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Measurements of gas exchange and momentum transfer in a circular wind–water tunnel

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ABSTRACT

Gas exchange and momentum transfer have been studied by means of a small circular wind–water tunnel. Gas transfer velocity and friction velocity first grow linearly with wind speed. At a critical wind speed, with the onset of rough waves, both increase abruptly. The experiments clearly show the enhancement of air–water exchange processes by waves. Experimental conditions were varied–nitrogen/carbon dioxide atmosphere, temperature, circulation of water unconfined/confined by a dam–and found to influence gas exchange. This, and the differing results of various authors demonstrate that gas exchange is not a function of wind speed alone, nor of friction velocity, but that further parameters, such as wave spectra, have to be considered as well.

1. Introduction

The transfer of gases of low to moderate solubility is controlled by diffusion through a thin boundary film in the liquid. It is primarily wind speed which controls the thickness of this film. The exact form, however, of the function gas exchange rate vs wind speed is not known up to now. Although laboratory experiments cannot directly be transferred to natural conditions, they allow specific investigation of the role of individual parameters and thus may lead to a better understanding of the mechanisms controlling gas exchange.

Previous laboratory experiments were usually performed in linear wind–water tunnels (e.g. Downing and Truesdale, 1955; Kanwisher, 1963; Hoover and Berkshire, 1969; Liss, 1973; Broecker et al., 1978). Flothmann et al. (1979) reported first results obtained by means of a small circular wind–water set-up. We present here further results, including measurements of momentum transfer.

2. Experimental techniques

2.1. *Circular wind–water tunnel*

The circular wind–water tunnel used in the present experiment (see Fig. 1) has a total diameter of about 75 cm and contains an annular water channel of 10 cm depth, 10 cm width and 40 cm inner diameter. Wind is produced by means of a rotor with four blades arranged at right angles. The system is covered by a gas-tight container and flushed by a dry gas—nitrogen or carbon dioxide in the experiments described here. Total gas volume is about 150 l, water volume 11 l (for a water depth of 7 cm). The whole set-up is situated in a heat-insulated box which can be temperature controlled between 0 °C and 40 °C. Temperatures of water and gas are measured with platinum resistors. Wind speed is assumed to be equal to the velocity of the rotor blades above the center of the water channel. Water velocities are determined by means of a hot-film anemometer probe.

As a result of circumferential continuity of the water surface the wave pattern and the wind profile are homogeneous along the channel, whereas in linear systems, hydrodynamic conditions—and therefore also gas exchange rate (H. C. Broecker, private communication)—change with fetch. Conditions of linear wind tunnels can partly be simulated in our system by introducing a dam into the water channel that acts as a beach.

The specific geometry of our wind tunnel leads to centrifugal forces causing radial velocity compo-

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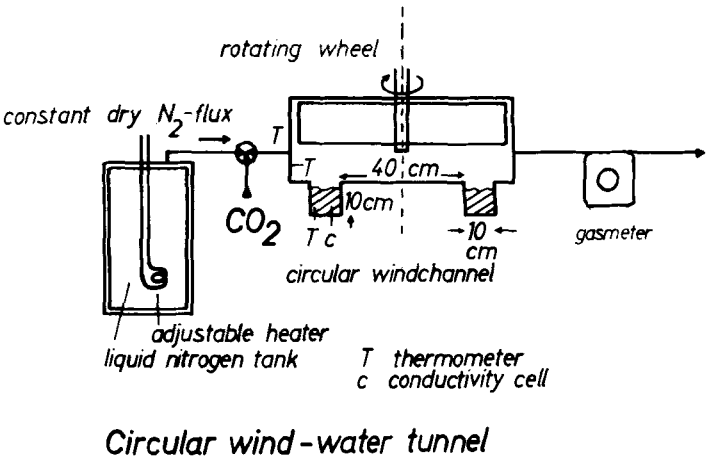


Fig. 1. Experimental set-up for gas exchange measurements with the circular wind–water tunnel. The container can be flushed both with dry nitrogen and CO_2.

nents near the interface, directed towards the center in the gas and outwards in the liquid phase. The velocity field is similar to that in a rotating fluid over a flat disc, described in Schlichting (1965). Above the viscous boundary layer in the order of 1 mm thickness, measurements with a hot-wire anemometer probe show constant wind speed rather than a logarithmic wind profile. The helical water flow implies a steady surface renewal which may enhance gas transfer.

2.2. *Gas exchange*

Gas exchange can be characterized by a transfer velocity w (cm/s), defined by

$$w = j/\Delta c \tag{1}$$

Δc = concentration difference between two specified levels (moles/cm^3);
j = gas flux density (moles/cm^2/s).

In the present experiments, except those for zero wind speed, the bulk of the water can be assumed to be well mixed; furthermore, for CO_2 (the gas we used in our experiments) the transfer resistance essentially is situated in the liquid boundary layer, and the concentration gradient in the gas phase is negligible. Natural reference levels for the transfer velocity therefore are the bulk water and the very surface of the water which is in equilibrium with the gas phase, so that $\Delta c = \alpha c_g - c_w$. The change with time of the CO_2 concentration in the water is then given by

$$H \frac{dc_w}{dt} = w(\alpha c_g - c_w) \tag{2}$$

where

H is the average depth of water (=volume/surface area);
c_w and c_g are the concentration in bulk water and gas, respectively; and
α is the solubility of CO_2 in the water.

We performed two types of gas exchange experiments using de-ionized water in all cases. For the *evasion* experiments water loaded with CO_2 was used and the chamber was flushed with nitrogen, at a nitrogen flux large enough (3 to 60 l/min) so that the CO_2 concentration in the gas phase could be neglected ($c_g \cong 0$). The CO_2 content of the water then simply decreases exponentially with time towards zero:

$$c_w = c_{w0}\, e^{-t/t_0},$$

with a decay time

$$t_0 = H/w. \tag{3}$$

In a few experiments gas invasion was studied. The water was initially CO_2-free ($c_w(t = 0) = 0$), while the chamber was filled with pure CO_2, so that

$$c_w = \alpha \cdot c_g\,(1 - e^{-t/t_0}).$$

So, by measuring c_w as a function of time, the transfer velocity w can be determined from eq. (3) both, for evasion and invasion.

The CO_2 concentration was determined by measuring the electrical conductivity which is proportional to $c_w^{1/2}$. This follows from the chemical equilibrium $[HCO_3^-] \cdot [H^+] = k_1 \cdot [CO_2]$ and the charge neutrality, $[H^+] \cong [HCO_3^-]$ since pH $\lesssim 5.5$ at the CO_2 concentration used. From this we obtain $[H^+]^2 = k_1 \cdot [CO_2]$. The conductivity is proportional to $[H^+]$ and therefore to $[CO_2]^{1/2}$. In de-ionized water and for the relatively high CO_2 concentration the amounts of bicarbonate and carbonate ions are negligible compared to the physically dissolved carbon dioxide, and we need not consider them for determining the transfer velocity (Bolin, 1960; Skirrow, 1975).

2.3. *Momentum transfer*

Momentum transfer was measured in the following way. Wind stress on the surface causes a drift velocity of the bulk of the water, and momentum conservation is given by

$$A_s H \frac{d\rho u_b}{dt} = A_s \tau_s - A_w \tau_w$$

where u_b is the bulk velocity of water, ρ is the density of water, A_s, A_w are the area of water surface and of walls, respectively, and τ_s, τ_w are the shear stress on water surface and walls, respectively.

$A_s \tau_s$ is the momentum influx from gas to water, $A_w \tau_w$ is the outflux to the channel walls, and both are equal at steady state. When the wind is turned off, say at time 0, wind stress decreases abruptly from τ_s to zero, and we have

$$\tau_s = \tau_w(t=0)\, A_w/A_s = H\rho \frac{du_b}{dt} \qquad (t=0) \qquad (4)$$

from which the friction velocity in the water $u_* = \sqrt{\tau_s/\rho}$ is found. Our velocity measurements are not very accurate, so the absolute values for u_* may have an appreciable error.

2.4. *Wave pattern*

For studying the *wave pattern* a He–Ne laser was mounted below the channel, the beam of which penetrates the water vertically and is refracted at the surface. The horizontal deflexion of the beam, observed on a screen in the top cover of the chamber, is proportional to the slope of the surface. So far, only qualitative observations have been possible, but the method will be developed for the determination of wave spectra.

3. Results

3.1. *Gas exchange with water, momentum transfer*

The transfer velocities as a function of wind speed obtained for different experimental conditions are summarized in Figs. 2 to 4. Different experimental conditions lead to distinctly different results. A quantitative explanation of the results is not yet possible at this stage; they will be discussed in qualitative terms, and the findings may indicate the direction for further research.

In *evasion* experiments the gas exchange rate increases linearly with wind speed up to about 8 m/s (Fig. 2). At this wind speed, the transfer velocity jumps abruptly by about a factor 5 and then increases further very rapidly with wind speed, again more or less linearly within the error limits (Fig. 3). Parallel to the discontinuity in gas exchange a distinct change in the wave pattern is observed, from no waves or small, regular waves (wavelength several cm, amplitude $\lesssim 0.2$ cm) to rough, rippled, irregular waves with amplitudes up to several cm (see Figs. 7, 8). In a few runs just below the critical wind speed, the wave pattern was unstable, changing from a smooth to a rough surface; the gas exchange rate varied correspondingly.

Friction velocity, like gas transfer velocity increases proportionally (Fig. 5) to wind speed up to a critical wind, and much faster beyond this (see also next paragraph). The highest observed friction velocity (in water) was about 15 cm/s at a wind speed of 10.9 m/s. The corresponding bulk water velocity was nearly 40% of the wind velocity, while below the jump the water moved at only about 3–4% of the wind speed. Interestingly, the wind–water velocity different ΔU was approximately constant, about 7.3 m/s above the jump. This shows the fast growth of wind–water-coupling. The drag coefficient measured at 0.1 m height $C_{0.1} = (u_{*g}/u)^2$ has a constant value of 6.3×10^{-3} for small wind velocities. Beyond the discontinuity it increases rapidly up to 170×10^{-3}.

When changing from evasion to invasion, i.e. from a nitrogen to a carbon dioxide atmosphere, the

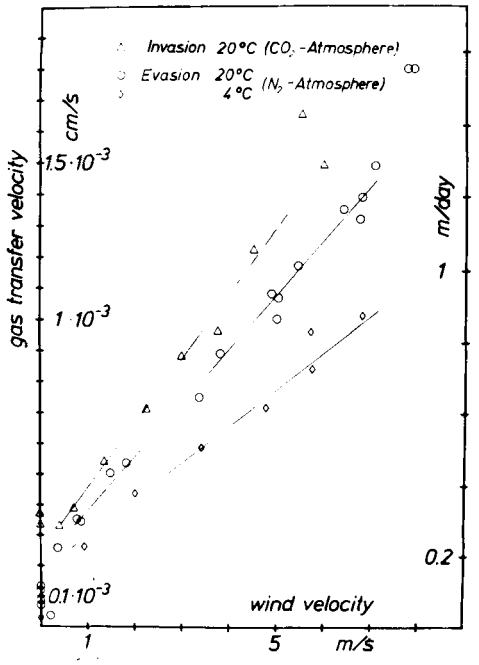


Fig. 2. CO_2–gas transfer velocity at low wind speed, Data for evasion experiments (N_2–atmosphere) at 4 °C and 20 °C and invasion experiments (CO_2–atmosphere) at 20 °C. Straight lines are least square fits of the data.

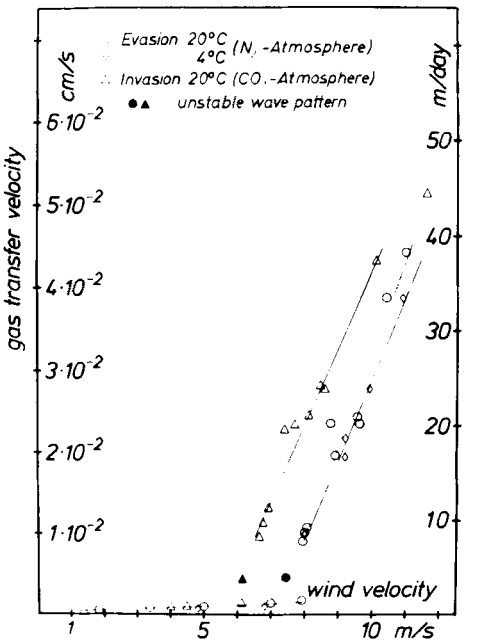


Fig. 3. CO_2–gas transfer velocity at high wind speed for the same experimental conditions as in Fig. 2. A periodic change of the wave pattern from rough to smooth surface was observed at the filled points.

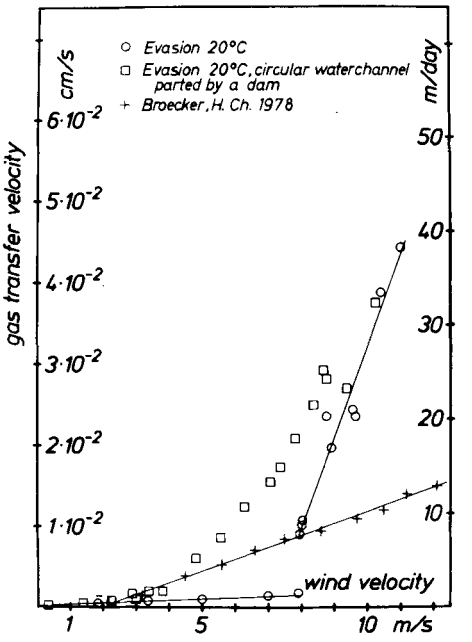


Fig. 4. Effect of fetch: Evasion results with water channel parted by a dam. Compared with results obtained with unconfined water channel and data of H. Ch. Broecker (1978) obtained in a linear wind–water tunnel.

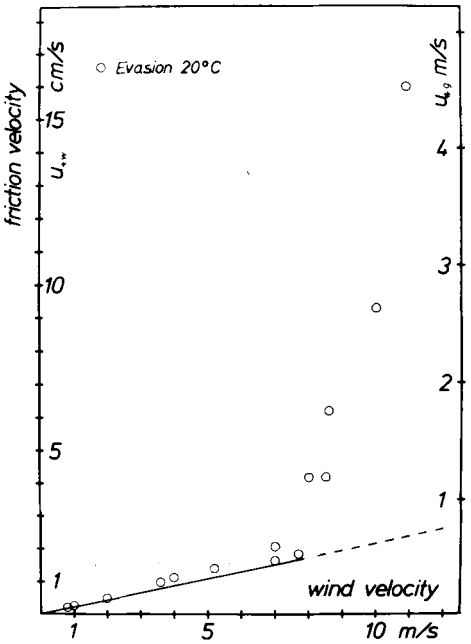


Fig. 5. Friction velocity u_* as a function of wind speed in the evasion experiments at 20 °C. Compare Fig. 3 for the gas exchange values at the same experimental conditions. Straight line theoretical values for smooth surface (see text).

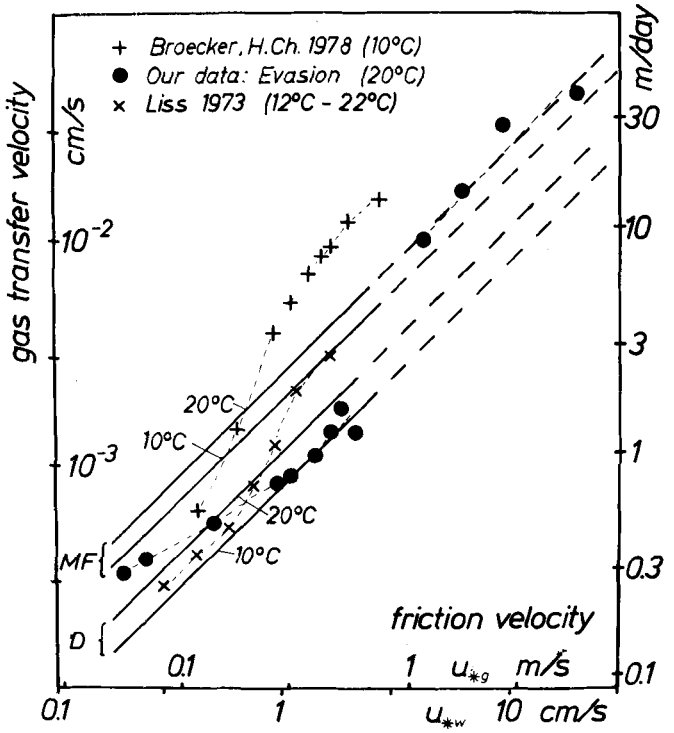
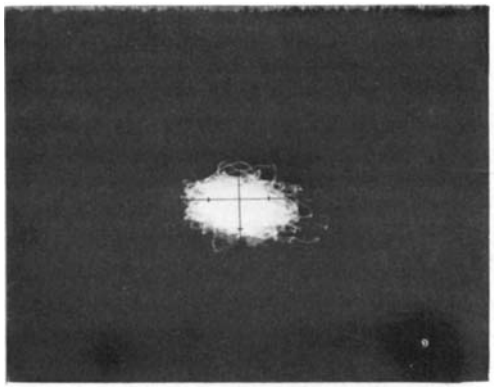
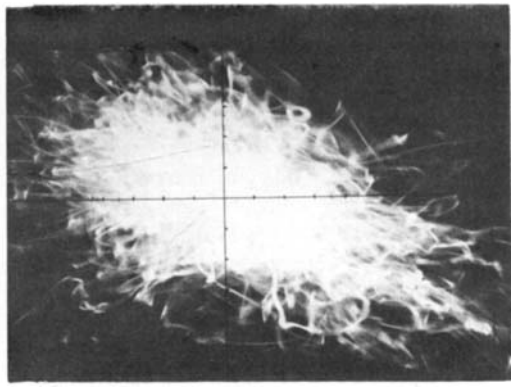


Fig. 6. Gas transfer as a function of friction velocity, as obtained by different authors. Friction velocity for the data of Liss as indicated by Deacon (1977), and personal communication by Broecker for his data. The gas transfer values of Liss were measured with oxygen. Straight lines: Theoretical results according to Münnich and Flothmann (1975) and Deacon (1977) indicated by MF and D, respectively.





Figs. 7 and 8. Photographs of the deviation of a helium–neon-laser beam through the inclination of the water surface by waves just before (Fig. 7, 7.8 m/s wind) and beyond (Fig. 8, 8.0 m/s) the discontinuity. Exposure time is 10 s, scale units 5°, x-axis parallel to wind, blowing from left to right.

discontinuity occurs already at $\Delta U \cong 6.0$ m/s instead of 7.3 m/s. No significant effect of water temperature is found; a rough wave pattern appears at the same wind speed at 4 °C as it does at 20 °C (with a nitrogen atmosphere in either case). One possible explanation for the dis-

continuity is the appearance of a Kelvin–Helmholtz instability (Lamb, 1932) which is expected to occur at a critical relative velocity

$$\Delta U_{crit} = (2/\rho_g)^{1/2} (g\rho_w \sigma)^{1/4} \quad (5)$$

where ρ_g, ρ_w are densities of gas and water, g in the gravitational acceleration, and σ is the surface tension. The calculated critical velocities are 6.8 m/s for nitrogen and water, and 5.4 m/s for CO_2 and water, respectively. The agreement with the experimental values is reasonable, mainly when considering that the actual wind velocity may be slightly smaller than the indicated value valid for the rotor blades. Eq. (5) gives a 2% lower critical velocity at 4 °C than at 20 °C, which is not in conflict with observation within the experimental uncertainty. This agreement is, however, no proof that we actually observe a Kelvin–Helmholtz type instability. The results are compatible, too, with the hypothesis of a definite critical shear stress, for which, by means of eq. (7) below, we calculate a critical wind velocity ratio of 0.83 for a CO_2 and a N_2 atmosphere. Experimentally we find a value of 0.82. The experimental data are not sufficient to determinate the nature of the discontinuity.

Francis (1949) studied wave formation in annular channels of different sizes similar to ours. He found that "there is an abrupt change of state of the waves at some limiting velocity in the range 24–27 feet/s (7.3–8.2 m/s), when the waves become rough and rippled", which was obviously the same phenomena we are observing.

Wu (1969) analysed oceanic data and found a discontinuity in drag coefficient and surface roughness at a wind velocity of 15 m/s, measured at 10 m height. He suggested a different explanation for that discontinuity, namely an increase in the wind velocity beyond the average wave phase velocity.

At zero wind speed, gas exchange is appreciably faster than for pure molecular diffusion into or out of the liquid. By surface cooling, evaporation leads to a gravity instable surface layer and thus induces convection (Flothmann et al., 1979). The very low gas exchange observed at $T = 4$ °C, where the water density has its maximum and where thus $d\rho/dT = 0$, supports this explanation. For invasion experiments at zero wind speed we find a higher gas exchange rate than for evasion. This is probably also due to an unstable stratification. A density measurement of water saturated with CO_2 at 1 atm., 20 °C, in fact showed a density increase of 0.04 ± 0.01%, equivalent in size with a 2 °C lower temperature.

3.2. *Alcohol–water mixtures*

Bohr (1900) reports that CO_2 transfer is several times higher in ethanol than it is in water. We did evasion experiments with an ethanol–water mixture (8% ethanol by weight) and found 3 to 20 times larger CO_2 transfer velocities than with pure water. Rough, irregular waves appeared already at about 2–3 m/s wind; no drastic change in the wave pattern was found with increasing wind. So far, however, we have not been able to get reproducible results with an alcohol–water mixture.

There are various possible reasons for the difference between water and the ethanol–water mixture. The formation of capillary waves depends on surface tension. An 8% ethanol solution has a surface tension of 50 dynes/cm instead of 72 dynes/cm for pure water which may be a reason for the earlier occurrence of waves and the higher gas exchange values.

In addition the evaporation rate is higher for the alcohol–water mixture because of the higher vapor pressure which leads to a greater evaporation-induced instability. This effect is further increased by the occurrence of a concentration gradient near the surface due to preferential evaporation of ethanol.

3.3. *Experiments with finite fetch*

When the water in the channel is divided by a beach, rough waves appear already at 3 m/s, and the transfer velocity as a function of wind speed exhibits no further jump, but is similar as observed in linear channels (Kanwisher, 1963; Broecker et al., 1978) (Fig. 4). This can be interpreted such that when fetch is limited, wave formation (and, therefore, a marked rise in gas exchange) occurs for lower wind velocities than in the unconfined circular channel. The same is true under natural conditions and has been explained by considering the influence of pressure fluctuations and viscosity (Phillips, 1957; Miles, 1957). The reason why the water surface is more stable against wave formation in the undivided circular channel is not yet clear. Possibly, it is because here, the water can circulate freely, while the disturbance of the flow with a beach or in a linear set-up may lead to an earlier appearance of waves. Rotational forces may also be of influence in the circular system.

4. Comparison with models

4.1. *Gas transfer*

Theoretical approaches have been presented by Münnich and Flothmann (1975) and by Deacon (1977), linking gas exchange to momentum transfer on the basis of the universal velocity profile over a hydrodynamically smooth surface. Both models predict that transfer velocity w is proportional to friction velocity and can be represented by

$$w = \beta^{-1} \left(\frac{D}{\nu}\right)^n u_{*w} \tag{6}$$

β is a constant, D diffusivity ν kinematic viscosity, and u_{*w} friction velocity in water. Münnich and Flothmann fitted the sub-logarithmic part of the universal profile by a surface-renewal model (Danckwerts, 1970) and obtained $n = 1/2$ and $\beta = 16$. For Schmidt numbers $\nu/D > 10$ Deacon gave a somewhat different relation which can be approximated well by eq. (6) with $n = 0.67$ and $\beta = 12.2$. Our evasion results at 20 °C without beach (friction velocity was measured only for these conditions) are compared with both models in Fig. 6.

Our experiments at different temperatures allow us to compute the exponent n in eq. (5). For CO_2 in water, D/ν changes by a factor of 2.43 ± 0.12 from 4 to 20 °C, and the ratio of slopes (dw/du) for low wind velocities and smooth surface (for which the models are only appreciable) is 1.44 ± 0.11 (Fig. 2) which corresponds to $n = 0.41 \pm 0.09$. Slopes are considered instead of absolute values because the non-zero transfer velocity at $u = 0$ is probably caused by an independent mechanism such as evaporation cooling. Taking absolute values instead of slopes would give an even smaller value. The exponent $n = 1/2$ assumed by Münnich and Flothmann therefore agrees with these experimental results, whereas Deacon's model better reproduces the absolute values (see Fig. 6).

The data of Broecker et al. (1978) and Liss (1973) are also shown in Fig. 6. At constant u_{*w} the gas transfer velocity data differ by up to one order of magnitude for different experimental conditions, so friction velocity alone is not sufficient to describe gas exchange, and other factors must also be taken into account. In our results, there is a discontinuity in the relation between w and u_*, the ratio w/u_* being higher by a factor 3 above the critical wind speed than below. Above the discontinuity there must be an additional process for mass transfer by which momentum transfer is not (or less) affected. Presumably this additional mechanism is related to the formation of capillary waves. This idea is supported by the findings of Kanwisher (1963) and Broecker et al. (1978) who both reported that a marked effect of wind velocity on gas exchange began only above some critical velocity where also capillary waves commenced. McIntyre (1971) proposed enhancement of gas transfer due to surface dilatation by ripples. This however can probably not fully explain the high gas transfer rates.

4.2. *Friction velocity*

Münnich and Flothmann (1975) showed that the universal velocity profile near a smooth wall, in the sublogarithmic range, can be fitted by

$$u(z) = \beta \cdot u_* \cdot (1 - e^{-z/\bar{z}})$$

where $\bar{z}$ is $\beta \cdot \nu/u_*$. Since the wind velocity is constant in our channel above the viscous boundary layer, we can apply this velocity law to determine the friction velocity in air as a function of wind speed u_∞:

$$u_{*g} = u_\infty/\beta$$

With

$$u_{*w} = \sqrt{\rho_g/\rho_w}\, u_{*g} \tag{7}$$

one obtains for the friction velocity in water $u_{*g} = 2.2\ 10^{-3}\ u$, which is about 20% less than observed experimentally for the smooth surface up to the discontinuity. To see a possible influence of centrifugal forces for momentum transfer, we calculated the torque exerted on a fluid by a rotating disc according to Schlichting (1965). From his results we obtain a corresponding shear stress proportional to $\rho \cdot \nu^{1/5} \cdot u_\infty^{g/5}$ and for the friction velocity $u_* = c \cdot u_\infty^{0.9}$ with $c = 4.2 \cdot 10^{-3}$ (cm/s)$^{0.1}$.

This is a nearly linear relation, and for $u_\infty = 5$ m/s we get $u_* = 1.1$ cm/sec or 20% less than the experiment. Both theoretical approaches give identical results. This could mean that the centrifugal forces do not seriously influence momentum transfer through the viscous sublayer. One would then expect little influence on gas transfer as well.

In invasion experiments with a CO_2 atmosphere u_{*w} is expected to be 1.20 times higher than with an N_2 atmosphere (evasion) and at same wind

speed (see eq. (7)). This is essentially because of the higher density of CO_2. One finds in fact a slope of gas transfer velocity against wind speed 1.19 ± 0.05 times higher for invasion than for evasion (see Fig. 2) as predicted by eq. (6). The discussion shows that the experimental results below the discontinuity are well predicted by theory assuming a smooth water surface. Beyond the discontinuity, the surface is rough and momentum transfer is enhanced by pressure forces.

5. Conclusions

Gas transfer depends strongly on the wind velocity, but a unique relationship obviously does not exist, neither with wind velocity directly nor with friction velocity. From hydrodynamics, gas transfer velocity w should be considered as a function of friction velocity u_* rather than of wind speed. For a smooth surface, models relating w to u_* describe reasonably well the observations. Our results further clearly show the importance of wave formation. Wind induced waves, particularly those of short wavelength and high slope, obviously enhance surface renewal. Therefore phenomena such as surface roughness and wave spectra must also be considered for understanding gas transfer.

The observed discontinuity in the plot of transfer velocity against friction velocity (Fig. 6) shows that there are at least two regimes governed by different mechanisms. The nature of the discontinuity and the rapid increase of transfer rates beyond it are only partly understood so far.

Data comparison shows the strong influence of the specific experimental conditions. Obviously, laboratory results cannot simply be applied to natural conditions until the relevant processes are better understood. Most probably in nature gas transfer increases more than proportional with wind speed, although the often quoted, quadratic relation cannot be inferred from the available data. Laboratory experiments predict a numerical value for the ratio of gas transfer velocity w to friction velocity from $1 \cdot 10^{-3}$ (smooth) to $1 \cdot 10^{-2}$ (rough sea) for CO_2 at 20 °C. (For other gases and temperatures one will get slightly different values according to eq. (6).) This does not include possible effects of spray and gas bubbles for strong winds. Detailed studies, both in the laboratory and in the field (Roether and Kromer, 1978; Peng et al., 1979) are necessary for a better understanding of gas exchange.

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ИЗМЕРЕНИЯ ОБМЕНА ГАЗОМ И КОЛИЧЕСТВОМ ДВИЖЕНИЯ В ЦИРКУЛЯРНОЙ АЭРОДИНАМИЧЕСКОЙ ТРУБЕ

С помощью небольшой циркулярной аэродинамической трубы изучался обмен газом и количеством движения. Скорость переноса газа и скорость трения сначала растут линейно со скоростью ветра. При критической скорости ветра, когда возникают крутые волны, обе скорости возрастают скачком. Эксперименты ясно указывают на усиление волнами процессов обмена между воздухом и водой. Условия экспериментов менялись—азотно-углекислая атмосфера, температура, циркуляция воды, беспрепятственная и с препятствием в виде плотины—и было найдено, что при этом газообмен менялся. Эти и различные результаты других авторов показывают, что газообмен не является функцией только скорости ветра или скорости трения, но что и другие параметры, такие, как спектры волн, должны быть также рассмотрены.