 31, 1994. The solid curve in (b) is for a low-pass filter with cutoff at 100 days and the dashed curve is for cutoff at 667 days. Reprinted from *J. Atmospher. Chem.*, **23** (1996), 137-161, Variations in atmospheric CO₂ at the Meteorological Research Institute, Tsukuba, Japan, Inoue and Matsueda, fig. 7 (©1996 Kluwer Academic Publishers. Printed in the Netherlands.) with kind permission from Kluwer Academic Publishers.

RADIOACTIVE NUCLIDES (⁹⁰Sr, ¹³⁷Cs, and Pu Isotopes)

3. Radioactive Materials in Air, Fallout, Rainwater, and Seawater

Global radioactive contamination derived from the fallout of nuclear weapon tests, waste disposal, or accidents at nuclear facilities has brought about scientific and social consequences. To evaluate the feedback effect on society and to trace the fate in the global environment, meteorological and oceanographic studies on the environmental radioactivity have been conducted for the last 40 years.