Air–Sea Gas Transfer: Mechanisms and Parameterization

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(Manuscript received 10 January 1995, in final form 4 October 1995)

ABSTRACT

The rate of air–sea gas transfer has been largely parameterized on the basis of studies performed in laboratory tanks: empirical formulas are proposed for three wind-velocity regions. Observed features of the transfer in these regions are associated with various wind, wave, and aqueous-flow conditions in straight and circulating tanks; some of these conditions do not even exist in the field. With these understandings, two formulas are proposed for the gas transfer: one for lakes and the other for the open sea; the rate is generally greater in the open sea.

1. Introduction

Gas transfer across the sea surface has long interested geochemists and oceanographers. Recent attention on the environment, such as the examination of global warming, has cast an urgent need for quantifying accurately the transfer velocity of liquid-phase controlled gases, including CO_2 (Dickinson and Cicerone 1986; Ramanathan 1988). Empirical formulas relating the rate of gas transfer to wind speed were deduced on the basis of two sets of laboratory observations (Liss and Merlivat 1986). The transfer rate was found to increase suddenly when capillary waves first appeared (Kanwisher 1963; Broecker et al. 1978); another sudden increase was observed at a higher wind speed, when the wave pattern changed from small, regular waves to rough, rippled, irregular waves (Jähne et al. 1979). As laboratory studies will continue to play a critical role in quantifying the air–sea gas transfer, the need to understand these phenomena is obvious.

Following studies on dynamic interactions in various wind–wave tanks (Wu 1975, 1978; Wang and Wu 1987; Tang and Wu 1992), the reported sudden increase of gas transfer at low wind speeds in a straight tank when capillary waves first appeared is suggested to be associated with the transition of the aqueous boundary layer from viscous to turbulent; the sudden increase at high wind speeds in a circulating tank when waves changed from smooth to sharply crested was caused by the removal of surface films due to wind mixing. Finally, the understanding gained from these laboratory observations is applied, along with investigations of wind wave breaking (Hwang et al. 1989) and air bubble production (Hwang et al. 1990) to parameterize all available measurements of air–sea gas exchanges; separate formulas are suggested for lakes and oceans.

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2. Gas-transfer phenomena observed in laboratory tanks

Laboratory observations on the gas transfer have constituted the basis of parameterizing the transfer rate across the sea surface. A formula consisting of three wind velocity regions was proposed by Liss and Merlivat (1986); each was suggested to be dominated by a different mechanism. These regions are divided by the following observed features, of which explanations are provided herein.

a. Sudden increase of gas transfer at low wind speed

A rapid increase in the gas transfer rate was identified in several laboratory studies; it coincided with the onset of waves on the water surface (Kanwisher 1963; Broecker et al. 1978). More specifically, Broecker et al. reported that following an almost unmeasurable rate at very low wind speed, the gas flux was strongly accelerated with the formation of capillary waves at a wind speed of about 2 m s^{-1}. Their results are reproduced in Fig. 1a, where K is the transfer velocity of gas and U the wind speed. Such an increase was first suggested to be caused by the increase of water surface area due to the presence of Crapper waves (MacIntyre 1971). Hasse and Liss (1980) doubted, however, whether these steep waves were sufficiently stable to introduce any significant effects. Liss (1983) then suggested that "capillary waves themselves may not be the cause of the observed enhancement, but may merely

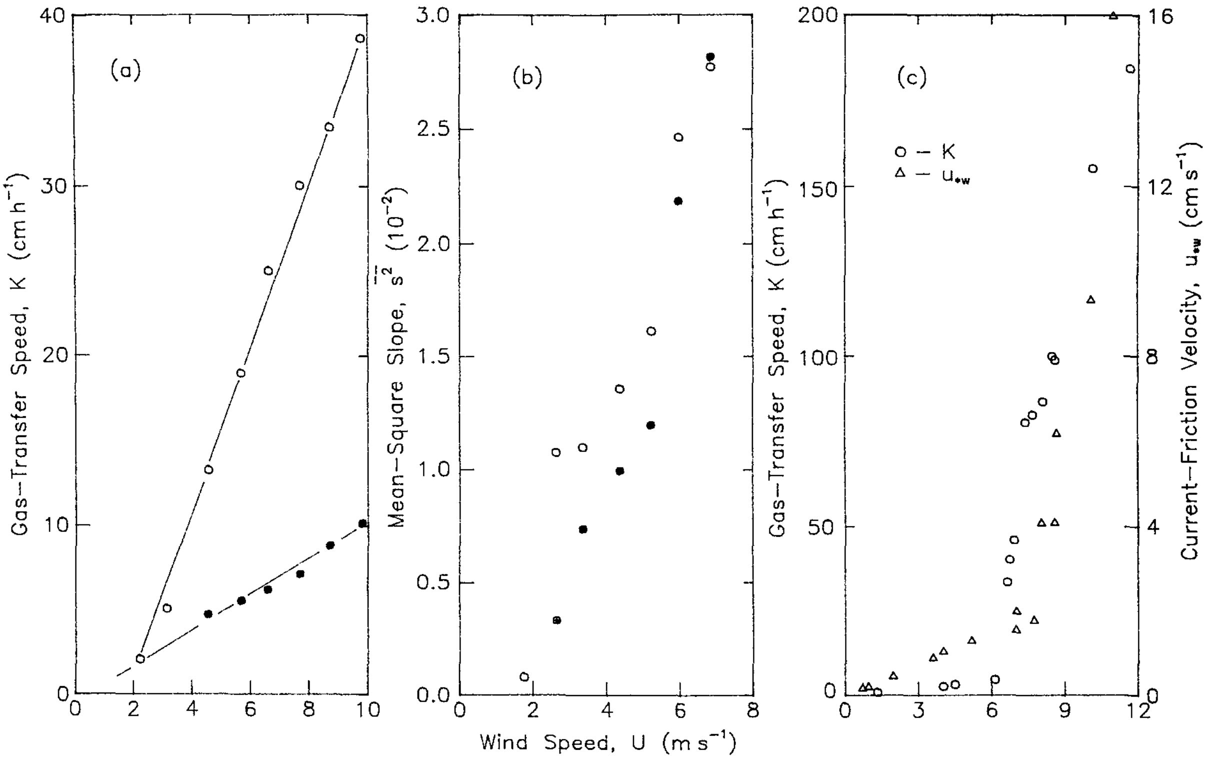

FIG. 1. Gas transfer rates and mean-square slopes measured in various wind–wave tanks. The data in (a) are reproduced from Broecker et al. (1979), in (b) from Tang and Wu (1992), and in (c) from Jähne et al. (1979); the open symbols represent clean-surface data and the solid symbols for slick-surface data.

act as indicators of a change in the nature of the gas transfer process in water close to the air–water interface.'' Subsequently, Coantic (1986) involved capillary waves in the surface renewal model, but found that this model could not provide an explanation for the enhancement. This result was further clarified by Hasse (1990), who pointed out the disparity between timescales associated with capillary wave motions and those with surface renewal processes. Note that all the experiments discussed above were carried out in straight wind–wave tanks.

Based on an earlier experiment on the inception of wind waves (Wu 1978), with simultaneous measurements of slopes of the wind-disturbed water surface and structures of the airflow boundary layer above it, the boundary layer at very low wind speed was considered to be in the pretransition regime (Wu 1968, 1975); its effective transition to fully turbulent occurred in the neighborhood of U_f = 2.5 m s^{-1}. The mean-square slope was negligibly small at very low wind speed (U_f < 1.6 m s^{-1}), increased almost linearly with wind speed in the range 1.6–2.5 m s^{-1}, and showed a more gradual increase beyond this range. The sudden rise of slopes at roughly U_f = 1.6 m s^{-1} was caused by the abrupt growth of initial waves (Ursell 1956). In the pretransition regime of the airflow boundary layer, these waves failed to grow in either length or slope with the wind speed.

In another study (Wu 1975), the transition of the aqueous boundary layer between various regimes was found to occur at a higher wind speed than that of the airflow boundary layer. In other words, the airflow at very low wind speed might already be turbulent while the aqueous flow remained viscous. This result is because not only the speed but also the kinematic viscosity of air are greater; both make the Reynolds number of airflows much greater than that of aqueous flows. In a straight (linear) tank, wind- and wave-induced downwind currents near the water surface introduce upwind (return) currents near the tank bottom. Conditions with and without bottom return flows were simulated by Wang and Wu (1987) in a circulating (oval shaped) wind–wave tank, through, respectively, the insertion and removal of a barricade on the water side. In both cases, the current was found to follow well the logarithmic distribution

$$U_w/u_{*w} = (1/\kappa)\ln(z/z_{0w}) \qquad (1)$$

in which U_w is the current speed at the depth z below the mean water surface, κ is the von Kármán universal constant, and z_{0w} is the roughness length. For the same wind speed, friction velocities were found to have nearly the same value for cases with and without return flows (0.76 and 0.80 cm s^{-1}), while the roughness length without return flows (1.02 mm) is much greater than that with (0.06 mm). This indicates that the transition of an aqueous boundary layer from viscous to turbulent regimes is unaffected by return flows since the transition is governed by the Reynolds number involving only the friction velocity. On the other hand, the structure of aqueous flows is much extended downward from the water surface in the case without a barricade, as the vertical coordinate is seen in Eq. (1) to be scaled by the roughness length. The condition without an aqueous boundary layer being influenced by return flows can still be realized even in a linear tank, when the water is deep enough to separate the boundary layer near the air–water interface from that along the tank bottom. A fully developed constant-flux layer was observed in the field to be about 100 cm deep (Churchill and Csanady 1983); it is thinner in a typical laboratory tank where it is thought to have a depth of about 50 cm (Wu 1975), but was fully developed in Wu's linear tank with 124-cm water depth. On the other hand, because of its shallow depth of 25 cm, the constant-flux layer was absent in Wang and Wu's (1987) circulating tank with a barricade. The water depth in Broecker et al.'s (1978) tank was about 50 cm; it might be too shallow to allow this layer to be developed.

Now, let us return to the sudden increase of gas transfer at low wind speed, when capillary waves first appear. Liss (1983) suggested that wave inception acted as an indicator of change in the nature of gas transfer processes in water. On the basis of results discussed above, we conclude that the change was very likely associated with the transition of the aqueous boundary-layer regime from viscous to turbulent. Thus, the observed sudden increase is due to a much more efficient transfer in turbulent aqueous flows. Although structures of these flows differ in various tanks having different water depths, our concern here is only the initiation of turbulent aqueous flows immediately below the interface.

b. Sudden increase of gas transfer at high winds

Experiments on the gas transfer and aqueous boundary layer were conducted by Jähne et al. (1979) in a circular wind–wave tank. As shown in Fig. 1c, the transfer rate was found to abruptly jump by about a factor of 5 at wind speed of about 8 m s^{-1}. A distinct change in the wave pattern was observed to accompany this jump; no small or regular waves were observed before the jump, whereas rough, rippled, irregular waves were observed afterward. In the same experiment, the friction velocity of aqueous flows (u_{*w}) was also measured, and was likewise found to manifest an abrupt change, as shown in Fig. 1c, at a wind speed similar to that that gave rise to the jump in the gas transfer rate.

Efficient damping of capillary waves by surface films observed in laboratory tanks and the field were reviewed by Wu (1971, 1989). Herein, the study of Tang and Wu (1992) is summarized, as their experiment was conducted in an oval-shaped tank having characteristics similar to that used by Jähne et al. (1979). In both experiments, the film on the water surface, if present, was able to circulate around the tank without being pushed to the downwind end and then mixed into the water body as in a straight tank. Equally important was the observation by Tang and Wu that the film could be produced by leaving either seawater or tap water in the tank overnight. In fact, the water surface in their experiment needed to be cleaned through an overflowing weir immediately before each clean-surface test. Slopes of the wind-disturbed water surface were continuously measured by Tang and Wu with an optical instrument. Their results, reproduced in Fig. 1b, indicate that at a wind speed of about 2.6 m s^{-1}, there was nearly a 70% reduction of the mean-square slope over a slick surface. At a wind speed of 6 m s^{-1}, results for clean and slick surfaces are seen in Fig. 1b to be quite similar, due to the disruption of surface films.

Rates of gas transfer in the presence of surface films were also measured by Broecker et al. (1978); see their results reproduced in Fig. 1a. Before the inception of waves, rates of gas transfer with and without films were reported to be identical. At higher winds, the rate of gas transfer is suppressed with the addition of films. In this case, capillary waves, which first appeared at wind speeds of 2–3 m s^{-1} without films, were absent even at higher wind speeds. Taking all these together, the sudden increase of gas transfer accompanied by the appearance of sharply crested waves at a wind speed of about 8 m s^{-1} in the circulating tank is suggested to be due to the removal of surface films. This effect would not occur in a straight tank, because films were pushed to the downwind end of the tank under much lower wind speed.

The above suggestion is also consistent with field observations of Barger et al. (1970); their results were interpreted to indicate that a surface film was disrupted at a wind speed of about 7 m s^{-1} (Wu 1971, 1989), which falls between the 6 m s^{-1} transition velocity that occurred in Tang and Wu's (1992) tank experiment and the 8 m s^{-1} transition velocity in Jähne et al.'s (1979). Being just a surface phenomenon, the breaking up of films is unrelated to the vertical structures of air and aqueous flows for which differences exist between laboratory tanks and the field. Such a disruption is also substantiated by one portion of Jähne et al.'s experiments; with the introduction of a dam, choppy waves started to appear at a much lower wind speed of about 3 m s^{-1}, and the transfer velocity varied continuously with the wind speed exhibiting no jump. As in our facility (Tang and Wu 1992), films were very likely present in Jähne et al.'s tank. The water surface remained covered by films without the dam, with a circulating water surface; it was cleared with the dam, with films being pushed to the downwind end. These reasonings are, of course, consistent with the similarity displayed between the gas transfer and aqueous boundary-layer results, Fig. 1c.

3. Quantitative parameterization of field results

a. Comments on current formulation

Formulas proposed by Liss and Merlivat (1986), widely used to estimate the rate of air–sea gas transfer, have the following three segments:

$$K = \begin{cases} 0.17U_{10}, & U_{10} < 3.6 \text{ m s}^{-1} \\ 2.85U_{10} - 9.65, & 3.6 \text{ m s}^{-1} < U_{10} < 13 \text{ m s}^{-1} \\ 5.90U_{10} - 49.3, & U_{10} > 13 \text{ m s}^{-1}, \end{cases} \quad (2)$$

where K is expressed in cm h^{-1}, and U_{10} in m s^{-1} is the wind speed at 10 m above the mean water surface. The division of the above region was adopted from laboratory observations; they are in order the so-called "smooth surface region" observed by Jähne et al. (1979), "rough surface region" by Ledwell (1984), and "breaking wave (bubble) region" by Broecker et al. (1978). The actual rate of transfer in the second region was deduced from field measurements of Wanninkhof et al. (1985) by assuming $U_{10} = 1.29U_1$, in which U_1 is the wind velocity at 1-m elevation.

Let us take a close look at the regions discussed above. The smooth-surface region at low wind speed was based on Jähne et al.'s (1979) observations with no more than a few waves in their tank. The gas transfer rate is very small in this region, where the airflow boundary layer might not even be turbulent. There is no such region in the field because of its large Reynolds number. This is true even at very low wind speeds over the sea surface ($U_{10} < 0.5$ m s^{-1}), under which the convective boundary layer prevails (Stull 1988). The mean shear stress associated with forced convective motions in this case may be negligible in comparison with the velocity variance associated with free convective motions; the latter actually govern the transfer process.

As for the division of the second and third gas transfer regions, waves start to break in large wind–wave tanks (Ursell 1956) at wind speeds lower than 13 m s^{-1} depicted in Eq. (2) for small tanks, and at an even lower speed of about 4 m s^{-1} in the ocean (Wu 1982). Breaking waves entrain air to produce bubbles, which actually influence the gas transfer (Merlivat and Memery 1983). The length of breaking waves in laboratory wind–wave tanks is also much shorter, by about two orders of magnitude, than that in the ocean. Furthermore, since the breaking wave crest length generally spans nearly one-half the tank width (Hwang et al. 1989), the ratio between this length and the wavelength is about an order of magnitude greater than that in the ocean. Both factors together make the breaking length per unit water-surface area in wind–wave tanks, and therefore the concentration of bubbles (Hwang et al. 1990), under the same wind speed orders of magnitude greater than in the ocean. Breaking waves in laboratory tanks are also much shorter in period, by about one order of magnitude (Hwang et al. 1989). Consequently, in addition to a much greater influence of bubbles per unit area of the water surface in laboratory tanks, the influence per unit time is also much greater than in the ocean. In terms of the bubble influence on gas transfer, laboratory conditions at wind speeds higher than 13 m s^{-1} may be matched by only those accompanying extreme storms in the field.

b. Summary of available data

From the above discussion, only a portion of the laboratory results, those in the second region shown in Eq. (2), are relevant for studying the air–sea gas exchange. There are a number of factors involved in scaling aqueous flows, which govern gas transfer, at various fetches (Wu 1988a). Inasmuch as there are still so many uncertainties in this scaling, especially between drastically different conditions in tanks and the field, it is advisable to use just the field results for determining the rate of gas transfer, with laboratory results providing the physical basis and the functional form for quantification.

The transfer velocity of radon was measured by Peng et al. (1979) during the Atlantic and Pacific cruises of the GEOSECS program. Portions of their data collected under steady winds required for the radon method were corrected by Deacon (1981) for water temperature effects to the 20°C condition. Later, similar measurements were performed by Kromer and Roether (1983) in the Atlantic Ocean, and by Hartman and Hammond (1985) in San Francisco Bay. Another experiment with radon was conducted by Glover and Reeburgh (1987) over the Bering Sea shelf, with data being averaged over two different periods. Wanninkhof et al. (1985) performed an experiment in Rockland Lake; escape rates of sulfur hexafluoride (SF_6) first dissolved in lake water were determined. Recently, Upstill-Goddard et al. (1990) conducted similar experiments with both CO_2 and SF_6 in two small lakes. Experiments were also carried out by Watson et al. (1991) in a shallow area of the southern North Sea off the Dutch coast; both SF_6 and ^{3}He were used. In all of these studies, the measured rates were transferred to those of CO_2 at 20°C water temperature according to Schmidt number scaling. We also converted wind speeds measured at different elevations to those at the standard anemometer height of 10 m above the mean water surface; the logarithmic wind-speed distribution and the wind stress coefficient proposed by Wu (1980) were used. All data converted as described above are compiled in Fig. 2a.

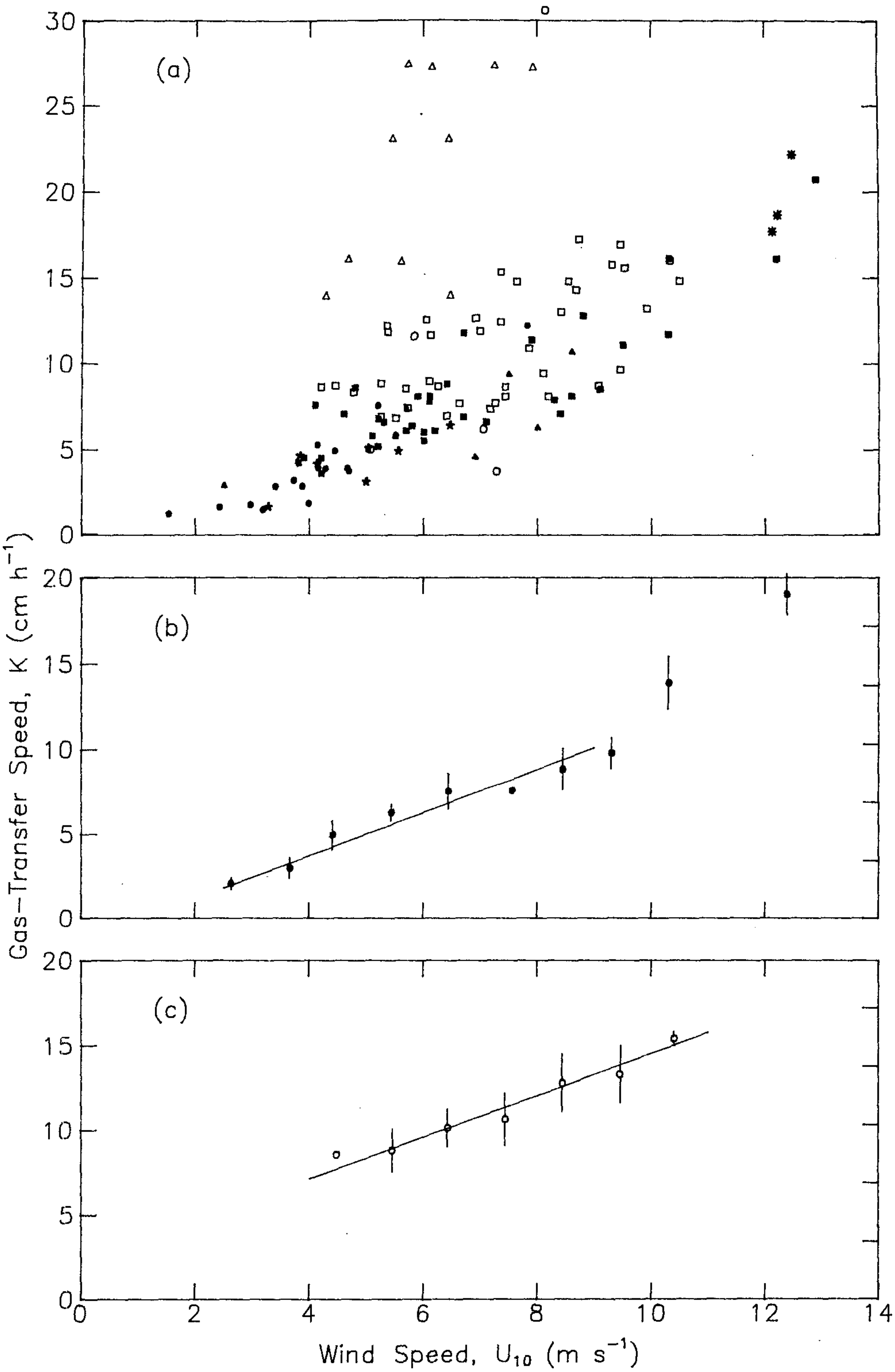

FIG. 2. Variation of gas transfer rate with wind speed. The data in (a) were obtained by: Wanninkhof et al. (1985) ◐, Peng et al. (1979) and Deacon (1981) □, Kromer and Roether (1983) ○, Hartman and Hammond (1985) ☆, Glover and Reeburgh (1987) △, Upstill-Goddard et al. (1990) lake ■, and pool ▲, and Watson et al. (1991) ✲. The band averaged lake results are presented in (b), and ocean results in (c).

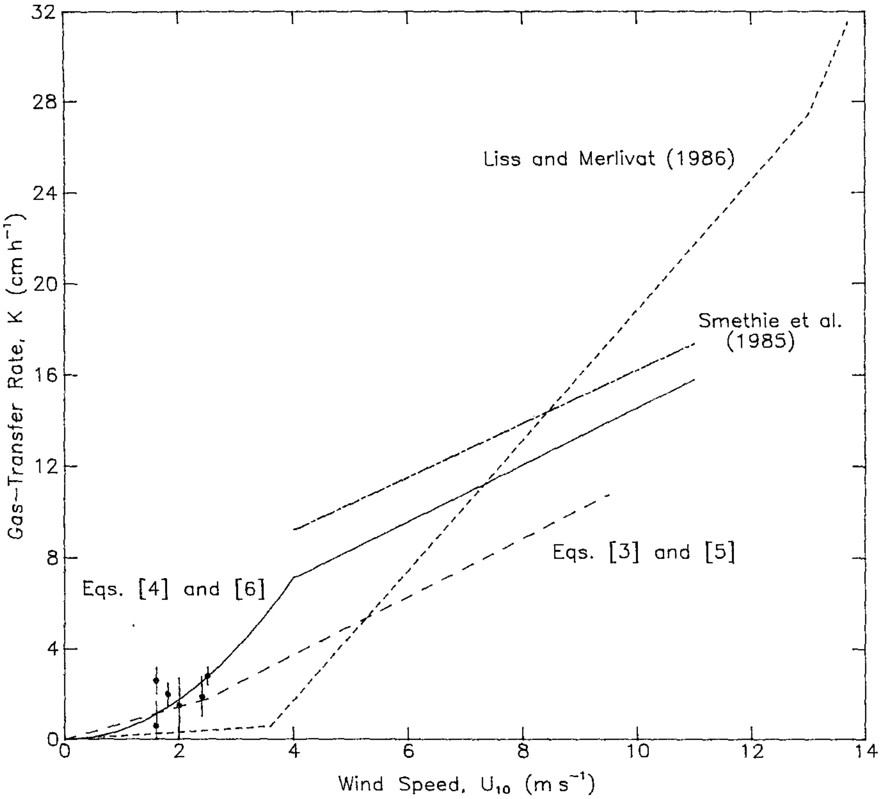

FIG. 3. Comparison of gas transfer formulas as are suggested by Liss and Merlivat (1986), Smethie et al. (1985), and in the present study: Eq. (3) for lakes and Eqs. (4) and (5) for the open sea; the data are from Clark et al. (1995).

c. Quantification of field results

The data presented by Glover and Reeburgh (1987) are seen in Fig. 2a to have a trend distinctly different from, and values much larger than, those of the others. Leaving out this set and one data point of Kromer and Roether (1983) outside the frame of Fig. 2a, the remaining bulk of the data are divided into two groups: those obtained in the open sea (Peng et al. 1979; Deacon 1981; Kromer and Roether 1983; Glover and Reeburgh 1987) shown as open symbols in Fig. 2a, and those from lakes, bays, and nearshore regions (Wanninkhof et al. 1985; Hartman and Hammond 1985; Upstill-Goddard et al. 1990; Watson et al. 1991) shown as solid symbols. The data in each group are then sorted into unit wind speed bands, and averaged. For both groups, the averaged data in each band and their standard deviation within the band are presented in Figs. 2b,c. Most interestingly, the data in each group are seen to be quite consistent, having rather small error bars.

Transfer rates for the open sea are seen in Fig. 2c to follow very well a linear variation with the wind speed, represented by the fitted straight line. The lake data in Fig. 2b can also be represented by a linear variation, with an upward turn at high winds. These two linear variations and their wind speed regions of application can be written as

$$\text{Lake:}\quad K = 1.28U_{10} - 1.38,\qquad 2.5\ \text{m s}^{-1} < U_{10} < 9.5\ \text{m s}^{-1} \tag{3}$$

$$\text{Ocean:}\quad K = 1.24U_{10} + 2.16,\qquad 4\ \text{m s}^{-1} < U_{10} < 11\ \text{m s}^{-1}. \tag{4}$$

The lake and ocean data are shown to differ in magnitude, but have nearly the same rate of variation with the wind speed. The gas transfer rate is justifiably greater in the open sea than in the lake, as a greater portion of the momentum flux from the wind is consumed at long fetches in generating aqueous flows, rather than waves (Wu 1988a).

Equations (3) and (4) are also diagrammed in Fig. 3, along with Eq. (2) suggested by Liss and Merlivat (1986). Referring to both Figs. 2b and 3, the results obtained in the lake, a small confined water body, appear to retain some characteristics of the gas transfer in laboratory tanks at low and high wind speeds. The transfer rate is insignificant at very low wind speed, and increases sharply with higher wind speed. Due to a large Reynolds number the main segment is pushed toward a wind speed lower than the 3.6 m s^{-1} shown in Eq. (2). The intensified rate, presumably due to bubbles, also occurs at a lower wind speed near 9 m s^{-1}, following effects of the fetch discussed earlier.

Let us now concentrate on the data collected under oceanic conditions. In this case, the fraction of momentum flux transmitted from the wind to aqueous flows increases with the wind speed at low wind speed (Wu 1988a), and reaches nearly a constant value for wind speeds beyond 4 m s^{-1} (Snyder et al. 1981; Wu 1988a). Consequently, it is feasible to represent the bulk of oceanic data for wind speeds greater than 4 m s^{-1} in Fig. 2c with a unique slope.

In the region of low wind speed ($U_{10} < 4$ m s^{-1}), because a smaller fraction of the momentum flux is transmitted to aqueous flows, the rate of gas transfer should be below the extension of the straight line toward lower wind speeds ($U_{10} < 4$ m s^{-1}). As shown in Fig. 2a, the data in this region follow an exponential variation with the wind speed suggested by Wanninkhof et al. (1985). Data were collected by Clark et al. (1995), especially for very low winds; see Fig. 3. Putting a greater emphasis on the well-established rates described by Eqs. (3) and (4) and taking into account the recent data, we propose to adopt the exponential function suggested by Wanninkhof et al. for ocean conditions and a simple linear function for lake conditions. The proposed functions as diagrammed in Fig. 3 can be written as

$$\text{Lake:}\quad K = 0.73U_{10} \quad U_{10} < 2.5 \text{ m s}^{-1} \tag{5}$$

$$\text{Ocean:}\quad K = 0.445U_{10}^2 \quad U_{10} < 4 \text{ m s}^{-1}. \tag{6}$$

Note that these low-wind speed segments cover nearly the same wind-speed region as, but provide much greater transfer rates than, that suggested by Liss and Merlivat (1986). At high wind speeds, there is no upward deviation seen; as discussed earlier, the breaking of ocean waves starts at a wind speed of about 4 m s^{-1}, and, more importantly, breaking ocean waves do not appear to exert an influence as drastic as that depicted in Eq. (2).

d. Comparison with other formulas

A formula for the air–sea gas transfer rate was also proposed by Smethie et al. (1985):

$$K = 1.167U_{10} + 4.55. \tag{7}$$

It is diagrammed in Fig. 3 for the wind-speed region covered by their data. This formula is seen to agree well in trends with the expressions proposed herein, but differs from those by Liss and Merlivat (1986). The cause of the difference in the magnitude between Smethie et al.'s and the present formulas is not clear, but the sets of data selected here appear to have much less scatter than do those of Smethie et al.

The formula of Liss and Merlivat (1986) was derived from the measurements of Wanninkhof et al. (1985), which can be seen in Fig. 2a to have been made mainly at wind speeds below 5.5 m s^{-1} with only one data point at 7.8 m s^{-1}. In other words, all the data were obtained below the wind speed at which Liss and Merlivat's formula intersects with the present expression for oceans as shown in Fig. 3. A formula was also proposed recently by Wanninkhof (1992) that follows well the expressions suggested by Liss and Merlivat (1986), as both proposals were based on the same dataset reported by Wanninkhof et al. (1985). This set of data is represented quite well by the present lake expression.

4. Concluding remarks

Two interesting coincident changes of gas transfer behaviors and surface wave patterns were observed in laboratory wind–wave tanks of different configurations, linear and oval shaped. Fortunately, physical processes of wind–wave interaction have been studied in tanks of both shapes (Wu 1975, 1978; Wang and Wu 1987; Tang and Wu 1992) to explain the probable causes of these changes. Our interpretations further indicate that the gas transfer is closely related to the aqueous boundary layer regime as well as the characteristics of the water surface, as first suggested by Liss (1983). Wind and wave, as well as aqueous-flow conditions in the ocean, differ greatly from those in laboratory tanks; differences also exist between those in contained (lakes) and open (ocean) water bodies. Our investigations on breaking waves (Hwang et al. 1989) and entrained air bubbles (Hwang et al. 1990) further indicate that only laboratory results at intermediate wind speeds were obtained under aqueous-flow conditions similar to those in the ocean. This condition actually consists of the principal region in the formulation of Liss and Merlivat (1986); it becomes the sole region in the ocean. Formulas are herein provided for all wind-speed regions.

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