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Influence of Gas Transfer Velocity Parameterization on Air-Sea CO_2 Exchange in the East (Japan) Sea

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Gas flux across the air-sea interface is often determined by the product of gas transfer velocity (k) and the difference of concentrations in water and air. k is primarily controlled by wind stress on the air-sea interface, thus all parameterizations of k involve wind speed, a rough indicator of wind stress, as one of the independent variables. We attempted to explore the spatial and temporal variations of k in the East (Japan) Sea using a database from Na *et al.* (1992). Three different parameterizations were employed: those of Liss and Merlivat (1986), Wanninkhof (1992), and Wanninkhof and McGillis (1999). The strong non-linear dependence of k on wind speed in all parameterizations leads us to examine the effect of time resolution, in which the binned wind speeds are averaged, on the estimation of k. Two time resolutions of 12 hours (short-term) and one month (long-term) were chosen. The mean wind speeds were fed into the given parameterizations, resulting in six different transfer velocities of CO_2 ranging from 12 to 32 cm/h. In addition to the threefold difference depending on the choice of parameterization, the long-term average of wind speed results in a value of k up to 20% higher than the short-term (12 hours) average of wind speed due to the non-Rayleigh wind distribution in the East (Japan) Sea. While it is not known which parameterization is more reliable, this study proposes that the time-averaged wind speed should not be used in areas where non-Ralyleigh wind distribution prevails such as the East (Japan) Sea. The net annual CO_2 flux was estimated using the value of k described above and the monthly ΔfCO_2 of Oh *et al.* (1999); this ranges from 0.034 to 0.11 Gt-C/yr.

Key words: Air-sea gas exchange, Gas transfer velocity, The East (Japan) Sea

INTRODUCTION

Determination of gas fluxes between reservoirs (e.g., atmosphere, ocean, biosphere, soil, etc.) is one of the key questions involved in understanding the biogeochemical cycles in our planet, and furthermore in predicting the impact of human activities, which have already been diagnosed as being responsible for the rapid change of global climate during last several decades (IPCC, 2001). The atmosphere resides at the center of those reservoirs, interacting directly with the other reservoirs. Among them, the ocean plays a significant role in the budget of the gases that influence global climate directly or indirectly (e.g. CO_2, N_2O, DMS), as well as atmospheric chemistry (e.g. methyl halide).

The ocean covers about 75% of the Earth surface where gas exchange takes place. It is the micro-layers at the uppermost layer of the ocean or the lowest level of the atmosphere that regulates gas fluxes from or to the atmosphere depending on the solubility of the gas. The driving force for gas flux is the difference of concentrations across this micro-layer (ΔC), and the rate depends on the physical and chemical conditions at the layer. The latter is called gas transfer velocity or piston velocity (k). The product of those two terms provides the amount of gas emitted or absorbed in a unit area:

$$F = k\ \Delta C \qquad (1)$$

Modern technology facilitates the direct detection of most of the gases comprising the atmosphere *in situ*, but it has not been possible up to now to measure k directly in the field. Consequently, the determina-

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tion of gas flux in the ocean relies on an empirical parameterization of k. Several parameters have been investigated to determine their relation to the variation of k in the field and the laboratory (Liss and Merlivat, 1986; Wanninkhof, 1992; Erickson, 1993; Wanninkhof and McGillis, 1999; Nightingale *et al.*, 2000). Momentum flux due to wind is known to be a major parameter responsible for controlling k and playing a triggering role with respect to other parameters such as the wave field and whitecap coverage that also cause k to vary (Bock *et al.*, 1999). Accordingly, all parameterizations are basically a function of wind speed obtained at a certain height.

Accumulation of wind data obtained from commercial ship and research vessel observations and buoy records make it possible to map the climatological short- and long-term wind field in the ocean. These data have been used to estimate global scales of gas flux in the ocean. In addition, recent progress in the development of algorithms for deriving surface wind data from remotely sensed data originating from satellites will provide a high-quality short-term wind data set covering the whole world (Bentamy *et al.*, 2002).

A considerable number of studies in the literature have reported global estimations of gas flux using conventional and remotely sensed wind fields in the ocean (Tans *et al.*, 1993; Takahashi *et al.*, 1999; Donelan and Wanninkhof, 2002). However, those estimates were based on the rather large geographic scale of the open ocean, of which a large fraction is oliogotropic, so that scaling down the impact of biogenic gases on the gas flux is unavoidable. Coastal and marginal seas are in general eutropic, resulting in dissolved biogenic gases being enriched, which in turn leads to enhancement of the gas flux out of the ocean (Tsunogai *et al.*, 1997; Liu *et al.*, 2000). Consequently, accurate prediction of k in this region is required because of its more sensitive response to the estimation of gas flux than the open ocean.

In this paper, we have estimated gas transfer velocities in a marginal sea, the East (Japan) Sea, surrounded by the Korean peninsula, the Japanese archipelago, and the East Asian continent. We have employed three parameterizations, frequently adopted in the literature: those of Liss and Merlivat (1986), Wanninkhof (1992), and Wanninkhof and McGillis (1999) (Fig. 1). The lack of wind speed measurements has obliged the researchers to use long-term averaged wind speed for the estimation of gas transfer velocity. It, however, has been known that the averaging period of wind speed could influence on the variability of gas transfer velocitiy (Bates and Merlivat, 2001; Wanninkhof *et al.*, 2002). We have used synoptic wind data extracted from weather maps over 18 years, which enable us to verify short- and long-term scales of wind speeds and thus gas transter velocities. Furthermore, this data set could be useful for a future comparative study using remotely sensed data.

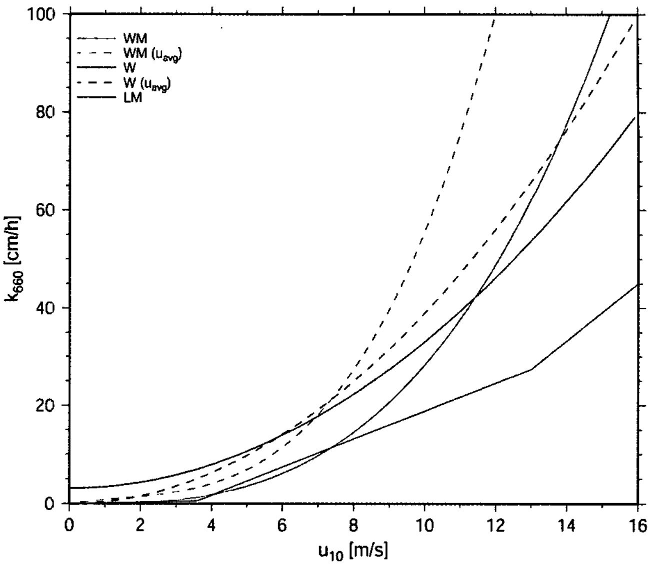

Fig. 1. The employed relationships between transfer velocity and wind speed. u_{10} is the wind speed at 10 m above sea surface. All transfer velocities (k) are normalized to the Schmidt number (Sc) of CO_2 in seawater at 20°C. The blue line shows the parameterization of Liss and Merlivat (1986). The black solid and dashed lines represent for the short- and long- term parameterizations of Wanninkhof (1992), respectively. The red lines are the parameterizations from Wanninkhof and McGillis (1999).

DATA

We used bi-diurnal wind speed measurements for the period 1978-1995 in the East Sea (Na *et al.*, 1992). Na *et al.* (1992) calculated the wind speed by using the Cardone model, which accepts atmospheric pressure, air temperature, and sea surface temperature as input data. The authors gridded atmospheric pressure and air temperature from the weather map, forecasted twice a day by the Japan Meteorological Agency, and calculated the sea surface temperature by using a 10-day mean temperature obtained by the Maizuru Marine Observatory. The resultant wind speed data are supplied in 1/2-degree square resolution every 12 hours. Although the data were obtained rather indirectly compared to shipboard measurement or satellite remote sensing, the wind data from Na *et al.* (1992) are superior in respect of the continuity in time and

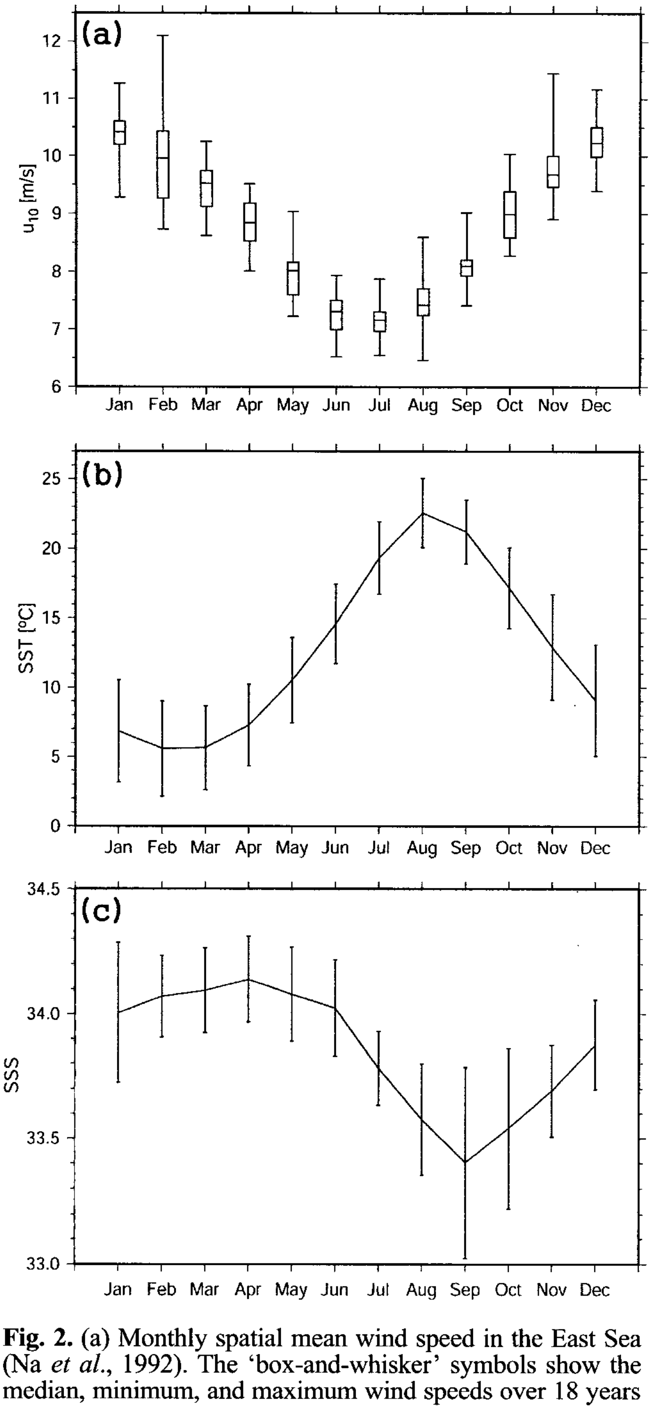

Fig. 2. (a) Monthly spatial mean wind speed in the East Sea (Na *et al.*, 1992). The 'box-and-whisker' symbols show the median, minimum, and maximum wind speeds over 18 years ('78-95). (b) SST from World Ocean Atlas '98 (WOA 98). *k* is dependent on SST as well as wind speed. The error bar denotes the standard deviation. (c) Monthly mean salinity used to calculate the solubility of gas, also derived from WOA 98.

space at the present time (Fig. 2(a)).

Sea surface temperature and salinity subsets were extracted from the World Ocean Atlas 98, which was generated by objective analysis of the World Ocean Database 98 (WOD 98) by NODC/NOAA. The WOD 98 contains observed and standard level profiles measured by various instruments such as CTD, XBT, and moored buoys. Over 320,000 stations in the East Sea, investigated for ~70 years (1925-present), were included in the WOD 98 (Fig. 2(b) and (c)).

METHODS

Gas transfer velocities

k is a function of the interfacial turbulence, the kinematic viscosity of the water (ν), and the diffusion coefficient of the gas (*D*). The relationship between ν and *D* is expressed as the Schmidt number (*Sc*), that is, $Sc=\nu/D$. Most parameterizations predict that *k* is proportional to $Sc^{-1/2}$ for an interface with waves (Ledwell, 1984). However, the relationship between *k* and wind speed is still under debate, ranging from linear to cubic equation representations.

The current estimates of global CO_2 uptake ranging from 1 to 3 Gt-C yr^{-1} are highly dependable on the choice of parameterization of *k* with wind speed (Boutin *et al.*, 2002). To investigate the effect of the parameterization on the gas exchange in the East Sea, we considered three frequently used parameterizations to derive transfer velocities: those of Liss and Merlivat (1986), Wanninkhof (1992), and Wanninkhof and McGillis (1999) (Fig. 1).

The parameterization of Liss and Merlivat (1986; hereafter LM) consists of three linear segments, intended to explain three different regimes: smooth surface, rough surface, and breaking wave regimes. The LM parameterization is essentially based on wind tunnel experiments and normalized with the lake results obtained from a purposely-added SF_6 experiment in order to overcome the small-scale problems of wind tunnels (Wanninkhof *et al.*, 1985). This parameterization predicts the lowest gas transfer velocities among those commonly adopted parameterizations.

The parameterization of Wanninkhof (1992) assumes a quadratic dependence between gas transfer and wind speed (*u*), that is, $k=fu^2$. The coefficient *f* was determined using bomb and natural-^{14}C gas exchange data (Broecker *et al.*, 1985; Cember, 1989). Further, a relationship between short-term wind and gas transfer was derived from the above relationship for long-term wind speed by assuming the Rayleigh distribution of wind speed. The Rayleigh distribution function is regarded as a reasonable approximation for a global ocean wind speed frequency distribution. This can be expressed in terms of the averaged wind speed:

$$P(u) = \frac{u[\exp(-u^2/2\Delta u^2)]}{2\pi\Delta u^2}$$

$$\Delta u = u_{avg}(\pi/2)^{-1/2} \qquad (2)$$

where $P(u)$ is the probability distribution function, u is the steady wind speed, and u_{avg} is the average climatological wind speed (Wentz *et al.*, 1984).

Wanninkhof and McGillis (1999; hereafter WM) proposed a cubic relationship between gas transfer and wind speed ($k = f\,u^3$) based on direct CO_2 flux measurements using eddy correlation (McGillis *et al.*, 1999). The authors argued that the strong non-linearity could be attributed to the retardation of gas transfer at low winds by surfactants, and enhanced bubble entrainment at high winds. The cubic equation was forced to fit the global mean flux derived using the ^{14}C inventory.

Computation of gas transfer velocities

In addition to the choice of parameterization, the averaging period of wind speed could have an influence on the uncertainty of the estimates of CO_2 uptake because most of the parameterizations suggest a non-linear dependency on wind speed (Wanninkhof *et al.*, 2002). To examine the difference between short-term and long-term parameterizations, we derived monthly mean transfer velocities (k) in two ways; for short-term parameterization we calculated transfer velocities using the 12-hour wind speed first and then averaged them for 18 years (1978-1995) for each month. For the long-term parameterization, meanwhile, we calculate the monthly mean wind speed by averaging the wind speed for 18 years first and then applying the mean wind speed to the parameterization. As we calculated the transfer velocities of short- and long-term wind speeds

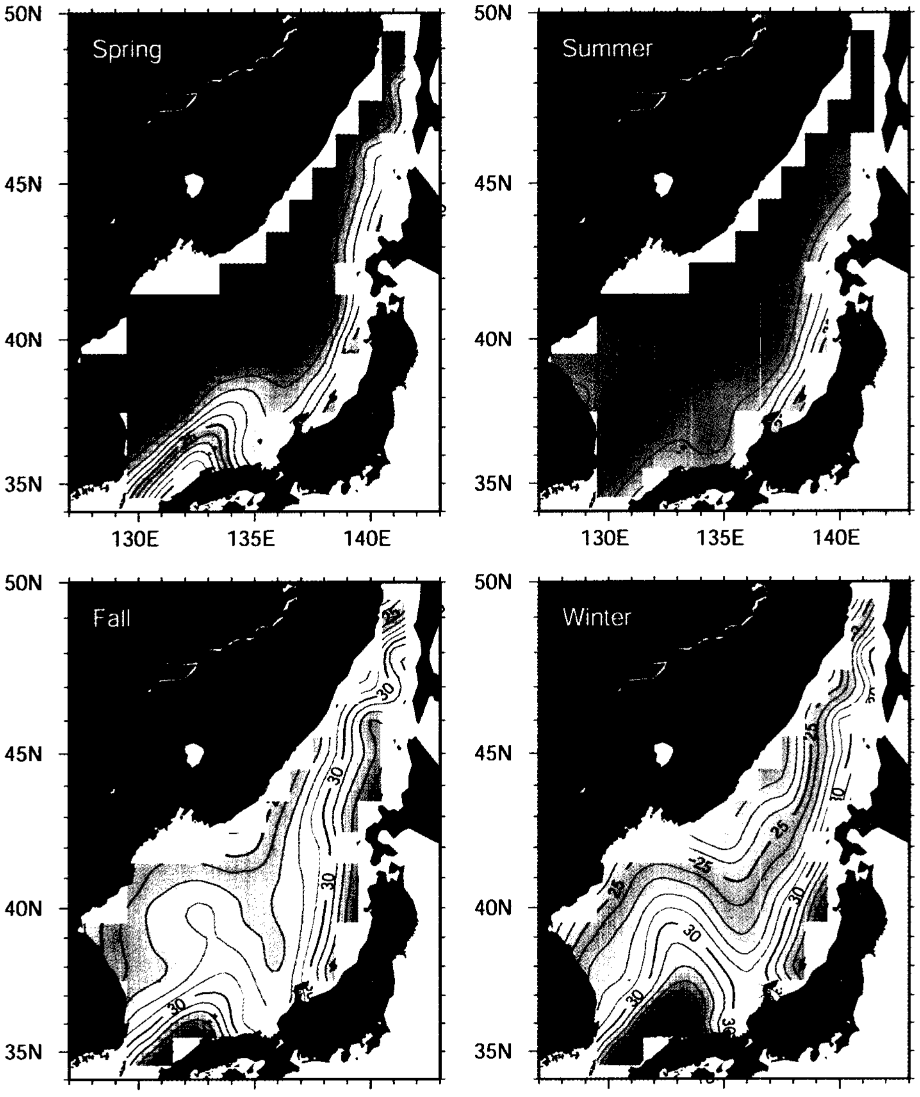

Fig. 3. Seasonal mean transfer velocities of CO_2 in cm/h. Parameterization for short-term wind speed from Wanninkhof (1992) was employed to show the spatial variation according to the season.

for the above three parameterizations, we compared 6 sets of transfer velocities in total.

To calculate gas transfer velocities we fed 1/2-degree square field temperature data into the parameterizations for short-term wind as well as for long-term wind because the daily variation of temperature is much smaller than that of wind speed. The above 1/2-degree square field was derived by linear interpolation of the 1-degree square values of WOA 98 to take advantage of the fine resolution of wind speed.

The boundaries of the study area were determined by considering the geographical distribution and the availability of data around the East Sea. The southern boundary was set at 35°N above the Korea Strait, while the northern one was placed at 50°N because the wind speed and sea surface temperature were not available above 50°N. The western and eastern boundaries were easily confined by the Eurasian Continent and Japanese Islands.

RESULTS AND DISCUSSION

Seasonal and spatial variability of gas transfer velocities

The six sets of transfer velocities show similar seasonal and spatial variability even though they have significant differences in magnitude (Fig 3, 4). We show the transfer velocities derived from the parameterization of Wanninkhof (1992) for short-term wind speed, possessing intermediate values compared to those of LM and WM, as examples of the seasonal and spatial variability of transfer velocities. In spring and summer, transfer velocities range from 15 to 25 cm/h. Owing to the relatively higher wind speed along the Japanese islands, the eastern part of the East Sea has higher transfer velocities than the western part. This trend is also found in fall and winter. However, transfer velocities in fall and winter are about 10 cm/h higher than those in spring and summer on average. Rather strong transfer velocities, due to the intensified wind speed near Vladivostok, are found over the western Japan Basin in winter. Although fall and winter have nearly identical transfer velocities of ~28 cm/h on average, the East Sea seems to play different roles in air-sea CO_2 exchange in those two seasons (See section '*Estimetion of CO_2 flux in the East Sea*').

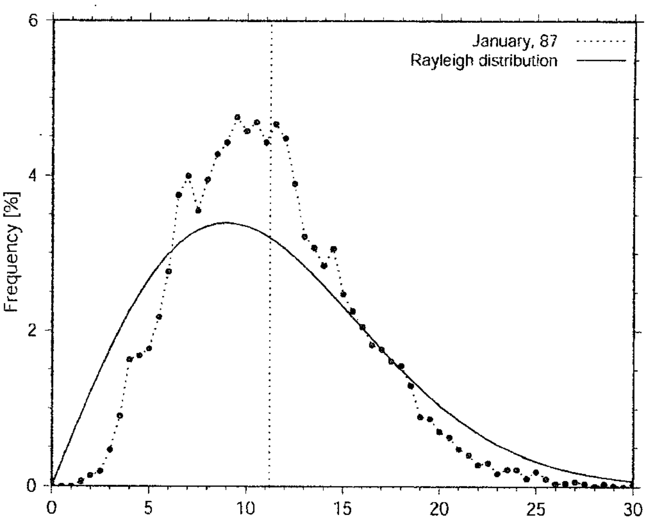

Fig. 5. Wind speed distribution in January, 1987 (dotted line with solid circles). The solid line shows the Rayleigh distribution (Wentz *et al.*, 1984) with the mean wind speed of 11.24 m/s in January. The overestimation of strong wind frequency results in much larger gas transfer if long-term wind is applied rather than short-term wind.

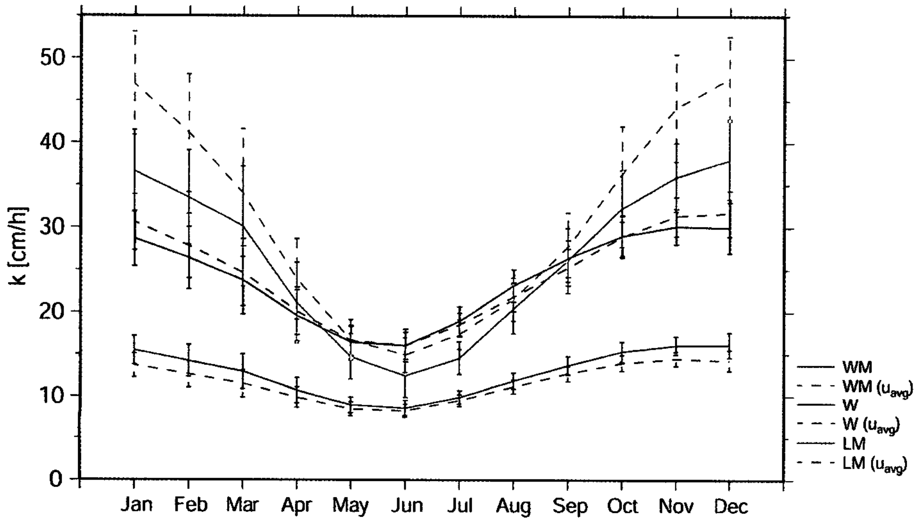

Fig. 4. Monthly variation of transfer velocities by the parameterization of Liss and Merlivat (1986; blue), Wanninkhof (1992; black), and Wanninkhof and McGillis (1999; red). The solid and dashed lines were calculated by feeding short- and long-term winds, respectively.

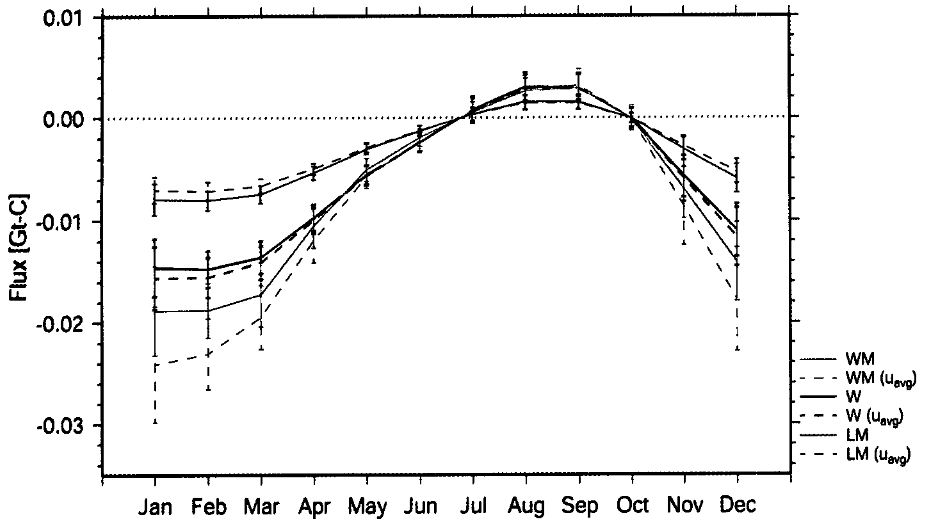

Fig. 6. Monthly variation of CO_2 fluxes. The color codes are the same as those of figure 4. The net uptake of CO_2 varies from 0.0343 to 0.106 Gt-C/yr according to the parameterization (Table 2).

Influence of parameterization on transfer velocities

The six transfer velocities calculated from the combination of three different parameterizations and two (short/long-term) wind data sets represent well the seasonal variability of wind speed. However, those parameterizations show large differences in absolute values; the rather narrow range of 10~17 cm/h in summer broadens to 15~48 cm/h in winter. It is easy to predict the larger difference between those parameterizations in winter from the relationship between wind speed and transfer velocity (Fig. 1). The stronger wind, the larger difference in *k*!

The parameterization of LM shows the lowest value of k throughout the year (Fig. 4). This is because the weakest wind speed in July (>7 m/s) is strong enough for the other parameterizations to give higher *k* values than that of LM. The parameterization of LM yields consistently lower transfer velocities with long-term wind speed than those with short-term wind speed. Since most relationships between transfer velocities and wind speeds are nonlinear, gas transfer velocities measured under variable winds will be higher than those measured under steady wind conditions for the same average wind speed. Thus, the relationship of LM based on short-term measurements with variable winds underestimates gas transfer velocities if long-term averaged winds are used (Wanninkhof, 1992).

The two parameterizations of Wanninkhof (1992; hereafter W) for long and short-term wind speeds have similar values throughout the year except for slightly higher values in winter. The discrepancy in winter becomes greater if the parameterizations of Wanninkhof and McGillis (1999) are applied. This phenomenon could be attributed to the non-Rayleigh distribution of wind speed in the East Sea (Fig. 5). Since the parameterizations of W and WM were obtained by assuming that the Rayleigh wind speed distribution (Wentz *et al.*, 1984) is valid for all regions of the ocean, the skewed wind distribution in the East Sea results in overestimation of the transfer velocities. Since the cubic relationship of WM assumes a stronger dependency of transfer velocities on wind speed than the quadratic one of W, it is expected that there will be a greater discrepancy between transfer velocities by long- and short-term wind speed parameterizations where the Rayleigh wind distribution is not valid (Wanninkhof *et al.*, 2002). The estimation of air-sea transfer velocities would be improved by employing regional algorithms based on the wind speed distribution in the East Sea.

Table 1. Transfer velocities calculated by using long- and short-term wind speed

	Long-term wind	Short-term wind	Long/short ratio
k_{LM}	11.70 cm/h	12.80 cm/h	0.91
k_W	24.43 cm/h	24.02 cm/h	1.02
k_{WM}	31.05 cm/h	26.33 cm/h	1.18

In summary, transfer velocities in the East Sea are highly dependent on the choice of parameterization and wind speed, and range from 11.70 to 31.05 cm/h on average (Table 1). Long-term averaged wind measurements seem to significantly overestimate (~20%) transfer velocities when parameterization assuming a higher dependency on wind speed is selected.

Estimation of CO_2 flux in the East Sea

We estimated CO_2 flux in the East Sea by combining the above transfer velocities with the only available fugacity field simulated by Oh *et al.* (1999). Oh *et al.* (1999) devised a model predicting the variation of surface fCO_2 by combining the solubility and biological pumps. The model, assuming the total alkalinity is constant through time, first calculates fCO_2 using input data such as total carbon dioxide (TCO_2), temperature and salinity at the sea surface, and mixed layer depth, and then predicts the integrated TCO_2 in the mixed layer for the day. TCO_2 for the next day is calculated by subtracting the export production and gas exchange at the air-sea interface. fCO_2 for each day was calculated repeatedly using the newly calculated TCO_2 and the other parameters. We calculated 1/2-degree square values of ΔfCO_2 from 1-degree square fields since the transfer velocities were calculated at 1/2-degree square resolution from the above wind speed, temperature, and salinity. The emission from July to September is only 0.007 Gt-C, which is 11% of the absorption during the rest of the year (Fig. 6). The largest flux was found in January owing to the large fugacity difference and transfer velocities.

The net CO_2 fluxes vary from 0.0343 Gt-C/yr to 0.106 Gt-C/yr according to the choice of parameterizations (Table 2). The fluxes also depend on the averaging period for the wind. The parameterization of WM for long-term wind, which assumes the highest dependence on wind speed, gives some 20% larger flux than that for short-term wind. The previous study by Oh *et al.* (1999), based on the short-term parameterization of Wanninkhof (1992), suggested that the net flux from the East Sea to the atmosphere should be 0.032±0.012 Gt-C/yr. Their flux is only half our value of 0.0705 Gt-C/yr although both studies utilized basically the same wind (Na *et al.*, 1992), temperature, and salinity data (NODC/NOAA). The difference could be attributed to the averaging period of wind speed and the spatial resolution. Oh *et al.* (1999) calculated transfer velocities using daily averaged wind speed at 1-degree square resolution. Considering that we applied a 12-hour interval wind speed at 1/2-degree square resolution, their wind speed averaged over longer time and larger space would result in a lower flux than ours because of the non-linear relationship between wind speed and transfer velocity. Underestimation of CO_2 flux caused by the low sampling frequency of wind speed was also found by Bates and Merlivat (2001). They reported that the same short-term parameterization gave three-time larger flux if hourly winds were used rather than daily winds.

Table 2. The net absorption of CO_2 by the East Sea

	Long-term wind	Short-term wind	Long/short ratio
F_{LM}	0.0343 Gt-C/yr	0.0383 Gt-C/yr	0.90
F_W	0.0744 Gt-C/yr	0.0705 Gt-C/yr	1.06
F_{WM}	0.106 Gt-C/yr	0.0866 Gt-C/yr	1.22

CONCLUSION

We have investigated the spatial and temporal variations of k in the East Sea using the wind database compiled by Na *et al.* (1992). Three different relationships were adopted to examine the influence of parameterizations on gas transfer. We also attempted to explore the effect of time-averaged wind speed on k by feeding two wind data sets with time resolutions of 12 hours (short-term) and one month (long-term) into the adopted parameterizations. The monthly mean of CO_2 transfer velocities ranges from 12 to 32 cm/h in the East Sea. In addition to the threefold difference depending on the choice of parameterization, the long-term average of wind speed results in values of k up to 20% higher than short-term average of wind speed. The difference in mean transfer velocities between long- and short-term winds grows in winter because the assumption of Rayleigh wind distribution is not valid especially in that season. This study suggests that the time-averaged wind should overestimate (or underestimate) the gas transfer velocity in the East Sea, unless a new regional algorithm is employed. Synoptic and short-term wind measurements from satellite observations could constitute a proper replacement for long-term climatological wind measurements for gas exchange studies. The anuual flux of 0.071 Gt-C/yr using the parameterization of Wanninkhof (1992) corresponds to 3% of the global estimation of 2.1 Gt-C/yr (Donelan and Wanninkhof, 2002). Considering that the area of the East Sea is only 0.3% of the global ocean, this study suggests that the future studies should be focused on the accurate estimation of the CO_2 flux through the marginal seas.

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