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A Practical Bi-parameter Formula of Gas Transfer Velocity Depending on Wave States

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The parameter that describes the kinetics of the air-sea exchange of a poorly soluble gas is the gas transfer velocity which is often parameterized as a function of wind speed. Both theoretical and experimental studies suggest that wind waves and their breaking can significantly enhance the gas exchange at the air-sea interface. A relationship between gas transfer velocity and a turbulent Reynolds number related to wind waves and their breaking is proposed based on field observations and drag coefficient formulation. The proposed relationship can be further simplified as a function of the product of wind speed and significant wave height. It is shown that this bi-parameter formula agrees quantitatively with the wind speed based parameterizations under certain wave age conditions. The new gas transfer velocity attains its maximum under fully developed wave fields, in which it is roughly dependent on the square of wind speed. This study provides a practical approach to quantitatively determine the effect of waves on the estimation of air-sea gas fluxes with routine observational data.

Keywords:
· Gas transfer velocity,
· wind speed,
· wind wave,
· significant wave height.

1. Introduction

Various air-sea fluxes including momentum, heat, moisture and gas play a key role in air-sea interaction, and global climate change. The gas flux at the air-sea interface is typically expressed as the product of the gas transfer velocity k_L, solubility s, and the difference of the partial pressure of the gas such as CO_2 between air and water:

$$F = k_L s\left(P_{CO2w} - P_{CO2a}\right) \tag{1}$$

where P_{CO2w} and P_{CO2a} are the partial pressure of CO_2 in water and air, respectively. The air-sea momentum flux or wind stress at the sea surface (τ) can be expressed as:

$$\tau = \rho_a C_D U_{10}^2 \tag{2}$$

$$C_D = u_*^2 / U_{10}^2 \tag{3}$$

where u_* is the friction velocity of the air, ρ_a is air density. U_{10} is the wind speed at 10 m height above the sea surface in neutral stratification condition; C_D is the drag coefficient. Many studies have shown that air-sea exchange is regulated by turbulence associated with wind and wind waves at the air-sea interface (Jähne *et al.*, 1987; Komori *et al.*, 1993). However, it is often difficult to find a suitable parameter that is robust enough to describe turbulence intensity in natural environmental conditions. Alternatively, wind speed has been mostly chosen as the parameter since wind is the primary forcing of the air-sea boundary layer and easy to obtain from routine observational data.

In order to extrapolate fluxes over long time and space scales, gas transfer velocities are usually assumed to be a function of wind speed alone (Liss and Merlivat, 1986; Wanninkhof, 1992; Nightingale *et al.*, 2000a, b; Sweeney *et al.*, 2007). These relationships show a wide range of scatter, especially at high wind speed, and give rise to large discrepancies in the estimation of air-sea gas fluxes. Such a scatter could be caused by the uncertainties in the measurement of gas transfer velocities and in the determination of the wind speed. It could also be caused by other factors that influence gas transfers, but have not been taken into account. For instance, in addi-

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tion to wind, it is believed that wind waves and their breaking may also directly influence the air-sea boundary-layer processes (Monahan and Spillane, 1984; Jähne *et al*., 1987; Ocampo-Torres and Donelan, 1995). Thus, the effect of wave field on air-sea gas transfer should be considered in the parameterization of gas transfer velocity (Wanninkhof, 1992; Zhao *et al*., 2003; Woolf, 2005).

In the ocean, observations have shown that C_D is not a constant but highly variable. Jones and Toba (2001) presented a comprehensive review on various effects that can cause the scattering in the measurements of C_D. It has been assumed that the only systematic variation is with wind speed (Wu, 1980; Smith, 1980; Yelland *et al*., 1998).

Toba *et al*. (2006) suggested that the dynamical conditions at sea can be described by two nondimensional parameters in terms of wind waves: wave age β_* and windsea Reynolds number R_B or R_H. The wave age ($\beta_* = g/u_*\omega_p$) expresses the state of wind wave development. Here g is the acceleration due to gravity and ω_p is the angular frequency at the spectral peak of wind waves. The wave age can also be defined in terms of U_{10}, as $\beta = g/\omega_p U_{10}$. With the development of wind waves, the wave age and significant wave height (SWH) increase with fetch. A fully developed wave field has $\beta = O(1)$, which is usually less than 1.2 (Pierson, 1991; Jones and Toba, 2001).

The so-called windsea Reynolds numbers R_B and R_H, regarded as the fundamental parameters that control the behavior of air-sea transfers, are defined as:

$$R_B = u_*^2 / \omega_p \nu_a \, ; \quad R_H = u_* H_s / \nu_a \tag{4}$$

where H_s is the SWH of wind waves, ν_a is the air kinematic viscosity. Zhao and Toba (2001) collected a large amount of data, including a variety of wave states and wind speeds up to 20 m s^{-1}, and tested statistically a number of parameterizations. They showed that R_B and R_H are the best parameters among those tested to describe the whitecap coverage. Zhao *et al*. (2003) proposed a formula for gas transfer velocity as a function of R_B:

$$k_L = 0.13 R_B^{0.63} \tag{5}$$

where k_L is normalized to Schmidt number (Sc) of 660 in unit of cm h^{-1}. Woolf (2005) assumed that the contribution of waves to gas transfer velocity can be explicitly separated into two parts. From his equations (2), (4), (5) and (11), k_L can be expressed as:

$$k_L = 3.26 \times 10^{-4} R_H^{0.96} + 53.89 u_* \tag{6}$$

where k_L is in unit of cm h^{-1} for Sc = 660, and u_* in m s^{-1}. The first term on the right hand side of Eq. (6) represents the enhancement by breaking waves through bubble-mediated transfer. The second term is the contribution of non-breaking waves through the direct effect of wind shear on gas transfer.

However, R_B and R_H is difficult to determine from routine observational data due to the lack of information about u_* and ω_p, which severely limits the practical application of Eqs. (5) and (6). In this paper, replacing R_B and R_H, a new parameter R_{HU} which can be easily obtained from routine observational data is introduced to parameterize the gas transfer velocity. By adjusting the wave age, this approach is shown to be consistent in magnitude with the current parameterizations of gas transfer velocity under certain wave age conditions. In the condition of fully developed wave field, it also provides an upper limit of gas transfer velocity that approaches a quadratic dependence of wind speed.

2. New Parameterization in Terms of R_{HU}

In case of windsea, Toba *et al*. (2006) indicated that the existence of similarity laws implies that it is sufficient to select only one of the wave-property variables (ω_p or H_s), together with u_*, in order to completely describe the dynamical system. In terms of physical constants, the acceleration due to gravity g and the kinematic viscosity of air ν_a can be chosen to construct nondimensional variables. It does not need to consider the surface tension since it is only related to very high frequency waves. Therefore, Toba *et al*. (2006) constructed two fundamental nondimensional variables, R_B and R_H, to represent the dynamical processes near the air-sea interface. It is also quite reasonable to assume that u_* is equivalent to U_{10}, so R_H is proportional to $U_{10}H_s$. On the other hand, Woolf (2005) suggested that the dissipation rate is proportional to $U_{10}H_s$ if the energy input to waves that is related to the cube of wind speed. It is obvious that the dissipation rate dominates the turbulence near the air-sea interface. Therefore, in order to parameterize the gas transfer velocity from routinely available observations, a new parameter R_{HU} is introduced as:

$$R_{HU} = U_{10} H_s / \nu_a \, . \tag{7}$$

Similar to R_B and R_H, R_{HU} can be considered as a turbulent Reynolds number describing the turbulent intensity near the air-sea interface. The relationship between R_B and R_{HU} can be determined in two ways. For clarity, a parameter $b = R_B/R_{HU} = u_*^2/U_{10}H_s\omega_p$ is defined. The first method to quantify parameter b is to directly determine its value from observational data by the least square approach, in which case, the result is affected by the selected data. The second approach to derive b is from C_D

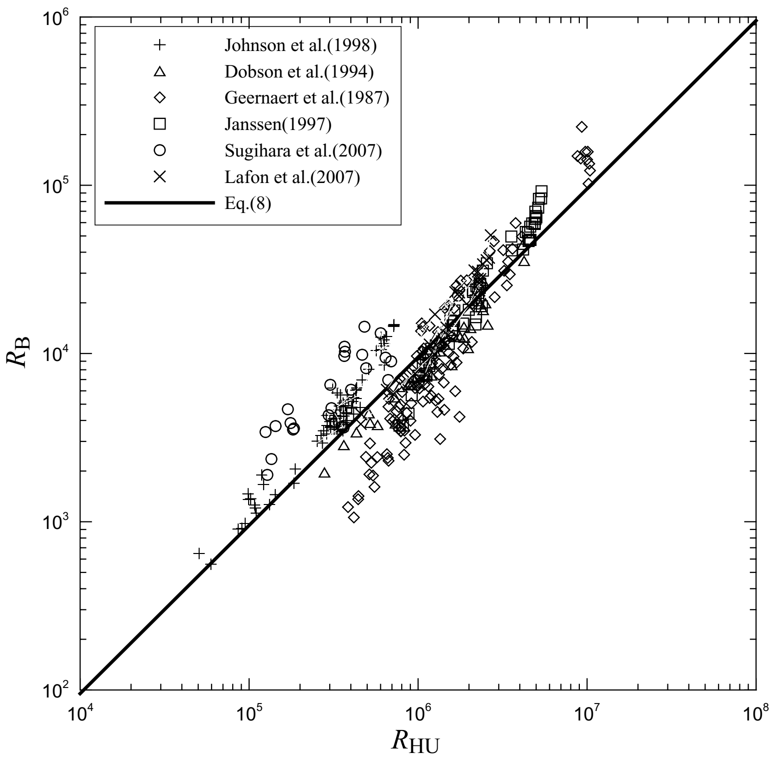

Fig. 1. Relationship between R_B and R_{HU} derived from the observational data. The solid line is Eq. (8) determined by the method of least square.

parameterizations and wind-wave growth relations, which are widely applied in wave studies.

Although many observations focus on the sea surface roughness or wind stress, only a few of them have measured wave parameters simultaneously. Some representative data obtained from field observations which contain information on waves are adopted in our analysis, as shown in Fig. 1. Without reduction in correlation coefficient (0.9), the relationship between R_B and R_{HU} can be expressed as:

$$R_B = 9.5 \times 10^{-3} R_{HU}. \quad (8)$$

Due to the high correlation coefficient, it is reasonable to conclude that R_B is in a linear relationship with R_{HU}, and parameter b can be taken as a constant. The parameter b will be discussed further below.

Based on observational data from laboratory and field programs, a large number of wind-wave growth relationships as a function of nondimensional fetch have been proposed. It is shown that these relationships are generally consistent with the Toba-3/2 power law (Toba, 1972) after eliminating the fetch (Guan *et al.*, 2004). Toba-3/2 power law is expressed as:

$$\frac{gH_s}{u_*^2} = B\left(\frac{gT_s}{u_*}\right)^{3/2}, \quad B = 0.062 \quad (9)$$

where T_s is the significant wave period, and B is an empirical constant. The relationship between T_s and ω_p can be written as $\omega_p = 2\pi/(1.05T_s)$ (Mitsuyasu, 1968). Equation (9) and the definitions of drag coefficient and wave age are used to rewrite b as b_1 in terms of drag coefficient and wave age. Therefore, the relationship between R_B and R_{HU} can be expressed as:

$$R_B = b_1 R_{HU} \quad (10)$$

where $b_1 = 1.11C_D^{3/4}\beta^{-1/2}$. Based on the field observational data from JMA (Japan Meteorological Agency) buoys, Zhao (2002) suggested that wave age is related to the nondimensional SWH via a 3/5 power law:

$$\beta = 2.56\left(\frac{gH_s}{U_{10}^2}\right)^{3/5} \quad (11)$$

Eq. (11) and the definition of drag coefficient are used to

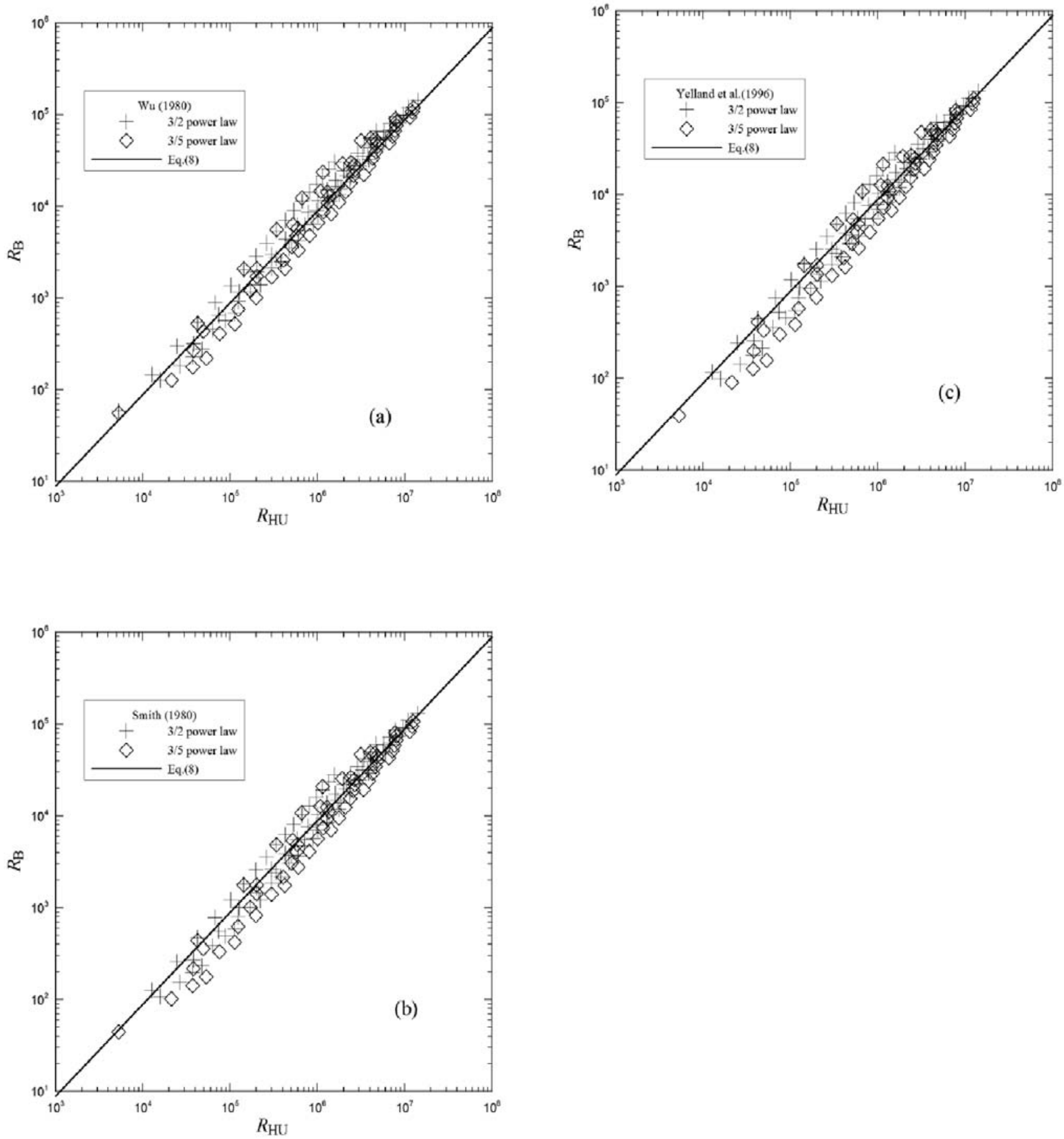

Fig. 2. Comparison of relationships between R_B and R_{HU} derived by 3/2 and 3/5 power law. From (a) to (c), C_D parameterizations used in calculations are Wu (1980), Smith (1980) and Yelland *et al*. (1998), respectively. Equation (8) is denoted as a solid line in the figures for comparison.

rewrite b as b_2 in terms of C_D and β. Thus R_B can be described by R_{HU}:

$$R_B = b_2 R_{HU} \tag{12}$$

where $b_2 = 4.79 C_D \beta^{-2/3}$. Although the proportionality factors b_1 and b_2 in Eqs. (10) and (12) are very different in form, it will be shown later that they are equivalent to each other in magnitude. It is also interesting to note that

Table 1. The values of b_1 and b_2 calculated from Eqs. (10) and (12) with three formulas proposed by Wu (1980), Smith (1980) and Yelland *et al.* (1998).

Authors	b_1 ($\times 10^{-2}$)			b_2 ($\times 10^{-2}$)		
	Max.	Min.	Aver.	Max.	Min.	Aver.
Wu (1980)	1.96	0.504	0.904	2.20	0.361	0.816
Smith (1980)	1.79	0.418	0.867	1.96	0.281	0.760
Yelland *et al.* (1998)	1.83	0.369	0.851	2.01	0.238	0.745

if it is taken $b_1 = b_2$, a relationship of C_D can be obtained as $C_D = 2.9 \times 10^{-3}\beta^{2/3}$, which predicts that C_D increases with the development degree of wind waves.

In order to quantitatively compare Eqs. (10) and (12), C_D must be specified first. Three representative formulas parameterized in terms of wind speed proposed by Wu (1980), Smith (1980) and Yelland *et al.* (1998) are employed in our calculations. At the same time, SWH must also be specified in the analysis. It is assumed that SWH can not be greater than that of a fully developed wave field that is specified by wind speed alone and independent of fetch. Following Carter (1982), the maximum of SWH is taken as:

$$H_{sm} = 0.025U_{10}^2. \tag{13}$$

Substituting Eq. (13) into Eq. (11), wave age $\beta \approx 1.1$, which agrees with the limitation suggested by Pierson (1991).

In order to compare Eqs. (10) and (12), wind speed U_{10} is specified varying from 1 to 20 m s^{-1}, H_s increases from $0.1H_{sm}$ to H_{sm} for each U_{10}, in which H_{sm} is determined by Eq. (13). Then R_{HU} can be calculated for ν_a = 1.53×10^{-5} m^{2}s^{-1} at 20°C. C_D is calculated from U_{10} for each of the three formulae proposed by Wu (1980), Smith (1980) and Yelland *et al.* (1998), and u_* is determined from Eq. (3). Wave age β is calculated from Eq. (11), which will then be used to determine ω_p from the definition of β. Finally, R_B can be calculated from u_* and ω_p. The comparisons between Eq. (10) and Eq. (12) are shown in Fig. 2 for the three C_D formulas stated above. Equation (8) is also shown in Fig. 2 for comparison. The representative values of b_1 and b_2 are depicted in Table 1. It can be seen that Eqs. (10) and (12) are quite consistent in magnitude, no matter which C_D formula is applied. This indicates that the two methods give similar results for a practical range of wave ages in determining the relationship between R_B and R_{HU}. It is also shown that both Eqs. (10) and (12) determined by this method agree with Eq. (8), especially at higher wind speeds. As shown in Table 1, the values of b_1 and b_2 vary within a relatively small range, and their average values are comparable in magnitude.

The coefficients of b_1 and b_2 are complicated parameters related to wind and wind waves. It is beyond the scope of this paper to discuss the details of these complex relationships. For simplicity, we assume that b_1 and b_2 can be approximately taken as a constant. Taking this constant as the average value of the six average values for b_1 and b_2 shown in Table 1, the relationship of R_B and R_{HU} can be expressed as:

$$R_B = 8.2 \times 10^{-3} R_{HU}. \tag{14}$$

Substituting Eq. (14) into Eq. (5), the gas transfer velocity can be parameterized by R_{HU} as:

$$k_L = 6.3 \times 10^{-3} R_{HU}^{0.63}. \tag{15}$$

By substituting the value of ν_a at 20°C, Eq. (15) can be further simplified as a function of $(U_{10}H_s)$:

$$k_L = 6.81\left(U_{10}H_s\right)^{0.63} \tag{16}$$

where H_s, U_{10} and k_L are in units of m, m s^{-1} and cm h^{-1}, respectively. Equation (16) shows that gas transfer velocity is proportional to the product of wind speed and SWH. For a given wind speed, it predicts that gas transfer velocity increases with SWH. In the open ocean, SWH can vary from several centimeters to a few tens of meters for different wave states. As a result, it leads to a significant difference in gas transfer velocity parameterizations between those that consider wave effect and those in which wave effect is neglected.

It must be kept in mind that the proportionality factor in Eq. (16) is highly uncertain. This uncertainty remains to be reduced by more observational data. Nevertheless, as will be discussed in the next section, Eq. (16) quantitatively agrees with various existing parameterizations by adjusting the wave age. Thus, Eq. (16) can be a practical way to consider the wave effect in the estimation of air-sea gas fluxes.

By using wind-wave growth relationships, the coef-

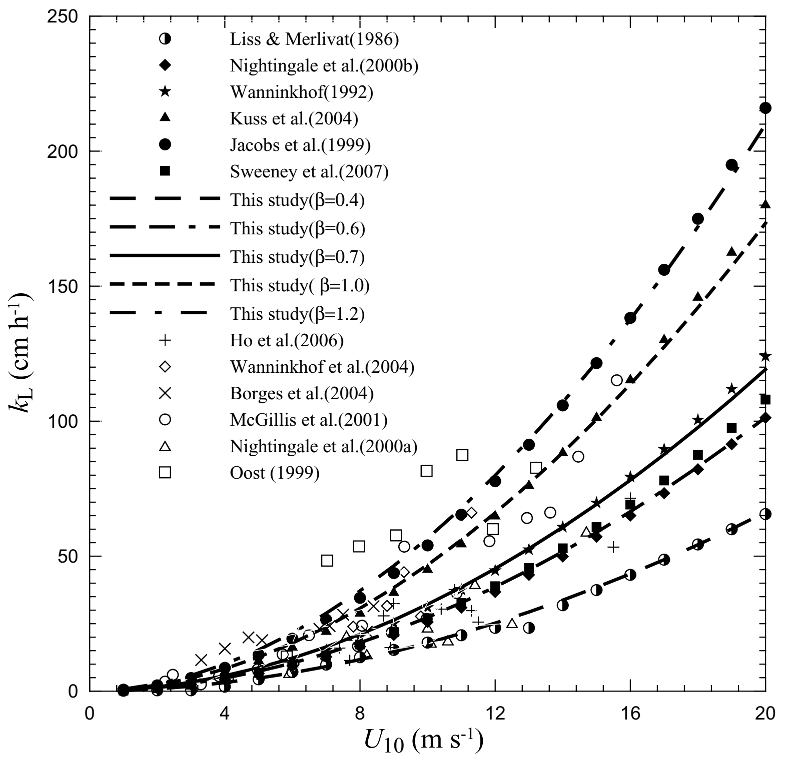

Fig. 3. Comparisons of gas transfer velocity of Eq. (16) at wave age of 0.4, 0.6, 0.7, 1.0 and 1.2 with other parameterizations in terms of wind speed. Some observational data are also plotted in the figure.

ficient b related R_B and R_{HU} can be written as some kind of combination of drag coefficient C_D and wave age β, such as b_1 and b_2, in which the parameters of U_{10} and H_s are not explicit. Although U_{10} and H_s can vary drastically in the field, the values of C_D ([0.5~1.5] × 10^{-3}) and β (0.1~1.2) are limited in a relatively narrow range, and their combinations will further reduce the variations of b_1 and b_2. As a result, U_{10} and H_s have little effect on the coefficient of b. It is also hopeful that a good average value of b is obtained in Eq. (14), so Eq. (16) is a robust representation of gas transfer velocity at various wave states.

3. Discussion

Figure 3 shows the comparison of Eq. (16) at various wave ages with some of the existing parameterizations in terms of wind speed. Some observational data are also shown for reference. It is evident that Eq. (16), at wave age β = 0.4, 0.6, 0.7, 1.0 and 1.2, is consistent with the relationships proposed by Liss and Merlivat (1986), Nightingale *et al.* (2000b), Wanninkhof (1992), Kuss *et al.* (2004) and Jacobs *et al.* (1999), respectively. Such agreements can be explained as follows.

The relationship of Liss and Merlivat (1986) was based on a combination of data obtained from a lake experiment and a wind/wave tank study. Due to the short fetches in lake and laboratory settings, young wave field with small wave ages is expected to be applicable to the relationship found in their study. At the same wind speed, their SWH is less than that at sea, leading to a low gas transfer velocity according to Eq. (16). The relationship of Liss and Merlivat (1986) gives the smallest values compared with the others for the same wind speed. Thus, the relationship of Liss and Merlivat (1986) is expected to agree with Eq. (16) at a small wave ages (e.g., β = 0.4) (Fig. 3).

Enhanced transfer might be expected in the open ocean in response to the occurrence of breaking waves in a more fully developed wave field. Wave age in the open ocean varies in a broad range, and is usually greater than that of lake and laboratory due to longer fetch. The relationship of Nightingale *et al.* (2000b) is a best fit to published dual tracer data obtained from the coastal and open ocean. In their figure 13 of Nightingale *et al.* (2000a), it is clearly shown that gas transfer velocity increases with SWH, which supports our argument. However, since no wave data in numerical form is provided in their paper, the value of SWH can only be roughly estimated from their figure. It is found that their wave ages range from 0.5 to 0.9. Thus, it is not surprising that their relationship

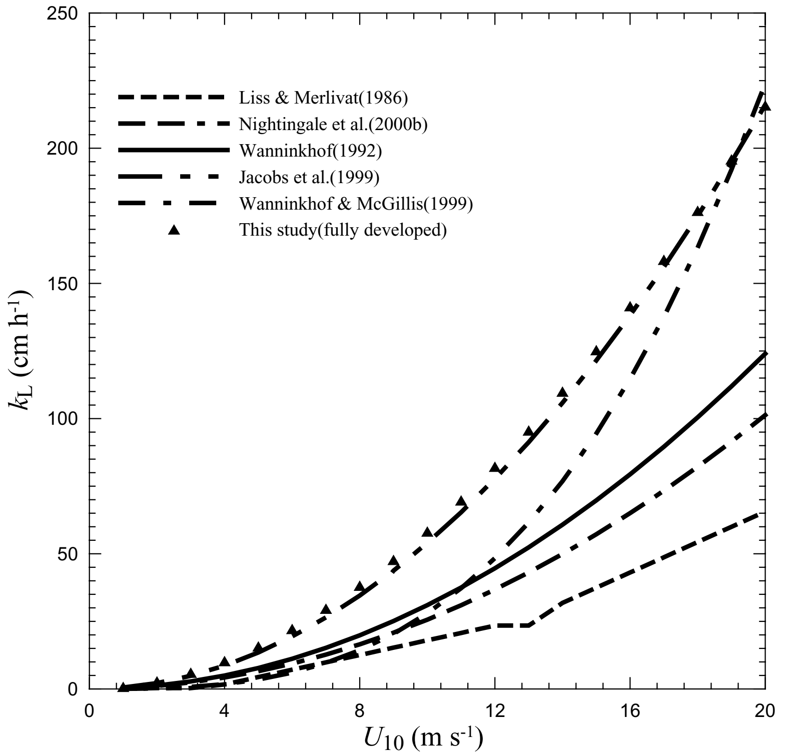

Fig. 4. Comparison of Eq. (17) as an upper limit of gas transfer velocity corresponding to the fully developed waves with some other parameterizations.

agrees well with Eq. (16) at a wave age of $\beta = 0.6$. The quadratic relationship between wind speed and gas transfer velocity proposed by Ho *et al.* (2006) obtained in the Southern Ocean is also well consistent with Eq. (16) at wave age $\beta = 0.6$ (not shown in the figure). As an interpretation of bomb ^{14}C measurements, Sweeney *et al.* (2007) proposed a relationship between gas transfer velocity and wind speed. As shown in Fig. 3, their result is slightly greater than Eq. (16) at $\beta = 0.6$.

The relationship of Wanninkhof (1992) is not directly associated with any particular experiments, but based on a modeled fit to the oceanic uptake of bomb-derived radiocarbon. It is surprising that his relationship is highly consistent with Eq. (16) at wave age $\beta = 0.7$. The observational data used by Kuss *et al.* (2004) was obtained in the eastern Gotland Sea (Baltic Sea). They did not provide wave information. Their relationship is consistent with Eq. (16) at wave age $\beta = 1.0$.

The relationship of Jacobs *et al.* (1999) was based on the data obtained in the North Sea during the air-sea gas exchange program ASGAMAGE. The detailed information about wave state was given by Oost *et al.* (2002). From their figure 10, it can be seen that their wave ages range mainly from 0.6 to 1.6, which indicates that the wave conditions in their study are near the fully developed waves. It is not surprising that the relationship of Jacobs *et al.* (1999) agrees well with Eq. (16) at wave age $\beta = 1.2$.

It is true that when the gas transfer velocities were measured in the field, especially for cases where dual tracer methods were used, they require some time during which wave age might vary. The same situation happened with regard to wind speed and any other environmental factors. Thus here assigned a certain value of wave age to the results of previous studies is just trying to describe the general wave states.

In the condition of fully developed wave field, substituting Eq. (13) into Eq. (16), we can obtain the upper limit of gas transfer velocity as:

$$k_L = 0.75U_{10}^{1.89}. \qquad (17)$$

The comparison of Eq. (17) with some other parameterizations is shown in Fig. 4. It shows that the observational data are smaller than those predicted by Eq. (17). It is also worth noting that Eq. (17) approximates to a quadratic dependence of wind speed that has been supported by past studies (Jacobs *et al.*, 1999; Kuss *et al.*, 2004; Ho *et al.*, 2006).

In practical application, the lower value from Eqs. (16) and (17) should be used in the estimation of air-sea CO_2 flux, and the formulae are for $Sc = 660$. They can be generally written as:

$$k_L = \min \begin{cases} 6.81(U_{10}H_s)^{0.63}(Sc/660)^{-0.5} \\ 0.75U_{10}^{1.89}(Sc/660)^{-0.5} \end{cases} \tag{18}$$

where "min" indicates gas transfer velocity will be chosen as the lower value of the two formulae.

We admit that there is great large uncertainty on the coefficient of $b = R_B/R_{HU}$ because it is determined with limited observational data, and some empirical relations that may only be valid in ideal conditions. For example, the drag coefficient of C_D introduced in this study contains the very controversial problem of wind-dependence of sea surface roughness. This ambiguity will certainly affect the accuracy of b. However, this approach provides a practical way to include the wave effect on gas transfer processes, and it can be improved in the future when more reliable observational data is available. With this bi-parameter formula of gas transfer velocity, it is hoped that the uncertainty in the estimation of CO_2 fluxes through the air-sea interface can be reduced to some extent.

4. Conclusions

The scope of this paper is limited to the consideration of the influence of wave state on gas transfer velocities. Although there is no doubt that other factors may affect the estimated transfer velocities, this study shows that variation in wave state is likely to be a major factor. With the application of Eq. (17) as an upper limit of gas transfer velocity, and SWH taken from the routine observational data, such as buoys, satellite altimeters and wave models, Eq. (16) can be used to estimate the gas transfer velocity no matter whether the wave state is wind wave or swell. With the combination of Eqs. (16) and (17), Eq. (18) is proposed as the final parameterization for k_L that can be used in general conditions with various Schmidt numbers. Although a thorough validation of this bi-parameter formula of gas transfer velocity remains to be carried out using observational data that include the information of wave state, it is shown that it can reconciles the differences among several existing parameterizations obtained at different wave states. This approach provides a practical, yet more accurate way to estimate air-sea gas flux by taking into account the effect of waves.

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