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LABORATORY MEASUREMENTS OF MASS TRANSFER OF CARBON DIOXIDE AND WATER VAPOUR FOR SMOOTH AND ROUGH FLOW CONDITIONS

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MANAGEMENT PERSPECTIVE

The exchange of mass and heat across air-water interfaces is an important process in geochemical global cycles. A better understanding of the corresponding exchange coefficients and their dependence upon the relevant physical phenomena is urgently needed in order to address the problem of properly parameterizing and predicting large scale processes that influence our environment. The transfer of carbon dioxide is of particular importance since it plays a major role in the global warming process. This paper describes some laboratory measurements of the mass transfer of carbon dioxide and water vapour and the determination of the corresponding exchange coefficients. The wind speed dependence of these coefficients is for the first time revealed from experimental work. The accuracy of the measurements is unprecedented.

SOMMAIRE À L'INTENTION DE LA DIRECTION

Le transfert de masse et de chaleur à l'interface air-eau est un processus important des cycles géochimiques à l'échelle de la planète. Il est urgent de mieux comprendre les coefficients de transfert correspondants et leur dépendance à l'égard des phénomènes physiques pertinents afin de mieux aborder la question d'une bonne paramétrisation et d'une bonne prévision des processus à grande échelle qui agissent sur notre environnement. Le transfert de gaz carbonique est particulièrement important étant donné qu'il joue un rôle primordial au niveau du réchauffement de la planète. Le présent document décrit quelques mesures expérimentales du transfert de masse du gaz carbonique et de la vapeur d'eau et de la mesure des coefficients de transfert correspondants. C'est la première fois que des expériences permettent d'indiquer la dépendance de ces coefficients à l'égard de la vitesse du vent. La précision de ces mesures est remarquable.

ABSTRACT

Mass transfer of carbon dioxide (CO_2) and water (H_2O) has been measured in a 32 m wind-wave tunnel, with a beach in the middle providing two sections of 16 m fetch. The wind speed (referred to 10 m) range covered was 1 to 24 m/s and the roughness Reynolds number (in the air) varied from 0.1 to 100. These correspond to aerodynamically smooth flow to rough flow. A minimum in the Dalton number for both CO_2 and H_2O is revealed for the first time. The minimum Dalton number, at wind speeds between 2 and 3 m/s, indicates a characteristic common point in the transfer mechanism for both gas and aqueous phase transfer controlled constituents. This point corresponds to the occurrence of the first wind generated wavelets.

RÉSUMÉ

Le transfert de masse du gaz carbonique (CO_2) et de l'eau (H_20) a été déterminé dans un tunnel de houle-vent de 32 m, avec une plage au centre formant ainsi deux fetchs de 16 m. La plage de vitesses du vent (établie à 10 m) était comprise entre 1 et 24 m par seconde, et le coefficient de rugosité de Reynolds (dans l'air), entre 0,1 et 100. Ces valeurs correspondent à un écoulement lisse aérodynamique et à un écoulement rugueux. Une valeur minimale du nombre de Dalton, pour le CO₂ et H₂0, est indiquée pour la première fois. Cette valeur minimale, pour des vitesses du vent comprises entre 2 et 3 m/s, révèle un point commun caractéristique du mécanisme de transfert des composantes, aussi bien celles dont le transfert est contrôlé dans la phase gazeuse que celles dont le transfert est contrôlé dans la phase aqueuse. Ce point correspond à la manifestation des premières vaguelettes générées par le vent.

INTRODUCTION

The transfer of mass, energy, and momentum across the air-water interface is a very important path in the global cycling system. Atmosphere and ocean act as a coupled thermodynamical system, in which the response of one phase to the forcing imposed by the other leads to variation in the transfer rates. The core of the problem of parameterizing the transfer process across the air-water interface is a phenomenological description of the way the resistance to transfer behaves. The resistance is largely in thin diffusive sublayers, on both sides of the interface. Gases of low solubility in water (like carbon dioxide) have their exchange controlled mainly in the water phase, while for those with high solubility or high reactivity the transfer is controlled in the air phase.

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In spite of numerous laboratory and field investigations on CO_2 and moisture transfer processes, their behaviour is not yet satisfactorily understood. A complete phenomenological description is still lacking. For gases whose transfer is mainly controlled in the water phase a characteristic response of the gas transfer velocity to the wind speed has been noticed since the first laboratory experiments. A linear increase (for low wind speeds), and an abrupt enhancement (for higher) in both the gas transfer and friction velocities as a function of wind speed were reported from measurements in a small circular wind-wave tunnel (Jähne et al., 1979). The gas transfer velocity enhancement was initially associated with the influence of small capillary waves. Further work showed that a more appropriate quantity to parameterize the transfer velocity is the mean square slope of all wave components (Jähne et al., 1984).

Empirical relations between wind speed and transfer velocity of carbon dioxide (CO_2) for instance, are already being used to estimate the CO_2 fluxes globally, from satellite wind data (Etcheto et al., 1991). Some measurements of air-sea exchange have been performed in rough and stormy seas (Watson et al., 1991). While they support the wind dependence suggested by Liss and Merlivat (1986), the number

of data points reported (only four) do not show much statistical significance. However, more accurate transfer coefficients are needed in view of the sensitivity of the coupled system to CO_2 concentrations. Furthermore, it has been demonstrated that the exchange coefficients cannot be described solely by the wind field (Jähne, 1991). Wave-related roughness as an obvious characteristic of a wind disturbed water surface has to be incorporated in the transfer description (Kitaigorodskii and Donelan, 1984).

 CO_2 transfer experimental results have been typically reported in terms of the actual mass transfer velocity K_{CO_2} (Liss, 1983). More recently, a nondimensional transfer velocity K_+ related to the friction velocity in the water has been introduced and found to correlate well with the mean square slope of the waves (Jähne et al., 1987). For the range of friction velocities explored, K_+ was found to increase with the friction velocity reaching a constant value for the highest friction velocities.

In the present work, for the case of CO_2 we decided to introduce a coefficient equivalent to the Dalton number in moisture transfer processes, which is essentially the ratio of the transfer velocity to the wind speed. We therefore will refer to the exchange coefficients D_{CO_2} (= K_{CO_2}/u) and D_{H_2O} (= K_{H_2O}/u) as Dalton numbers for the CO_2 and moisture transfer processes respectively.

There have been some suggestions indicating that the exchange coefficients (Dalton number for water vapour and Stanton number for heat, for instance) should vary with wind speed. The evaporation of water droplets in the near surface layer may be one of the causes for a significant enhancement of the Dalton number. Statistical analysis of the problem of spray evaporation (Bortkovskii, 1987) shows that the Dalton number should increase by a factor of two from 9 to 18 m/s wind speed. However, no apparent variation in the Dalton number has been observed in field measurements. An average value of 1.2×10^{-3} has been reported (DeCosmo et al., 1988) for wind speeds ranging from 7 to 14 m/s, during the Humidity Exchange over the Sea Main Experiment (HEXMAX). During the same experiment, for wind speeds from 4 to 18 m/s other researchers (Smith and Anderson, 1988) determined that the average

Dalton number was also 1.2×10^{-3} with values varying from 0.8×10^{-3} to 2×10^{-3} .

Although much progress has been achieved in the last 20 years or so, the apparent differences between theoretical approaches and experimental results regarding the dependence of the exchange coefficients on the wind speed urges some further considerations. An appropriate parameterization of the transfer velocity as a function of wind and wave parameters as well as water turbulence information is urgently needed (Jähne et al., 1989). This goal can only be reached with a better knowledge of the physics of the transfer process in the near surface layers.

The objective of the present work is to advance our knowledge on the nature of the transfer process across the air-water interface, in particular of H_2O and CO_2 . With an appropriate range of wind speeds, it will be shown that the exchange coefficients (Dalton numbers) depend on the wind speed, and their dependence is associated with the different flow regimes and relevant physical phenomena involved in the transfer processes across the air-water interface. As the main resistance for the transfer of H_2O is in the air phase, while for CO_2 it is in the water phase, a joint investigation will possibly shed some light on the relative importance of the different physical mechanisms involved.

2 THE EXPERIMENTS

2.1 <u>The Gas Transfer Flume</u>

The Gas Transfer Flume (GTF) of the Hydraulics Laboratory at the National Water Research Institute (see figure 1) has a test section that is 32.2 m long and 0.76 m wide; the water depth can be up to 0.25 m while the air duct is 0.60 m high (Merzi et al., 1990). It may be operated either open ('ventilating mode') or closed ('circulating mode'). The experiments reported in this paper were conducted in the circulating mode; the flume is tightly closed and the air circulates continuously. A vaneaxial direct drive fan driven by a 25 hp variable speed motor is capable of deliv-

ering wind speeds up to 22.5 m/s measured on the centerline of the tunnel. The total air volume in the flume is about 66 m^3 , while the water volume is 10 m^3 . A 2 m^3 tank ('tail box') at the downwind end of the test section and a smaller one at the upwind end ('head box') are connected by a 0.28 m diameter pipe which has an inline pump used to circulate and mix the water. The current induced by this pump in the test section may be varied continuosly from 0 to 0.60 m/s.

There are three instrumented measuring stations in the GTF at 5.3 m, 14.5 m, and 29.7 m from the upwind end of the test section respectively, each with a profiling system. Sensors at the three stations used in this work are: a) 0.2 mm diameter capacitance wires to measure wave height, and b) Pitot tubes to measure wind speed. At station 1, a relative humidity sensor and thermistors for air and water temperature were also used. The sampling ports for CO_2 measurements in water and air and for water vapor were also at station 1. A differential, non-dispersive, infrared analyzer -NDIR (LI-6262, LiCOR)- was used for the measurements of CO_2 and H_2O . In this instrument the concentration mesurements are based on the difference in absorption of infrared radiation passing through the sample and reference cells. At station 3, a laser slope gauge was used to measure the along-wind and cross-wind wave slopes.

An *inverted beach* was installed just upwind of station 2 in the GTF. The beach consisted of an aluminum plate with several layers of wire grids beneath. The plate is installed with a negative slope so that its upwind edge is about 5 cm above the still water surface and its downwind edge about 5 cm below. The plate spans the width of the test section and is 76 cm long. Waves approaching this beach loose their energy in the turbulence generated by interaction with the grid and are further damped by the rigid lid. This obstacle to waves divided the effective fetch in two for the experiments reported here, in such a way that the measurements at stations 2 and 3 correspond to a fetch of approximately 1 m and 16 m respectively, while those at station 1 are referred to the original 5.3 m fetch. The future use of more inverted beaches will enable us to

generate various wind wave fields (with different mean square slopes) for a particular wind speed. The analysis of the dependence of the mass transfer velocities on the mean square wave slope will be explored in a subsequent work. Two main advantages of the beach characteristics are the following: a) while it dissipates the wave energy, the breaking-generated turbulence does not enhance the mass transfer velocities as the contact with the air flow above is eliminated in the dissipation region, and b) the flat aluminum plate facing the flume top does not appreciably obstruct the wind flow.

During the experimental runs to determine the mass transfer velocity of H_2O and CO_2 , wave height η and reference wind speed u (measured at the air duct centerline approximately 30 cm above mean water level) were also measured at the three stations. The analog signals were digitized and recorded in a personal computer (IBM 286) at 20 Hz. Water and air temperature were monitored and recorded at 1 Hz. During separate runs wind speed profiles were obtained at the three stations, to estimate the friction velocity u_* , and the roughness scale z_o . Wind wave slopes were also monitored at station 3.

2.2 Wind Profiles and Wave Slope Measurements

Wind speed measurements as a function of height from the surface were carried out at the three stations for reference wind speeds up to about 16 m/s. The Pitot tube profiler systems sampled at levels 5 mm apart from the centerline downwards, at a sampling rate of 20 Hz for 10s at each level. Simultaneously, wave height was measured at the three stations. The friction velocity u_* and the roughness scale z_o were estimated and they are presented as a function of the reference wind speed uin figures 2 and 3. Due to the beach installed just upwind, station 2 has the shortest fetch and so the lowest friction velocity and roughness scale for a particular wind speed are observed at this station. A better representation of the wind field is achieved by a fetch-averaged friction velocity which is shown in figure 4 as a function of the measured

wind speed.

The relation between the roughness scale and the the root-mean-square of the surface displacement (η_{rms}) due to the waves is shown in figure 5. According to Kitaigorodskii (1968), a linear dependence is expected for fully rough flow. This is seen to occur at all stations for wind speeds in excess of 7 m/s.

Some runs were performed to measure wave slope at station 3. The meansquare of the along-wind (η_x) and cross-wind (η_y) wave slope are presented in figure 6. It is observed that the initial wavelets begin to appear at 2 m/s wind speed, although they can not be observed through the elevation measurements since a capacitance wire system was used. A sharp increase in the along-wind mean square slope occurs from 2 to 6 m/s wind.

2.3

Experimental Procedure for CO₂ Transfer Measurements

The water is impregnated with CO_2 by bubbling into the tail box while the circulation pump is operated at a rather high speed (40 to 50 cm/s) thereby rapidly mixing the entire water mass. To avoid any CO_2 concentration build up in the air during this water impregnation procedure, the flume flaps were open (ventilating mode), and the wind was maintained at low speed to keep the concentration at the ambient laboratory level as the initial condition. Input of CO_2 for about 15 min was observed to be sufficient for the concentration in the water to be near the upper limit of the NDIR analyzer.

The source of CO_2 to the tail box was shut off and once the water was well mixed (water phase CO_2 signal decreasing steadily for several minutes), the flume flaps were closed and the wind was set to a specified speed, to begin the experimental run.

A timed value was connected to the analyzer to allow sampling the air and water phase intermittently. The value switches from air phase to water phase measurements (and viceversa) every two minutes.

The air phase CO_2 concentration (C_a) is given directly by the analyzer in ppm, as air from the flume is pumped out through a ceiling port into the analyzer sample cell at about 60 - 70 ml/min. Standard gas mixtures of $CO_2 = 1000$ ppm, and pure Nitrogen were used for calibration.

For the water phase CO_2 measurements the following sparging method was employed. Water was piped out continuously from the tank, into the top of a 21 glass column 110 cm high, and was pumped out from the bottom of the columm, and returned to the tank, at a rate of 900 ml/min approximately. At the same time nitrogen was bubbled through the column at a rate of 60 ml/min, and the resultant mixture (N_2 and CO_2) went into the analyzer sample cell. The low N_2 flow assures that the gas bubbles were allowed sufficient time to equilibrate with the CO_2 in the water phase, then

$$C_g = H_{CO_2} C_w \tag{1}$$

can be applied in order to calculate the CO_2 concentration in the water phase (C_w) , where H_{CO_2} is the nondimensional Henry's Law constant (Yin and Hassett, 1986) and C_g is the concentration of CO_2 in the $(N_2 \text{ and } CO_2)$ gas mixture.

A high ratio of water flow to nitrogen flow prevents any appreciable depletion of the CO_2 in the sparging tower. This was checked previous to the experiments by allowing the CO_2 to come into equilibrium within the flume after leaving the wind on for several hours. Measurements of the CO_2 concentration in the water were made while varying the purge gas flow rate from 35 to 250 ml/min. For the water circulating flow and column volume used in the experiments, any N_2 purging flow rate of 80 ml/min or lower gave a constant value, while for any higher flow the CO_2 concentration readings decreased.

The CO_2 analog signal was digitized and recorded at 1 Hz. The length of the experiments varied between 1 and 3 hours, depending on wind speed.

Experimental Procedure for H₂O

Unfortunately, the H_2O signal from the NDIR analyzer showed such a slow response that the 2 minute sampling time in each phase was not enough for a stable reading. The slow response was due to condensation/evaporation of water vapor on/from the dust filter installed in the NDIR circuit upstream of the sampling volume. Instead, the rate of change of water vapour concentration in the air phase was measured with a fast response (2 s) relative humidity sensor of the capacitance type (Vaisala 'Humicap'). This sensor was calibrated at the start and end of each run using the NDIR analyzer. Relative humidity and absolute water content signals were digitized and recorded in the computer at 1 Hz.

3

2.4

DATA PROCESSING

An example of the original CO_2 data is shown in figure 7 as a time series of measured concentration from the NDIR analyzer. The intake to the analyzer was switched back and forth between air and water (actually N_2 from the sparging tower) every 2 minutes. The upper envelope (that decreasing with time) corresponds to the CO_2 and N_2 mixture concentration (water phase concentration obtained by sparging with N_2) while the lower portion represents the CO_2 concentration in the air (C_a). The actual CO_2 concentration in water (C_w) is calculated according to (1). Polynomial fitting to both air and water phases time series (see figure 8) allows us to reconstruct the time series, filling the gaps due to the 2 minute intermittency.

From figures 7 and 8 a time delay in the response for the water phase signal is readily apparent – the first two readings in the water phase are constant. While the concentration in the air phase signal is observed to increase as soon as the experimental run begins, the water phase mesurements show a practically constant value for the first 4 to 6 minutes, after which the loss of CO_2 from the water is then continuously detected. Note that the initial 8 minutes are not taken into account for

the polynomial fitting. A shifting procedure was done in order to find the proper time delay between the air and water phase signals, using a mass balance condition as the criterion (see figure 9). (The CO_2 concentration in air was transformed to the equivalent in water by considering the air density and the ratio of air to water volume within the flume). The sum of the CO_2 mass in air and water (a time series), will show a minimum standard deviation about its mean when the proper delay is adopted and the mass balance is satisfied (see figure 10). Typically mass balance better than 1% of total mass was achieved. This delay is introduced in our measurements as a combination of the residence time of the water in the glass column (of the order of $2 \min$), the time required for the CO_2 and N_2 mixture to reach the analyzer and replace the sample cell content (O(1 min)), and the water circulating time through the flume.

The estimation of the mass transfer velocity (K_{CO_2}) follows from

$$L\frac{\partial C_w}{\partial t} = K_{CO_2}(C_w - C_a) \tag{2}$$

where L is a length scale equal to the ratio between the total volume of water (V_w) and the surface area exposed to the air flow (A), and $\partial C_w/\partial t$ represents the time derivative of the CO_2 concentration in water. This term is easily evaluated through the difference between successive values of C_w . The rate of change of concentration of CO_2 and the air-water difference are compared in figure 11a.

Each run produces a time series of $\frac{\partial C_w}{\partial t} (= -\frac{\partial C_a}{\partial t} \times \frac{V_a}{V_w})$ and of $C_w - C_a$, where V_a is the total volume of air in the flume. The 3^{rd} order polynomial fit to these, allows us to calculate K_{CO_2} as a time series. Its constancy over the run (constant slope of figure 11a) provides a rigorous test of the accuracy of the experimental method (figure 11b).

The mass transfer velocity for water is estimated from an expression similar to (2), which is written in the form:

$$L\frac{\partial Q}{\partial t} = K_{H_2O}(Q_{sat} - Q) \tag{3}$$

where the length scale L is now the ratio of total volume of air and the water surface area exposed to the action of the wind. The specific humidity Q (g/Kg) is obtained from the relative humidity signal (H_{rel} in%) according to

$$Q = \frac{622}{p} \frac{H_{rel}}{100} \exp[1.81 + \frac{19.864 \times T_a}{T_a + 273}]$$
(4)

where p is the pressure in mb, and T_a is the air temperature in ^{o}C . The saturated value for specific humidity Q_{sat} is estimated from the average of the last (constant mean) portion of the record from the NDIR analyzer absolute water content signal, once converted from mmol/mol to g/Kg. Note that Q_{sat} is used instead of the surface value Q_{sfc} . The reason is that we do not know the surface temperature (cool skin) T_{sfc} , and we assume that Q_{sat} is its corresponding value. A running average (61 points) is applied to the specific humidity time series Q previous to the estimation of K_{H_2O} which follows from a regression analysis in accordance with (3). In figure 12 typical time series of Q (and its smooth version) and of the water content from the NDIR analyzer are shown. The slow response of the analyzer in comparison to Q is clearly noticeable. The runs lasted long enough however, for the signal to reach an equilibrium level, and that absolute quantity is considered as the saturation value. The relation between the time derivative of the smoothed water content and the difference between the smoothed water content at a specific time and its saturation value is shown in figure 13. The slope of the fitted straight line represents the mass transfer velocity for water K_{H_2O} (divided by the length scale L).

RESULTS

A summary of the experimental runs carried out during two different sessions to measure the CO_2 mass transfer velocity (K_{CO_2}) is given in tables 1 and 2. The average air and water temperature were $24^{\circ}C$ and $21^{\circ}C$ respectively. During runs number 44, 47 and 48, the Pitot tube at station 1 was not functioning. The wind speed dependence of the CO_2 transfer velocity seems to be stronger than linear as can be seen in figure 14, where the wind speed reported is an average over the fetch obtained from the three Pitot tubes at approximately 30 cm above mean water level.

Clearly distinguishable is an increase in the transfer velocity associated with the onset of initial waves, at wind speeds between 2 and 3 m/s. For lower speeds K_{CO_2} is practically constant. However, no abrupt enhancement is observed at the higher wind speeds, that could possibly be related to active breaking of gravity waves although some breaking occurred at wind speeds above 10 m/s at the end of the fetch.

A relation of the form $K_{CO_2} \propto u^N$ has been found, for wind speeds higher than 3 m/s (see figure 15). These data yield N=1.83. It is obvious that in the transfer velocity of gases under water phase control the wind speed dependence is stronger than linear, (compare to N=1.27 for the water transfer experiments below). However, such a power law fit to the mass transfer velocity obscures the detailed behaviour in different ranges of wind speed and wave slope.

In figure 16 the Dalton number for the CO_2 transfer process (D_{CO_2}) is presented as a function of the wind speed. A minimum is observed for wind speeds between 2 and 3 m/s. High Dalton numbers for low wind speed correspond to the flow over a smooth surface. A clear increase in D_{CO_2} with wind speed is observed for higher wind speeds, except between 8 and 11 m/s where a plateau can be distinguished. Such a detailed dependence of D_{CO_2} with wind speed is for the first time revealed from experimental results. The relative minimum is shown to be deeper than that observed for the case of H_2O . In terms of the maximum D_{CO_2} of about 1.2×10^{-5} , the minimum is only as much as 16% of the maximum. That is, a variation of a factor of 6 in the Dalton number for CO_2 has been observed.

The D_{CO_2} values reflect the influence of other factors besides the wind speed, such as the breaking events present at the end of the fetch for wind speeds of about 10 m/s or higher. No sharp transition in D_{CO_2} is seen at the speed where wave breaking begins in the tank. Circular tanks generate a more uniform wave field and so the change in wind speed from non-breaking to breaking waves may be quite abrupt. By contrast, in these measurements an increase in the wind speed simply shortens the fetch where wave breaking starts, so more and more area is subjected to breaking as the speed increases. Consequently, a smooth increase of D_{CO_2} with wind speed is observed.

The results for the mass transfer mesurements of H_2O are summarized in table 3. The wind speed dependence of the water transfer velocity K_{H_2O} is clearly observed in figure 17. K_{H_2O} increases with wind speed for all the range of speeds considered, while for the case of CO_2 , at the lowest wind speed its transfer velocity remains practically constant. A relation of the form $K_{H_2O} \propto u^N$ is also observed for these measurements (see figure 18), where N=1.27 for wind speeds higher than 3 m/s (N=1.05 when the fitting is performed over all wind speeds).

The wind dependence observed in D_{H_2O} shows some interesting similarities and differences with D_{CO_2} . A minimum is also shown at 2-3 m/s wind speed (see figure 19), and D_{H_2O} remains rather constant for wind speeds between 5 and 11 m/s. For higher wind speeds, as the waves develop and breaking begins, the influence of spray and entraining due to wave breaking can be noticed as they enhance the transfer process. However, the relative variation of D_{H_2O} over the wind speed range of these experiments (3.2/1.6=2), is less than that reported for D_{CO_2} (3/0.5=6).

In an attempt to visualize the relative behaviour between CO_2 and H_2O transfer processes, the ratio D_{CO_2} to D_{H_2O} has been estimated by fitting polynomials. This ratio is presented as a function of wind speed in figure 20.

For wind speeds lower than 3 m/s in both cases $(CO_2 \text{ and } H_2O)$ the Dalton number decreases gradually with wind speed. The ratio (D_{CO_2}/D_{H_2O}) decreases with wind speed, reflecting a decrease of D_{CO_2} at a higher rate than D_{H_2O} . This is a relative enhancement in the H_2O transfer process.

From the first appearance of waves, the surface gets rougher as the wind

increases. For these low to moderate wind conditions, when no large wave breaking is yet occurring, the roughness seen from below can be considered equal to that seen from above. However, microbreaking can occur even for these low wind speeds, and the diffusive sublayer in the aqueous phase can be drastically affected (Kahma and Donelan, 1988). Since this effect principally affects the diffusive sublayer below the surface, the transfer of CO_2 is enhanced relatively as the wind speed gradually increases. A steady increase in D_{CO_2}/D_{H_2O} is achieved until the wind reaches about 10 m/s, where breaking starts to occur.

Active breaking also affects the diffusive sublayer above the surface, as splashing can occur and the surface becomes fully rough. Furthermore, the water transfer is also enhanced by air entrainment, allowing some time for the water to diffuse into the air bubbles. Although the transfer of slightly soluble gases is also enhanced by this mechanism, as the diffusive sublayer in the water is disrupted while the occurrence of breaking produces bubbles and splashed droplets, a stronger enhancement can be observed for the moisture transfer process for wind speeds higher than 10 m/s. While both Dalton numbers increase with wind speed, there is a decrease in the rate of change of D_{CO_2}/D_{H_2O} . A fully rough surface can be distinguished by the linear dependence between the roughness scale z_o and the root mean square of the vertical displacement of the water surface η_{rms} . This seems to be the case for wind speeds higher than 7 m/s, where $z_o/\eta_{rms} \sim 1/6$ is readily observed.

5 DISCUSSION

5.1 Comparison with mass transfer velocities from other experiments

There is a considerable number of laboratory measurements of the transfer velocity of gases in general. However, there are not many where specifically CO_2 transfer has been measured (Jähne et al., 1987; Liss, 1983). Transfer velocity estimated with tracers other than CO_2 is usually reported as corrected for a tracer with Schmidt

number equal to 600, the equivalent to that of CO_2 at a temperature of $20^{\circ}C$. Since the Schmidt number dependence of the transfer process is not satisfactorily understood and the effect of the wave field in regulating the transfer process has been proven to be important (Jähne, 1991), we decided to compare our measurements of K_{CO_2} with only those obtained using CO_2 directly in other experiments, and correspondingly those of K_{H_2O} with measurements of the moisture transfer process.

In figure 21, the results of evaporation measurements made from a small tank (4.50m \times 0.30m \times 0.10m) placed in a wind tunnel (Liss, 1973), are compared to the values of K_{H_2O} obtained in our experiments. While it is true that our flume is larger than Liss's tank, and therefore a more developed wave field is simulated, the trend of K_{H_2O} to increase with wind speed is quite similar in both cases. At low and moderate wind speed, when no spray or bubbles are yet present, the transfer process is governed by the behaviour of the physical properties in the gas phase. The differences encountered (Liss's values are about 25% higher than ours) between these two experiments must be largely due to the increased turbulence induced by the small tank installed in the tunnel, enhancing the evaporation process. A smaller part of the difference must be ascribed to the different measuring heights for wind speed.

A comparison of the mass transfer velocity for CO_2 reported from experiments in different small tanks (Kanwisher, 1963; Hoover and Berkshire, 1969; Liss, 1973), with those obtained in the present work is shown in figure 22. It is not surprising that the less developed wind wave field generated in the small tanks affects the measurements resulting in lower values of K_{CO_2} . This is noticeable for wind speed higher that 4 m/s (the exception are the results from Kanwisher). Although microscale breaking may be present even during the early stages of wave development, and may disrupt the diffusive sublayer below the interface, it gets more frequent and important as the waves develop further and as the wind gets stronger. These results stress the importance of the wave field on the transfer process, and the fact that tank dimensions must be properly considered when making intercomparisons among different laboratory experiments.

The results of one set of experiments on CO_2 transfer (Broecker, et al., 1978) carried out in a tank with fetch slightly larger than that of the GTF are presented in figure 23 (the size of their tank was $18 \text{ m} \times 1.0 \text{ m} \times 0.5 \text{ m}$). Although the air and water temperature were about $10^{\circ}C$, for which the Schmidt number of CO_2 is approximately 1000, we include these results since they are the only previous measurements of K_{CO_2} in a large wind and wave tunnel. Therefore, the K_{CO_2} values have been corrected to a Schmidt number of 600. Their results are consistently higher than ours, possibly due to the longer fetch (18 m) for the waves to develop. The values of K_{CO_2} measured in a small circular tank (Jähne et al., 1979) are also shown in this figure. Since circular flumes can presumably be considered to have unconfined fetch, the wave field is more developed an homogeneous, therefore higher transfer velocities are expected. The sharp increase in K_{CO_2} for winds higher than 8 m/s is associated with a sudden transition from a very smooth flow regime, with the presence of a surface film inhibiting the appearance of waves, to a very rough flow regime once the waves are onset and grow continuously to reach steady and homogeneous state (Jähne et al., 1979). Linear flumes inherently simulate a fetch dependent wave field and average properties are responsible for the transfer measured. Therefore, no abrupt enhancing of K_{CO_2} is observed in our experiments and our measured transfer velocities are lower than those reported by Jähne et al (1979) for winds higher than 9 m/s.

5.2

On the Dalton Number Wind Speed Dependence

The wave field influence on the water side controlled gas transfer process has been recognized from previous experiments (Broecker, et al., 1978; Jähne et al., 1979). Furthermore, it is also clear that the mass transfer velocities cannot be parameterized solely by the wind speed. In spite of this, there have been some attempts to estimate CO_2 fluxes over the global oceans from satellite derived wind speed (Etcheto et al., 1991). In this section, besides stressing the importance of properly parameterizing the transfer of gases across the air water interface incorporating as much as possible the physics of the process, we discuss our results in the context of the Dalton number and its possible association with the characteristics of the wind and wave fields present in the experiments. A detailed dependence of the Dalton number (D_{CO_2} and D_{H_2O}) on wind speed is for the first time revealed from experimental work on the transfer of CO_2 and moisture across the air water interface. This must ultimately be related also to the wave field and other associated physical processes which influence the transfer process, like the presence of bubbles and spray droplets. It is therefore clearly shown that the mass transfer velocities K_{CO_2} and K_{H_2O} should not be determined solely from the wind speed.

By inspection of the Dalton number plots as a function of wind speed, it is possible to depict several regimes:

- I Where the Dalton number decreases with wind speed. This regime is associated with very low wind speed, 2.5 m/s or less.
- II Where the minimum in Dalton number is encountered.
- III The Dalton number increases steadily with wind speed. This regime is associated with moderate wind speeds.
- IV A region where the Dalton number remains practically constant.
- V Highest wind speed region where the Dalton number increases again with wind.

The shape of the Dalton number plots as a function of wind speed have some similarities that should be pointed out. A maximum is shown to be present at very low wind speeds, followed by a gradual decrease in both D_{H_2O} and D_{CO_2} (regime I) to reach a minimum at a wind speed between 2 and 3 m/s (regime II). In a smooth flow regime, the viscous sub-layer thins for increasing wind (more precisely, as the roughness Reynolds number increases). On the other extreme (regime V), both Dalton numbers increase steadily from about 12 m/s wind to the highest measured speeds (16 m/s). Right after the minimum is encountered, both D_{CO_2} and D_{H_2O} tend to increase as the wind gets stronger (regime III). This common tendency does not go very far as the H_2O transfer process is not further enhanced for a region in the wind from about 5 to practically 12 m/s. There is however, a somehow narrower wind range where a practically constant D_{CO_2} can also be discerned. This corresponds to regime IV.

The decrease of the Dalton numbers with wind speed at low wind speeds is the characteristic behaviour in smooth flow and is generally predicted by most heat and mass transfer models (Donelan, 1990). The sudden increase associated with the growth of roughness elements (gas phase limited transfer) or with the onset of micro-breaking (liquid phase limited transfer) has been described by previous works, although no general theory exists to describe both processes at play in regime III. Regions IV and V have not been reported before, but may help clarify the mass transfer mechanisms that lead to a region of constant Dalton number for both air and liquid phase limited transfer, even though the obvious forcing parameters -turbulence near the interface and wave breaking- are clearly increasing with wind speed.

Some ideas have been put forward to support the possible variation of D_{H_2O} at high wind speed. Variation of intensity of breaking due to increasing wind may influence the net flux of water by changing both the surface evaporative and the spray droplet components. However, there may be compensating effects as well, as the surface evaporation may be reduced due to sheltering where flow separation occurs while the spray production is increased (Katsaros and DeCosmo, 1988). Through an analysis of the dynamics and thermodynamics of spray formation and evaporation (Bortkovskiĭ, 1987) concludes that spray evaporation is insignificant below 9 m/s, but D_{H_2O} is doubled at 18 m/s, no measurements have so far supported these theoretical

results. Although the measured values of D_{H_2O} in our experiments show an increasing tendency at high wind speeds, the increase is only about 25%. D_{H_2O} varies from a rather constant 2.5×10^{-3} at wind speeds from 6 to 12 m/s, to 3.2×10^{-3} at about 15 m/s-definitively not in a good quantitative agreement with Bortkovskii's theoretical results.

In order to make a proper comparison with some of the results from experiments at sea, the values of D_{H_2O} have been adjusted to a height of 10 m and are presented in figure 24 along with some of the results reported in the literature. The characteristic wind speed dependence of D_{H_2O} from the experiments is still observable from the D_{H_2O} values adjusted to 10 m height, although it is less pronounced. The latest HEXMAX results are believed to support a constant D_{H_2O} , proposed some years ago (Smith, 1989) on the basis of the analysis of the results of a number of laboratory and field experiments. D_{H_2O} estimates from HEXMAX (Smith and Anderson, 1988) for a range of wind speeds between 4 and 18 m/s are shown scattered between approximately 0.8×10^{-3} and 2.0×10^{-3} about an average = 1.2×10^{-3} (see figure 24), even though the drag coefficient was found to increase significantly with the wind speed. The results from other experiments as well as those from HEXMAX are scattered about our D_{H_2O} values. The averages are shown in the figure.

It is apparent that our laboratory measurements are completely consistent with field estimates of D_{H_2O} . However, the scatter in the field measurements is such that the details of the variations of D_{H_2O} are completely obscured. It may well be that field mesurements, such as those reported in figure 24, may be inherently incapable of the precision required to reveal the underlying source of variation of the Dalton numbers. On the other hand, it is often argued that laboratory measurements cannot be scaled up to field conditions.

The task at hand is to understand the processes that lead to the observed dependence of the Dalton numbers of both gas-phase and liquid-phase controlled substances. We have, for historical reasons, been examining the non-dimensional Dalton numbers as a function of wind speed. It is clear that a physically consistent approach would involve the search for suitable non-dimensional ratios of physical variables that describe the mechanisms that operate in regimes I-V. Properly chosen, these nondimensional ratios should describe the Dalton numbers in both laboratory and field. This is a goal that we are currently pursuing. Its realization, however, we must leave for another day.

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CO_2 experiments summary session I										
run	t [sec]	$T_a [^{\circ}C]$	$T_w [^oC]$	$\bar{u_1}$ [m/s]	$\bar{u_2}$ [m/s]	$\bar{u_3}$ [m/s]	u [m/s]	K_{CO_2} [m/s]		
44	3600	23.29	20.44		16.40	15.63	16.02	.202E-03		
47	3600	23.51	21.40	·	16.39	15.64	16.02	.199E-03		
69	3600	23.91	21.70	13.26	14.49	13.75	13.87	.153E-03		
48	3600	24.05	21.54		12.59	11.67	12.13	.122E-03		
51	3600	23.33	21.78	8.95	9.35	8.59	9.15	.761E-04		
53	5400	23.37	21.87	6.09	6.00	5.38	6.05	.293E-04		
66	5400	24.33	21.81	5.0 <u>4</u>	4.59	4.10	4.82	.201E-04		
59	10800	22.77	20.18	3.51	2.45	1.96	2.98	.705E-05		

Table 1: Summary for CO₂ experiments session I, apr 1991

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CO_2 experiments summary session II									
run	t [sec]	$T_a \ [^\circ C]$	$T_w [^{\circ}C]$	$ar{u_1} \mathrm{[m/s]}$	$\bar{u_2} [{ m m/s}]$	$\bar{u_3} [{ m m/s}]$	<i>u</i> [m/s]	K_{CO_2} [m/s]	
151	7200	24.62	20.18	1.03	1.05	.95	1.01	.928E-05	
149	7200	26.38	21.58	1.05	1.09	1.36	1.17	.504E-05	
159	7200	23.58	20.42	1.16	1.26	1.25	1.22	.438E-05	
148	7200	25.74	21.20	1.39	1.69	1.87	1.65	.577E-05	
146	7200	24.48	20.74	1.83	2.05	2.31	2.06	.515E-05	
144	5400	23.60	18.51	2.34	2.52	2.61	2.49	.580E-05	
166	5400	24.62	21.62	7.03	7.56	7.13	7.24	.557E-04	
162	5400	24.08	21.19	7.04	7.61	7.17	7.27	.525E-04	
164	5400	23.02	20.90	7.18	7.73	7.26	7.39	.523E-04	
160	5400	23.58	20.76	7.20	7.72	7.28	7.40	.571E-04	
165	3600	23.63	21.20	10.10	10.97	10.40	10.49	.829E-04	

Table 2: Summary for CO₂ experiments session II, may-jun 1991

H_2O experiments summary										
run	t [sec]	p[kPa]	$T_a [°C]$	$T_w \ [^oC]$	$\bar{u_1} [{ m m/s}]$	$ar{u_2} \mathrm{[m/s]}$	<i>ū</i> ₃ [m/s]	<i>u</i> [m/s]	K_{H_2O} [m/s]	
84	10800	100.97	23.86	20.71	.92	1.18	.05	.72	.0020	
85	7200	100.95	23.93	20.76	1.15	1.71	1.00	1.28	.0032	
92	7200	100.99	23.94	20.01	1.28	1.43	1.60	1.44	.0035	
88	5200	100.14	23.36	18.68	1.40	1.56	1.77	1.57	.0037	
86	7200	100.40	20.91	18.15	1.93	2.26	1.75	1.98	.0040	
87	7200	100.25	22.13	18.39	2.43	2.66	2.17	2.42	.0045	
81	7200	100.18	23.72	21.12	2.53	2.96	2.44	2.64	.0049	
80	7200	99.46	24.63	21.37	3.44	3.71	3.42	3.52	.0070	
79	7200	99.82	23.85	21.11	4.11	4.59	4.01	4.24	.0084	
83	3600	100.20	24.13	21.41	4.93	5.41	4.93	5.09	.0115	
78	3600	99.89	23.49	20.09	5.98	6.51	5.95	6.15	.0146	
77	1800	99.89	23.31	19.82	7.77	8.45	7.79	8.01	.0199	
76	3600	99.96	22.80	19.55	9.57	10.54	9.73	9.95	.0239	
75	1800	100.04	21.93	18.50	11.30	12.53	11.72	11.85	.0300	
74	1800	100.04	21.53	18.19	12.93	14.40	13.66-	13.66	.0379	
73	1200	100.04	21.55	17.79	14.47	16.15	15.52	15.38	.0499	

Table 3: Summary for H_2O experiments, apr-may 1991

GAS TRANSFER FLUME



Fig 1



Figure 2





Figure 3



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Figure 5





Figure 6







Fig 9

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Fig 2



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Mean reference wind speed [m/s]

Fig 14





Fig15





Fig 16



Fig 17





Fig 18



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Fig 22

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Wind speed [m/s]

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Fig 24



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