

Air-Sea CO₂ Flux by Eddy Covariance Technique in the Equatorial Indian Ocean

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(Received 21 August 2006; in revised form 16 November 2006; accepted 14 January 2007)

Direct measurements of the air-sea CO₂ flux by the eddy covariance technique were carried out in the equatorial Indian Ocean. The turbulent flux observation system was installed at the top of the foremast of the R/V MIRAI, thus minimizing dynamical and thermal effects of the ship body. During the turbulent flux runs around the two stations, the vessel was steered into the wind at constant speed. The power spectra of the temperature or water vapor density fluctuations followed the Kolmogorov -5/3 power law, although that of the CO₂ density fluctuation showed white noise in the high frequency range. However, the cospectrum of the vertical wind velocity and CO₂ density was well matched with those of the vertical velocity and temperature or water vapor density in this frequency range, and the CO₂ white noise did not influence the CO₂ flux. The raw CO₂ fluxes due to the turbulent transport showed a sink from the air to the ocean, and had almost the same value as the source CO₂ fluxes due to the mean vertical flow, corrected by the sensible and latent heat fluxes (called the Webb correction). The total CO₂ fluxes including the Webb correction terms showed a source from the ocean to the air, and were larger than the bulk CO₂ fluxes estimated using the gas transfer velocity by mass balance techniques.

Keywords:

- Eddy covariance technique,
- eddy correlation technique,
- CO₂ flux,
- air-sea interaction,
- Webb correction,
- spectral analysis.

1. Introduction

The ocean is one of the main sinks of anthropogenic CO₂. Precise measurements of the CO₂ gas transfer across the air-sea interface provide a better understanding of the global carbon cycle. However, many of the mechanisms that control the air-sea gas transfer are not well understood.

The eddy covariance (correlation) technique is the only direct measurement of the momentum, heat, and trace gas (e.g. water vapor, CO₂, and DMS) fluxes across the air-sea interface. This technique can evaluate the trace gas flux on a shorter time scale (e.g. several minutes or hours) than mass balance techniques using the tracers of the natural and bomb-produced ¹⁴C, ²²²Rn/²²⁶Ra, and SF₆/³He.

Jones and Smith (1977) reported the first measurement of CO₂ fluxes by the eddy covariance technique using an open-path CO₂ gas analyzer over the coastal sea. Their result shows an order of magnitude larger CO₂ fluxes than the bulk CO₂ fluxes estimated using the gas

transfer velocity by mass balance techniques. Subsequent observations over coastal seas in a variety of environments are not consistent with the bulk CO₂ fluxes (Wesely *et al.*, 1982; Smith and Jones, 1985; Ohtaki *et al.*, 1989; Jacobs *et al.*, 1999). McGillis *et al.* (2001) reported the measurement of CO₂ fluxes evaluated by the eddy covariance technique over the open ocean using a closed-path CO₂/H₂O gas analyzer. Their result shows almost the same values as the bulk CO₂ fluxes estimated using the gas transfer velocity, which was simultaneously evaluated in situ by the mass balance technique using SF₆/³He dual tracers.

One important problem is the question of what physical and chemical processes influence the gas transfer across the air-sea interface, such as bubble enhancement, surfactant, surface film, wave, wind speed, and atmospheric stability. However, the present parameterization of the gas transfer velocity indicates that this velocity is a function of the wind speed only. Wanninkhof and McGillis (1999) suggested that this relationship depends on the time scale of the evaluated CO₂ fluxes. The eddy covariance technique can evaluate the trace gas flux on a shorter time scale than mass balance techniques, and also includes the uncertain processes (e.g. bubble enhance-

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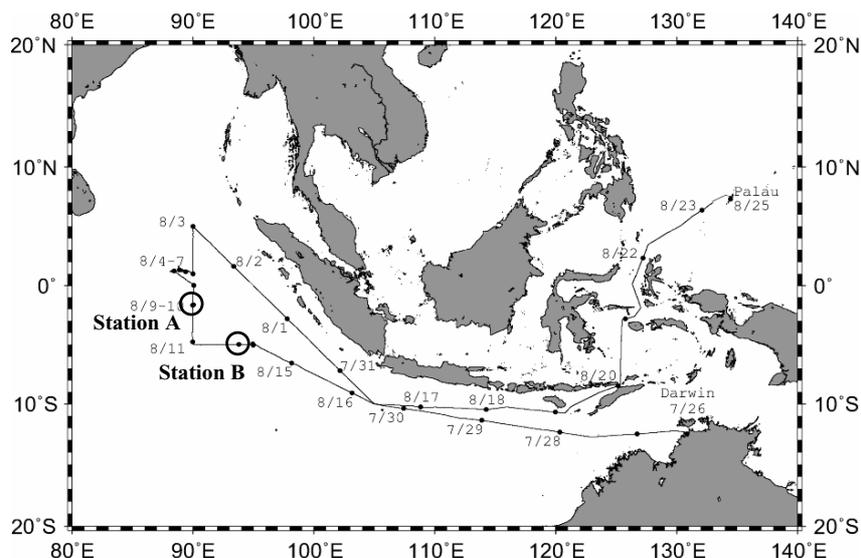


Fig. 1. Cruise track of the R/V MIRAI during MR05-03 leg 2. The two circles represent station A (90°E, 1.5°S) and B (95°E, 5°S) at which special observations of the turbulent flux were performed.

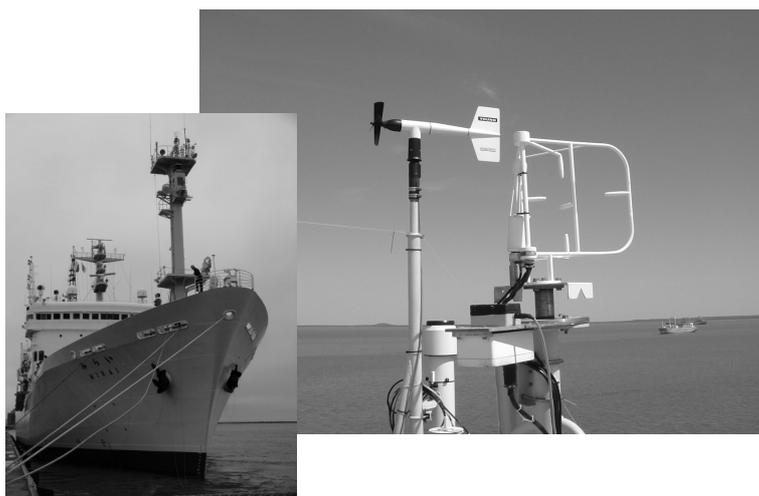


Fig. 2. Systems for turbulent flux observation and ship motion correlation on the top of the foremast of the R/V MIRAI, JAMSTEC.

ment, surfactant, surface film, wave, and atmospheric stability) that the parameterization does not consider.

This paper focuses on the direct measurement of CO₂ fluxes evaluated by the eddy covariance technique over the open ocean, and compares these fluxes with the bulk CO₂ fluxes estimated using the gas transfer velocity by mass balance techniques.

2. Methods and Observation Procedures

2.1 Study site

The measurement was conducted aboard the R/V

MIRAI of JAMSTEC (Japan Agency for Marine-Earth Science and Technology) in the equatorial Indian Ocean. The cruise track is shown in Fig. 1. The general meteorological data (air temperature, humidity, short- and long-wave radiation, wind speed and direction, and air pressure) were measured at about 24 m height above the sea surface. The properties of the bulk seawater temperature and salinity were measured at about 4.5 m depth below the sea surface. The fugacities of CO₂ in bulk seawater and air were also measured at the same height or depth (Murata and Takizawa, 2003).

During this cruise, special observations of the tur-

bulent flux were performed at the two stations. The first run at station A (90°E, 1.5°S) was performed from 9 (15:00) to 10 (3:00) August 2005; the second run at station B (95°E, 5°S) was performed from 12 (16:30) to 13 (4:30) August 2005. During both runs, the vessel was steered into the wind at a constant speed of 8 knots for about an hour, and then returned to keep the same position. This leg was repeated for 12 hours at the two stations.

2.2 Turbulent flux measurement system

We installed the turbulent flux system and ship motion correction system on the top of the foremast (about 24 m above the mean sea surface) of the R/V MIRAI (Fig. 2). The turbulent flux system consisted of a sonic anemometer-thermometer (KAIJO, DA-600-3TV) and an infrared open-path CO₂/H₂O gas analyzer (LI-COR, LI-7500). We cleaned the LI-7500 optical windows to avoid contamination by salt particles and raindrops before special observations, and checked the AGC value (“clean window” baseline value) before and after these observations. The ship motion correction system consisted of a two-axis inclinometer (Applied Geomechanics, MD-900-T), a three-axis accelerometer (Applied Signals, QA-700-020), and a three-axis rate gyro (Systron Donner, QRS11-0050-100). Analog output signals from these fast response instruments were sampled and digitized at the rate of 10 Hz by a data acquisition system (National Instruments, LabVIEW). The digitized signals were averaged to 2.5 Hz during the data processing.

2.3 Ship motion correction

One of the greatest difficulties in measuring turbulent flux by the eddy covariance technique over the open ocean is the apparent wind velocity caused by the ship motion. Fujitani (1985) suggested that the true wind velocity (V_{true}) can be expressed as follows:

$$V_{\text{true}} = TV_{\text{observed}} + \Omega T(R - r) + V_{\text{ship}}, \quad (1)$$

where V_{observed} is the raw wind velocity measured by the sonic anemometer, T is the coordinate transformation matrix, Ω is the angular velocity vector of the ship around the reference coordinate, R is the position vector of the sonic anemometer with respect to the ship coordinate, r is the position vector of the motion sensor, and V_{ship} is the velocity vector of the ship.

We adopted the simplified system proposed by Takahashi *et al.* (2005). This system was installed in the ship motion correction system at the same position as the sonic anemometer (i.e. $R = r$). The above equation can therefore be simplified as follows:

$$V_{\text{true}} = TV_{\text{observed}} + V_{\text{anemo}}. \quad (2)$$

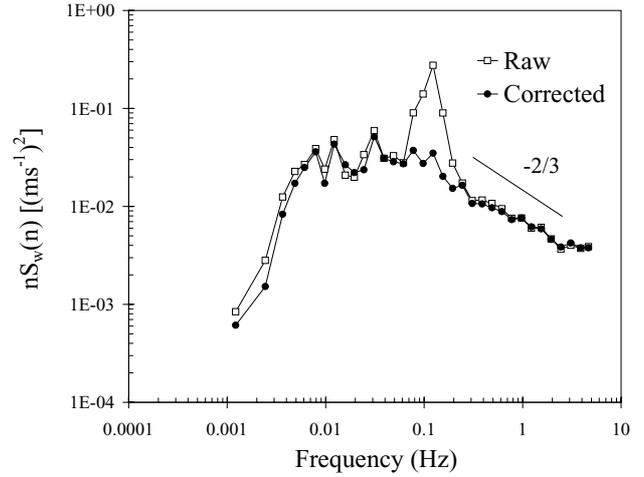


Fig. 3. Power spectra of the raw and corrected vertical wind velocity fluctuations. Raw data are open squares (\square) and corrected data are closed circles (\bullet).

The yaw, pitch, and roll angles measured by the inclinometer and rate gyro lead to the coordinate transformation matrix. V_{anemo} is obtained as the time integration of the three-axis accelerations. Figure 3 shows the power spectra of the raw and corrected vertical wind velocity fluctuations. The power spectrum of the raw vertical wind velocity fluctuation shows a false peak around 0.1 Hz. This frequency is the same as the cycles caused by the ship motion. On the other hand, the power spectrum of the corrected vertical wind velocity fluctuation does not show the false peak.

2.4 Turbulent CO₂ flux by the eddy covariance technique

The air-sea turbulent CO₂ flux by the eddy covariance technique is given by:

$$Fc(\text{eddy}) = \overline{w\rho_c} = \overline{w'\rho'_c} + \overline{w}\overline{\rho_c}, \quad (3)$$

where w is the vertical wind velocity and ρ_c is the CO₂ density. The prime indicates the fluctuation as deviations from the mean value (overbar) over a sampling period. The CO₂ flux was calculated as fifteen minute statistics. The first term on the right hand side of Eq. (3) is the raw CO₂ flux measured by the turbulent flux system. The second term on the right hand side is the CO₂ flux due to the mean vertical flow. Webb *et al.* (1980) suggested that the flux due to the mean vertical flow cannot be neglected for trace gases such as water vapor and CO₂. To evaluate the magnitude of the influence of the mean vertical flow, Webb *et al.* (1980) assumed that the vertical flux due to the density fluctuation of the dry air (ρ_a) should be zero:

$$\overline{w\rho_a} = \overline{w}\overline{\rho_a} + \overline{w'\rho'_a} = 0. \quad (4)$$

Using the ideal gas equation, the final expression for the mean vertical flow is as follows:

$$\bar{w} = \frac{\mu}{\rho_a} \overline{w' \rho'_v} + (1 + \mu\sigma) \frac{\overline{w' T'}}{\bar{T}}. \quad (5)$$

For the total CO₂ flux, it is as follows:

$$F_c(\text{eddy}) = \overline{w' \rho'_c} + \mu \frac{\bar{\rho}_c}{\rho_a} \overline{w' \rho'_v} + (1 + \mu\sigma) \frac{\bar{\rho}_c}{\bar{T}} \overline{w' T'}, \quad (6)$$

where μ is the ratio of the molecular weights of dry air and water vapor, σ is the ratio of water vapor and dry air densities, ρ_v is the water vapor density, and T is the absolute temperature. Practically speaking, the sensible ($\overline{w' T'}$) and latent ($\overline{w' \rho'_v}$) heat fluxes evaluated by the eddy covariance technique are used to calculate CO₂ fluxes due to the mean vertical flow (Webb correction terms). The actual values of these terms are discussed in Subsection 3.2. A detailed derivation of Eq. (4) can be found in Webb *et al.* (1980).

2.5 Air-sea bulk CO₂ flux

The air-sea bulk CO₂ flux is given by (Liss and Merlivat, 1986):

$$F_c(\text{bulk}) = k_{\text{CO}_2} S_{\text{CO}_2} \Delta f\text{CO}_2, \quad (7)$$

where k_{CO_2} is the gas transfer velocity of CO₂ modified by the Schmidt Number, S_{CO_2} is the solubility of CO₂, and $\Delta f\text{CO}_2$ is the difference between the fugacities of CO₂ in bulk seawater and air. We adopted the gas transfer velocity proposed by Wanninkhof (1992), which was estimated by mass balance techniques using tracers of the natural and bomb-produced ¹⁴C, ²²²Rn/²²⁶Ra, and SF₆/³He. The time scale of this mass balance-based gas transfer velocity ranged from several days to years. We estimated the bulk CO₂ flux at each station using the mean wind speed and $\Delta f\text{CO}_2$ for a period of 12 hours, during which the flux observation runs were performed.

3. Results

Figure 4 shows the time-series data of the difference between the fugacities of CO₂ in bulk seawater and air ($\Delta f\text{CO}_2 = f\text{CO}_{2,\text{sea}} - f\text{CO}_{2,\text{air}}$), and the wind speed including special observation periods of the turbulent flux.

The $\Delta f\text{CO}_2$ was positive and almost constant during both station runs. The mean $\Delta f\text{CO}_2$ was $+27.2 \pm 0.5 \mu\text{atm}$ at station A, and $+16.6 \pm 2.5 \mu\text{atm}$ at station B. The wind speed was light ($< 7 \text{ ms}^{-1}$) during both runs, varying from 1.4 to 5.9 ms^{-1} at station A and 1.6 to 6.6 ms^{-1} at station B.

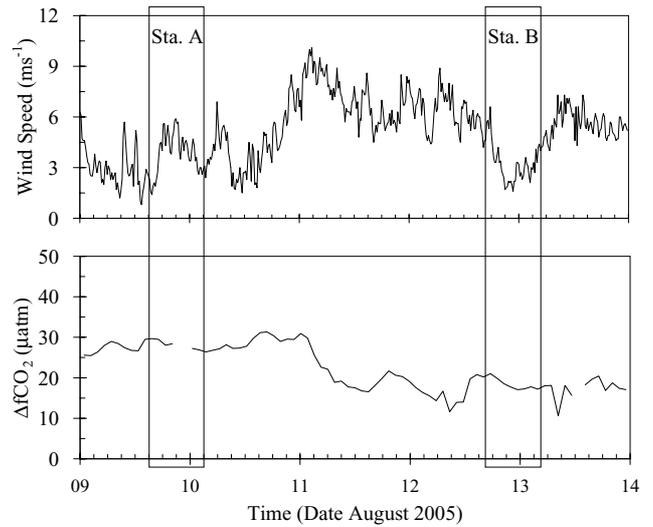


Fig. 4. Time-series data of the air-sea CO₂ fugacity difference ($\Delta f\text{CO}_2 = f\text{CO}_{2,\text{sea}} - f\text{CO}_{2,\text{air}}$, lower) and wind speed at 24 m above the sea surface (upper).

3.1 Spectrum and cospectrum

Figure 5 shows an example of the power spectra of the temperature (Δ), water vapor density (\blacksquare), and CO₂ density (\circ) fluctuations observed by the turbulent flux measurement system, normalized by the variance of each parameter. The abscissa is the nondimensional frequency, $f = nz/U$, on a logarithmic scale. Here, n is the frequency [Hz], and U is the wind speed [ms^{-1}] at the measurement height z [m].

The shapes of the power spectra of the temperature, water vapor density, and CO₂ density fluctuations were basically similar. The power spectra of the temperature and water vapor density fluctuations followed the Kolmogorov $-5/3$ power law over a wide frequency range above 0.04. On the other hand, the power spectrum of CO₂ density fluctuation followed the $-5/3$ power law in the frequency range 0.04–1.6, while white noise appeared above the nondimensional frequency range 1.6.

This CO₂ white noise is due to the inadequate sensitivity of the gas analyzer to the small CO₂ density fluctuation. The CO₂ density fluctuations over the ocean are an order of magnitude smaller than those over the vegetation on land.

Figure 6 shows an example of the cospectra of the parameters mentioned above and the vertical wind velocity as functions of the nondimensional frequency. Each flux is equivalent to a frequency integral of the cospectrum of each parameter. The shapes of the cospectra of the temperature, water vapor, and CO₂ fluxes were also basically similar, giving the same result as the power spectra. The cospectrum of the CO₂ flux showed a negative flux (rep-

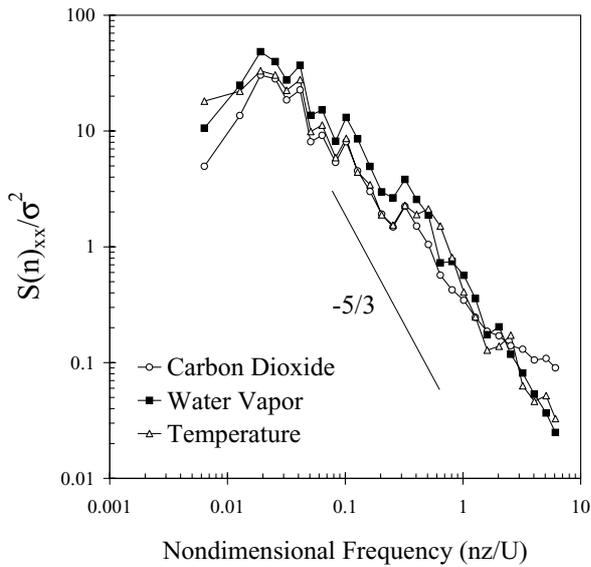


Fig. 5. Power spectra of carbon dioxide density (○), water vapor density (■), and temperature (△) fluctuations normalized by the variance during 12 (16:45–17:00) August 2005.

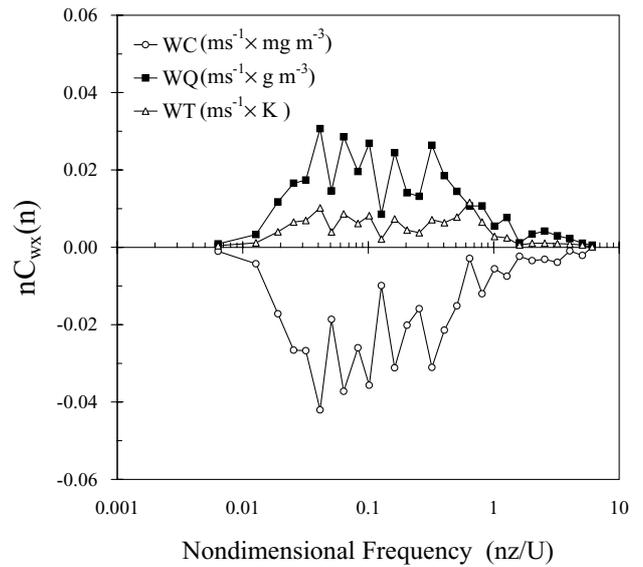


Fig. 6. Cospectra of vertical wind velocity fluctuation and carbon dioxide density (WC, ○), water vapor density (WQ, ■), and temperature (WT, △) fluctuations during 12 (16:45–17:00) August 2005.

representing a CO₂ sink) over the entire frequency range, while those of the temperature and water vapor fluxes were positive fluxes (representing energy sources).

The cospectrum of the CO₂ flux does not show the influence of white noise, which was observed in the high frequency range of the power spectrum in Fig. 5. This is because the instrumental white noise of CO₂ density fluctuation does not correlate with the vertical wind velocity fluctuation. This cospectrum was mainly limited to the frequency range 0.006–1.6, and the most dominant frequency range was centered at 0.04, as were the cospectra of the temperature and water vapor fluxes. The CO₂ flux in the frequency range 1.6–6.1 showing white noise represented only about 2 percent of the entire CO₂ flux. As this cospectrum does not show any influence from the ship motion, the turbulent flux (integrated value as the covariance) evaluated directly by the eddy covariance technique is considered to be reasonable.

3.2 CO₂ flux

Figure 7 shows the time-series data of the raw and total CO₂ fluxes evaluated directly by the eddy covariance technique, and those of the CO₂ fluxes due to the mean vertical flow corrected by the sensible and latent heat fluxes (Webb correction terms) at the two stations. The open circle (○), solid circle (●), open triangle (△), and solid square (■) represent the raw and total CO₂ fluxes, and the Webb correction terms for the sensible and latent heat fluxes, respectively. The Webb correction terms for

Table 1. Mean values and standard deviations of the wind speed and the air-sea CO₂ fugacity difference ($\Delta f\text{CO}_2$), and the estimated bulk CO₂ fluxes at the two stations.

	Wind speed [ms ⁻¹]	$\Delta f\text{CO}_2$ [μatm]	Bulk CO ₂ flux [10 ⁻³ mg m ⁻² s ⁻¹]
Station A	3.4 ± 1.0	+27.2 ± 0.5	+0.51
Station B	4.7 ± 1.6	+16.6 ± 2.5	+0.59

the sensible and latent heat fluxes were evaluated directly by the eddy covariance technique, and calculated as the second and third terms of Eq. (6).

The raw CO₂ fluxes were negative, and the mean value was $-0.052 \text{ mg m}^{-2}\text{s}^{-1}$ at station A and $-0.063 \text{ mg m}^{-2}\text{s}^{-1}$ at station B. The Webb correction terms were positive, and the mean values were $+0.023 \text{ mg m}^{-2}\text{s}^{-1}$ (Webb correction term for the sensible heat flux) and $+0.039 \text{ mg m}^{-2}\text{s}^{-1}$ (Webb correction term for the latent heat flux) at station A, and $+0.017 \text{ mg m}^{-2}\text{s}^{-1}$ and $+0.047 \text{ mg m}^{-2}\text{s}^{-1}$ at station B, respectively. The Webb correction term for the latent heat flux was larger than the Webb correction term for the sensible heat flux. The sum of Webb correction terms for the sensible and latent heat fluxes was almost the same as or rather larger than the raw CO₂ flux. As a result, the total CO₂ fluxes were positive, the mean value being $+9.6 \times 10^{-3} \text{ mg m}^{-2}\text{s}^{-1}$ at station A and $+0.9 \times 10^{-3} \text{ mg m}^{-2}\text{s}^{-1}$ at station B.

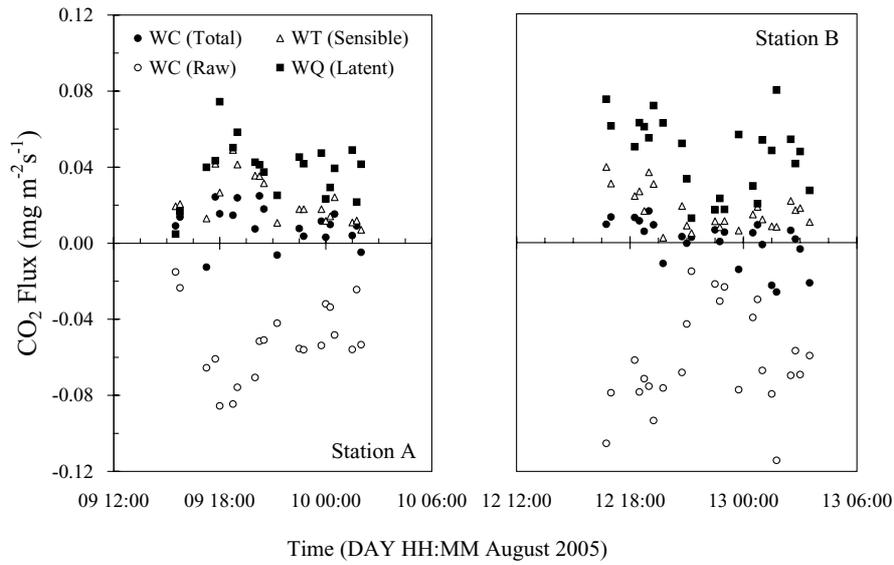


Fig. 7. Time-series data of total (●) and raw (○) CO₂ fluxes, and mean vertical flow CO₂ fluxes corrected by sensible (△) and latent (■) heat fluxes at station A (left) and station B (right).

Table 2. Mean values (upper) and ranges (lower) of the CO₂ fluxes evaluated directly by the eddy covariance technique, and mean values of the estimated bulk CO₂ flux at the two stations. The sensible and latent heat fluxes evaluated by the eddy covariance technique are used to calculate of the mean vertical flow CO₂ fluxes (Webb correction terms). Units are 10⁻³ mg m⁻²s⁻¹.

	CO ₂ flux by eddy covariance technique				Bulk CO ₂ flux
	Raw CO ₂ flux	Webb correction term for the sensible heat flux	Webb correction term for the latent heat flux	Total CO ₂ flux	
Station A	-51.95 -85.54~-15.11	+22.94 +7.11~+49.05	+38.62 +4.81~+74.38	+9.62 -12.57~+24.93	+0.51
Station B	-62.66 -114.21~-15.06	+17.05 +2.58~+39.69	+46.50 +12.84~+80.06	+0.90 -25.88~+16.55	+0.59

On the other hand, the bulk CO₂ fluxes estimated using the gas transfer velocity proposed by Wanninkhof (1992) were $+0.51 \times 10^{-3}$ mg m⁻²s⁻¹ at station A and $+0.59 \times 10^{-3}$ mg m⁻²s⁻¹ at station B (Table 1). As a result, the total CO₂ flux evaluated by the eddy covariance technique was an order of magnitude larger than the estimated bulk CO₂ flux at station A. However, the total CO₂ flux evaluated by the eddy covariance technique was similar to the estimated bulk CO₂ flux at station B.

4. Discussion

Direct measurements of the air-sea CO₂ flux by the eddy covariance technique were performed over the open ocean with careful quality control. The Δf CO₂ was positive and almost constant at both stations. The wind speeds were light, varying over a rather narrow range 1.4–6.6

ms⁻¹ during special observations of the turbulent flux. The shapes of the normalized power spectra of the temperature, water vapor density, and CO₂ density fluctuations were basically similar. The total CO₂ fluxes were positive and the mean vertical flow CO₂ fluxes by the Webb correction were almost the same as or rather larger than the raw CO₂ fluxes.

Table 2 shows the mean values and ranges of the CO₂ fluxes evaluated by the eddy covariance technique, and the mean values of the estimated bulk CO₂ flux at the two stations. The sensible and latent heat fluxes evaluated by the eddy covariance technique are used for the calculation of the mean vertical flow CO₂ fluxes (Webb correction terms). The total CO₂ flux by the eddy covariance technique and the bulk CO₂ flux disagree at station A, but become similar at station B. However, the

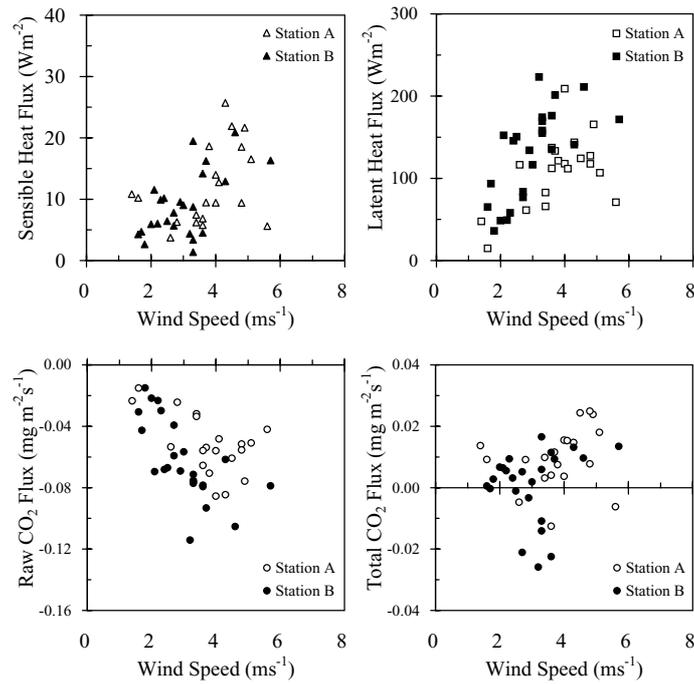


Fig. 8. Sensible heat, latent heat, raw CO₂, and total CO₂ fluxes as a function of wind speed using data from both stations.

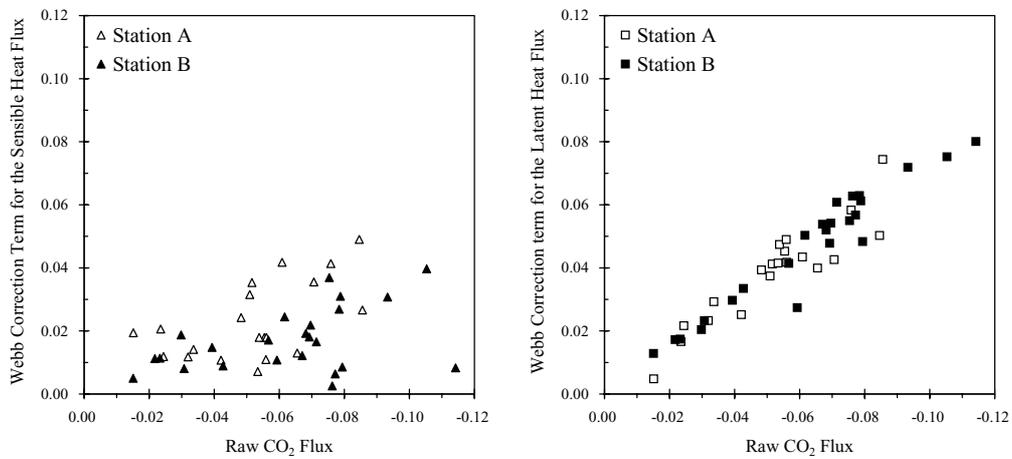


Fig. 9. Webb correction terms for sensible and latent heat fluxes as a function of raw CO₂ flux using data at both stations.

ranges of the total CO₂ flux by the eddy covariance technique at both stations are almost the same.

Figure 8 shows the sensible heat, latent heat, raw CO₂, and total CO₂ fluxes as a function of the wind speed using the data at both stations. Although there is some scatter, the sensible heat, latent heat, raw CO₂ fluxes increase with increasing wind speed, due to the increasing turbulent intensity. On the other hand, the total CO₂ flux depends roughly on the wind velocity as well as the sen-

sible heat, latent heat, and raw CO₂ fluxes at both stations. However, the total CO₂ fluxes at station B include large negative values, even after the Webb correction is applied.

Figure 9 shows the Webb correction terms for the sensible and latent heat fluxes as a function of the raw CO₂ flux using the data at both stations. The Webb correction term for the latent heat flux correlates closely with the raw CO₂ flux at both stations, although there is a

poorer correlation between the Webb correction term for the sensible heat flux and the raw CO₂ flux. The large negative total CO₂ fluxes at station B may be due to small sensible heat fluxes, even given the large raw CO₂ fluxes. However, the cospectra of sensible heat fluxes are basically similar to those of latent heat and raw CO₂ fluxes.

McGillis *et al.* (2001) suggested that CO₂ flux measurements by the eddy covariance technique should be performed under large $\Delta f\text{CO}_2$ and high wind speed environments to be successful. However, we need to know what certain parameters to control the CO₂ fluxes over the various oceans. In fact, there are very few large $\Delta f\text{CO}_2$ areas over the ocean (Takahashi *et al.*, 2002). In this study, we observed the CO₂ fluxes by the eddy covariance technique in areas of small $\Delta f\text{CO}_2$ and low wind speed over the ocean. The CO₂ density fluctuation shows white noise in the high frequency range, which has no influence on the CO₂ fluxes.

Our goal is the precise evaluation of the ocean uptake of anthropogenic CO₂. The eddy covariance technique is the only direct air-sea flux measurement that allows us to understand the process controlling the air-sea CO₂ fluxes over short time and space scales. To reach our goal, the following issues remain for future studies:

(1) Flux evaluation by observations at fixed points and for a long term (days and weeks) over the open ocean;

(2) Development of a turbulent flux measurement system without the need for the Webb correction;

(3) Confirmation of the transport direction measuring the vertical CO₂ concentration profile in air.

The time scale needed to estimate the gas transfer velocity by mass balance techniques is generally long, from days (²²²Rn/²²⁶Ra and SF₆/³He) to years (natural and bomb-produced ¹⁴C). On the other hand, the eddy covariance technique can evaluate the trace gas flux on the time scale of minutes or hours. In this study, we have attempted to evaluate the CO₂ fluxes by the eddy covariance technique on a time scale of 12 hours at the same position over the open ocean. The present results did not always correspond to the bulk CO₂ fluxes estimated using the gas transfer velocity by mass balance techniques.

This study found that the upward mean vertical flow CO₂ fluxes by the Webb correction are of the same order as the downward raw CO₂ fluxes, leading to the upward total CO₂ fluxes. This is the same situation as the coastal sea results reported by Ohtaki *et al.* (1989). The total CO₂ fluxes are order of magnitude larger than the bulk CO₂ fluxes. We need to attempt flux observations using turbulent flux measurement system without the need for the Webb correction, and to measure the vertical CO₂ concentration profile in air.

Further work is now in progress in an attempt to measure CO₂ flux more precisely by the eddy covariance technique.

Acknowledgements

We wish to express our heartfelt thanks to all crews of the R/V MIRAI lead by Captain Kita. We are deeply indebted to the staff of JAMSTEC, Marine Work Japan, LTD. (MWJ), and Global Ocean Development Inc. (GODI). Mr. Moro of MWJ and Dr. Murata of JAMSTEC are acknowledged for their invaluable assistance in obtaining *f*CO₂ data. Special thanks are given to Dr. Yoneyama and Dr. Hase of JAMSTEC for their arrangements.

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