

**Air–sea gas exchange at hurricane wind speeds**

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# First air–sea gas exchange laboratory study at hurricane wind speeds

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## Abstract

In a pilot study conducted in October and November 2011, air–sea gas transfer velocities of the two sparingly soluble trace gases hexafluorobenzene and 1,4-difluorobenzene were measured in the unique High-Speed Wind-Wave Tank at Kyoto University, Japan. This air–sea interaction facility is capable of producing hurricane strength wind speeds of up to  $u_{10} = 67 \text{ m s}^{-1}$ . This constitutes the first lab study of gas transfer at such high wind speeds. The measured transfer velocities  $k_{600}$  spanned two orders of magnitude, lying between  $11 \text{ cm h}^{-1}$  and  $1180 \text{ cm h}^{-1}$  with the latter being the highest ever measured wind induced gas transfer velocity. The measured gas transfer velocities are in agreement with the only available dataset at hurricane wind speeds (McNeil and D’Asaro, 2007). The disproportionately large increase of the transfer velocities found at highest wind speeds indicates a new regime of air–sea gas transfer, which is characterized by strong wave breaking, enhanced turbulence and bubble cloud entrainment. It was found that tracers spanning a wide range of solubilities and diffusivities are needed to separate the effects of enhanced surface area and turbulence due to breaking waves from the effects of bubble and spray mediated gas transfer.

## 1 Introduction

Ocean regions, where strong winds usually occur, play an important role in global  $\text{CO}_2$  budgets, see Bates et al. (1998). Therefore, a better understanding of gas transfer at high wind speed conditions is essential. Field measurements of air–sea gas exchange velocities under hurricane wind speed conditions are sparse due to difficulties of sampling under extreme wind conditions. During hurricane Frances in 2004, McNeil and D’Asaro (2007) measured three transfer velocities of  $\text{O}_2$  using unmanned floats at wind speeds larger than  $25 \text{ m s}^{-1}$ , with the highest wind speed being  $50.4 \text{ m s}^{-1}$ .

High wind speeds are associated with the presence of breaking waves. These enhance turbulence near the water surface and generate spray and bubble plumes, which

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increases gas fluxes, see for instance Monahan and Spillane (1984) and Farmer et al. (1993). Breaking waves enhance gas transfer by several mechanisms. The water surface, across which gas is transferred, is enlarged by waves, and by breaking, waves enhance near surface turbulence. Bubbles and spray provide a limited, mostly short-lived volume of air or water associated with an additional surface area, over which gas transfer can occur (Memery and Merlivat, 1985). And by floating through air and water and bursting through the water surface, bubbles and spray enhance turbulent mixing near the water surface.

Wind-wave tanks provide an alternative to measurements in the field. All the inconveniences and dangers associated with measurements in the field during hurricane wind speed conditions are virtually non-existent in a lab setup.

## 2 Air–sea gas transfer

The gas transfer velocity  $k$ , along with the net gas flux  $j$ , is commonly used to describe the gas transfer process across the air–sea boundary

$$j = k\Delta c = k(c_w - \alpha c_a) \quad (1)$$

with the tracer's air and water side concentrations,  $c_a$  and  $c_w$ , respectively, and the tracer's dimensionless solubility  $\alpha$ .

For a sparingly soluble tracer, a dependency of the transfer velocity  $k$  on the water sided friction velocity  $u_*$ , a measure for momentum input into the water, is commonly assumed in the form

$$k \propto u_* Sc^{-n} \quad (2)$$

with the tracer's dimensionless Schmidt number  $Sc = \nu/D$ , the ratio between the kinematic viscosity of water  $\nu$  and the tracer's diffusivity in water  $D$ . The Schmidt number exponent  $n$  is  $2/3$  in the case of a smooth water surface and  $1/2$  for a rough and wavy





is the rate of water inflow to replace water lost from the flume due to spray being blown out of the tank. The Kyoto High-Speed Wind-Wave Tank is an open facility, meaning fresh ambient air is blown over the water surface once and then removed from the system. Choosing a tracer that is not present in ambient air, the condition of a negligible air side concentration  $\alpha c_a \approx 0$ , can be met.

Equation (5) can be easily solved,

$$c_w(t) = c_w(0) \cdot \exp\left(-\left(k \cdot \frac{A}{V_w} + \frac{\dot{V}_w}{V_w}\right) \cdot t\right), \quad (6)$$

with  $c_w(0)$  being the water side concentration at time  $t = 0$ .

The time constant  $\tau$  of this equation is defined as

$$\frac{1}{\tau} := k \cdot \frac{A}{V_w} + \frac{\dot{V}_w}{V_w}. \quad (7)$$

This time constant  $\tau$  is acquired from an exponential fit of Eq. (6) to the time series of measured concentrations. The water volume  $V_w$ , the water surface area  $A$ , as well as the leak rate  $\lambda = \dot{V}_w/V_w$  are known or measured during an experiment. The transfer velocity can then be calculated as

$$k = \left(\frac{1}{\tau} - \lambda\right) \cdot \frac{V_w}{A}. \quad (8)$$

## 4 Experiments

### 4.1 Tracers

The tracers were chosen such that their diffusivity in water, and thus their Schmidt numbers, were similar, while their solubility differed. Because UV absorption spectroscopy

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was used to measure tracer concentrations, tracers which exhibit a high extinction coefficient in the UV range as well as distinctly different spectra were chosen. To keep the mass balance described in Sect. 3 simple, the tracers were required to not be present in ambient air, as well as not present in the tap water. The tracers chosen by these criteria were hexafluorobenzene (HFB) and 1,4-difluorobenzene (DFB). Table 1 lists properties of the tracers as well as carbon dioxide as a reference.

## 4.2 Experimental setup

### 4.2.1 The Kyoto High-Speed Wind-Wave Tank

The Kyoto High-Speed Wind-Wave Tank has a linear flume shape, see Fig. 2. The water flume is 80 cm wide, has a total length of 15.7 m with 12.9 m being exposed to the wind. The total height is 1.6 m, with up to 0.8 m being filled with tap water. The wind is generated by a radial fan. The maximum wind speed that can be reached is  $u_{10} = 67.1 \text{ m s}^{-1}$ . Before the wind enters the air side of the tank, it is passing through a honeycomb structure to minimize large eddies. The air is taken from the room surrounding the wind-wave tank and guided out of the building after it was blown over the water.

There is an external water tank available that holds up to  $7 \text{ m}^3$  of water, which is connected to the wind-wave flume by two pipes. A pump draws the water out at the downwind end of the flume and into the water tank, and another pump draws the water out of the tank and into the upwind end of the wind-wave flume. For all lower wind speed settings, the amount of water coming out of the lab's water supply lines is sufficient to replace the water lost due to spray. At the highest wind speed setting, the external tank was used as a buffer to keep the water level constant inside the wind-wave tank. Trace gases can be mixed into the water by operating both pumps and thus cycling the water between the external tank and the wind-wave flume.

## 4.2.2 Concentration measurement

Tracer concentrations in the water were monitored using UV absorption spectroscopy. Water was sampled at a fetch of about 6.5 m at a water height of approximately 35 cm with a rate of 7 to 10 L min<sup>-1</sup>. The approximate sampling location is marked in Fig. 2.

Because air bubbles, generated by breaking waves would have scattered the light out of the UV spectroscopic measuring cell, it was chosen to not spectroscopically analyze the water directly, but to equilibrate the water with a small parcel of air first, and analyze this air. The water extraction and equilibration setup is shown in Fig. 3. A membrane equilibrator called an oxygenator (*Jostra Quadrox* manufactured by *Maquet*, Hirrlingen, Germany) was used to equilibrate the water with the air. In Krall (2013), the performance and applicability of the oxygenator setup is validated from geometrical consideration as well as test experiments. Air is cycled around the closed air loop at a rate of about 150 mL min<sup>-1</sup>. During the measurements, the valves were set such, that no outside air could enter or leave the air loop. During preparation of the experiment, the valves allowed sampling of ambient air to estimate the background. In addition, the water temperature was monitored.

The gas sampling cell is made of a 1 m long quartz glass tube with an inner diameter of 3 mm. Light produced by a deuterium lamp enters the tube through a quartz glass lens with focal length of 5 cm and a quartz glass window. It leaves the measuring cell through another quartz glass window and lens to be focused on a glass fiber. This glass fiber is connected to a UV spectrometer (*Maya2000 Pro* by *Ocean Optics*, Dunedin, USA). This spectrometer can resolve wavelengths from 190.5 nm to 294.1 nm with a resolution of approximately 0.05 nm. About one spectrum was acquired per second.

During data evaluation, one absorbance value per tracer is calculated from each spectrum in a process described in detail in Krall (2013). Beer's law states that the absorbance  $A$  of a tracer is directly proportional to the concentration in the measured air parcel,  $c_a$ . According to Henry's law, the air side concentration is proportional to the

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water side concentration  $c_w$ , thus  $A \propto c_a \propto c_w$ . Because only the change in concentration over time is relevant to measure the gas transfer velocity, see Eqs. (6) and (8), no absolute calibration that converts absorbance into the water side concentrations is needed. Equation (6) can then be converted into the form

$$\frac{c_w(t)}{c_w(0)} = \frac{A(t)}{A(0)} = \exp\left(-\frac{t}{\tau}\right), \quad (9)$$

with the time constant  $\tau$  that is needed to calculate the gas transfer velocities, see Eq. (8).

### 4.3 Experimental conditions

A total of 21 experiments at 9 different fixed wind speeds were performed. The wind generator's rotational frequency  $f_{\text{fan}}$  was set and kept constant for each condition. The free stream wind speed  $u_{\text{inf}}$ , the air sided friction velocity  $u_*$  as well as the wind speed at a height of 10 m  $u_{10}$  that is commonly used as a reference were not measured during the presented campaign, but taken from a table kindly provided by the Japanese colleagues. Water height  $h_w$  was measured at the wind inlet before and after each experiment with no wind and no waves. Typically, both water height values differed by no more than 1%. This ensured that the rate of inflowing water  $\dot{V}_w$  was equal to the amount of water lost due to spray as required by the method, see Sect. 3. The conditions used are listed in Table 2. Transfer velocities of both tracers were measured in parallel in each of the experiments, with the exception of one experiment at  $f_{\text{fan}}=600$ , where only the absorbance time series of DFB could be evaluated.

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## 5 Results

### 5.1 Comparison with the gas exchange model and previous measurements

A total of 41 transfer velocities were measured, 21 of which for DFB and 20 for HFB. Figure 4 shows the measured transfer velocities for both tracers vs. the transfer velocities predicted by Eq. (2). The momentum transfer resistance parameter  $\beta$  was assumed to be 6.7, see Krall (2013). The Schmidt number exponent was chosen to be  $n = 0.5$  at medium to high wind speeds. However, for the two lowest wind speeds of  $7 \text{ ms}^{-1}$  and  $12.1 \text{ ms}^{-1}$  the exponent was set to a value of 0.55, to compensate for the smooth water surface visually observed at low fetches during this wind speed conditions. For the low friction velocities of up to  $u_* < 6 \text{ cm s}^{-1}$ , which corresponds to a wind speed of  $u_{10} < 35 \text{ ms}^{-1}$  and transfer velocities around  $80 \text{ cm s}^{-1}$ , the measured transfer velocities agree well with the transfer model's prediction. At higher wind speeds, the measured transfer velocities exceed the ones expected from Eq. (2) by up to around 340 % (HFB) and 220 % (DFB).

Figure 5 shows the transfer velocities, scaled to a Schmidt number of 600 using Schmidt number scaling (Eq. 3) in comparison with the data by McNeil and D'Asaro (2007) acquired on the open ocean, including their proposed parameterization. Within the margin of errors, both data sets agree surprisingly well.

### 5.2 Enhancement at highest wind speeds

Vlahos et al. (2011) present measured transfer velocities of dimethylsulfide (DMS), which show a decrease in the gas transfer velocity when bubble clouds are present at high wind speeds. For both tracers used in this study, this decrease is not observed. Up to a wind speed of  $35 \text{ ms}^{-1}$ , the gas transfer velocity is roughly proportional to  $u_{10}^{1.1}$ . For higher wind speeds, the proportionality changes to  $k \propto u_{10}^3$  for DFB and  $k \propto u_{10}^{3.6}$  for HFB, see Fig. 6. This clearly indicates the start of a new regime of air sea gas exchange starting at around  $35 \text{ ms}^{-1}$ .

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At highest wind speeds, the transfer velocity of HFB increases stronger than the one of DFB, as indicated by the different slopes in Fig. 6. To quantify this, an enhancement factor  $E_f$  can be defined by

$$E_f := \frac{k_{600,\text{HFB}} - k_{600,\text{DFB}}}{k_{600,\text{DFB}}} \times 100\%. \quad (10)$$

5 Figure 7 shows the enhancement factor  $E_f$ , averaged on a condition basis. Up to a wind speed of around  $40 \text{ m s}^{-1}$ , no enhancement is observed. Above  $40 \text{ m s}^{-1}$ , however, the transfer velocity of HFB is up to 40 % larger than that of DFB with a clear wind speed dependence. This enhancement is expected from bubble models, see Sect. 2, with the less soluble tracer HFB ( $\alpha = 1.0$  at  $20^\circ\text{C}$ ) having larger transfer velocities than the slightly higher soluble tracer DFB ( $\alpha = 3.08$  at  $20^\circ\text{C}$ ).

## 6 Conclusions

The transfer velocities at hurricane strength wind speeds were found to be extremely high. The measured transfer velocities were found to be in agreement with the only other data set of gas transfer at extreme wind speeds (McNeil and D'Asaro, 2007). In wind speeds higher than around  $35\text{--}40 \text{ m s}^{-1}$ , where frequent large scale wave breaking with bubble entrainment and spray generation occurs, the correlation between gas transfer velocities and wind speed was found to become steeper, indicating a new regime of air–sea gas exchange. The steepness of the relationship between the gas transfer velocity and the wind speed could be linked to the solubility of the tracer. The lower the solubility, the higher the transfer velocities measured.

The tracers used in this pilot study spanned only a small fraction of the Schmidt number – solubility parameter space, see Fig. 8. Because the covered parameter space is so small, general statements, applicable to all of the solubility and Schmidt number range would be highly speculative and is therefore omitted in this paper.

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The results of this pilot study confirm, that it is possible, to measure realistic air–sea gas exchange velocities in a wind-wave tank at hurricane wind speeds with the method described in this paper. However, due to the mentioned limitations in solubility and Schmidt number, a physical interpretation as well as physics based modeling will have to be suspended until further measurements with more tracers and detailed measurements of bubble and spray densities and of turbulence have been conducted. Choosing tracers covering the whole range of possible solubilities and Schmidt numbers is expected to expand the understanding of the processes involved in air–sea gas exchange at highest wind speeds. Partitioning the transfer velocity into a part due to enhanced turbulence at the water surface, and spray and bubble mediated gas transfer seems feasible in future studies at the Kyoto High-Speed Wind-Wave Tank.

## Appendix A

### Measured transfer velocities

Table A1 shows all measured transfer velocities, as well as mean water temperatures and Schmidt numbers of both tracers.

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**Table 1.** Molar mass, solubility, diffusivity in water and Schmidt numbers of the tracers hexafluorobenzene (HFB) and 1,4-difluorobenzene (DFB) for a temperature of 20 °C. Also shown is CO<sub>2</sub> for comparison. [1]: calculated from mole fraction solubility from Freire et al. (2005) and vapor pressure from Ambrose et al. (1990). [2]: Yaws and Yang (1992). [3]: Young (1981). [4]: Yaws (1995) [5]: calculated from diffusion coefficients taken from Yaws (1995) and water viscosity taken from Kestin et al. (1987). [6]: Jähne et al. (1987).

Name	$M$ g mol <sup>-1</sup>	$\alpha$ at 20 °C	$D$ 10 <sup>-5</sup> cm <sup>2</sup> s <sup>-1</sup>	Sc
HFB	186.1	1.0 <sup>[1]</sup>	0.736 <sup>[4]</sup>	1360 <sup>[5]</sup>
DFB	114.1	3.08 <sup>[2]</sup>	0.815 <sup>[4]</sup>	1225 <sup>[5]</sup>
CO <sub>2</sub>	44.01	0.83 <sup>[3]</sup>	1.68 <sup>[6]</sup>	601 <sup>[6]</sup>

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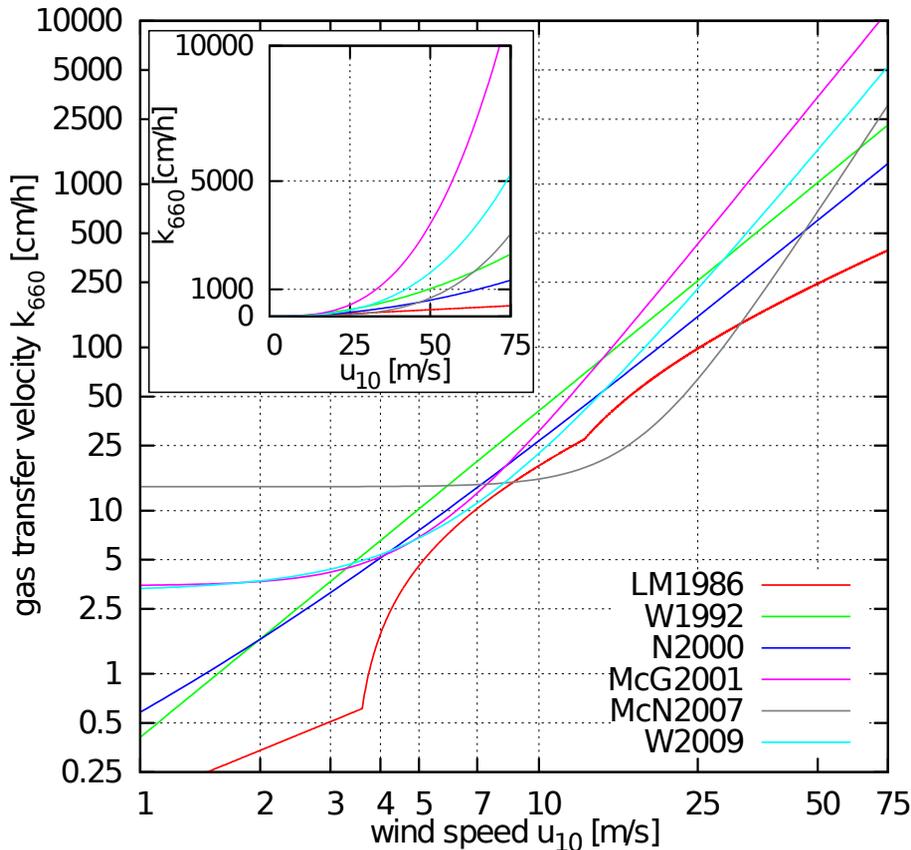
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**Table 2.** Experimental conditions used at the Kyoto High-Speed Wind-Wave Tank.  $f_{\text{fan}}$  is the frequency of the wind generating fan,  $u_*$  is the friction velocity,  $u_{\text{inf}}$  denotes the free stream velocity, and  $u_{10}$  is the wind speed at 10 m height.  $\dot{V}_w$  is the leak rate. The number of repetitions of each of the conditions is labeled with  $N$ . One free stream velocity  $u_{\text{inf}}$  was not measured (n.m.).

$f_{\text{fan}}$ rpm	$u_*$ $\text{cm s}^{-1}$	$u_{\text{inf}}$ $\text{m s}^{-1}$	$u_{10}$ $\text{m s}^{-1}$	$\dot{V}_w$ $\text{L min}^{-1}$	$N$	notes
100	0.836	4.72	7.0	0	3	
150	1.50	10.36	12.1	0	1	
200	2.34	10.29	16.7	0	2	
250	3.10	n.m.	23.75	0	1	
300	5.19	16.26	29.8	0	3	
400	7.24	22.17	40.7	0	3	
500	8.23	28.47	48.0	3.5	2	
600	9.37	34.75	56.4	14.5	4	only 3 experiments evaluable for HFB
800	11.5	43.29	67.1	192	2	$V_w$ decreased, external tank used

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**Fig. 1.** Some commonly used gas transfer – wind speed parameterizations in a double logarithmic plot. The insert shows the same, but with linear axes. LM1986: Liss and Merlivat (1986), W1992: Wanninkhof (1992), N2000: Nightingale et al. (2000), McG2001: McGillis et al. (2001) and W2009: Wanninkhof et al. (2009). The parameterization by McNeil and D’Asaro (2007) (McN2007) is the only one developed for hurricane wind speeds.

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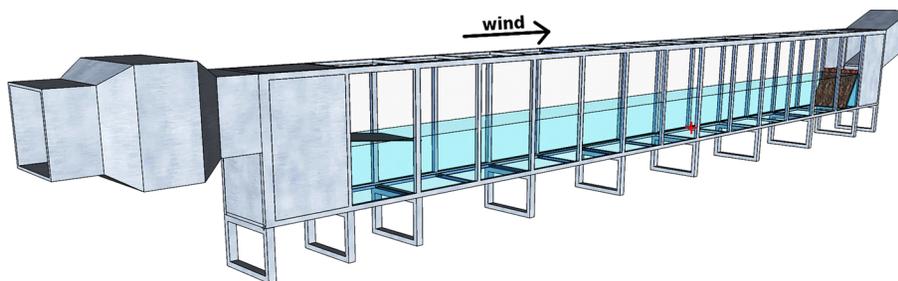
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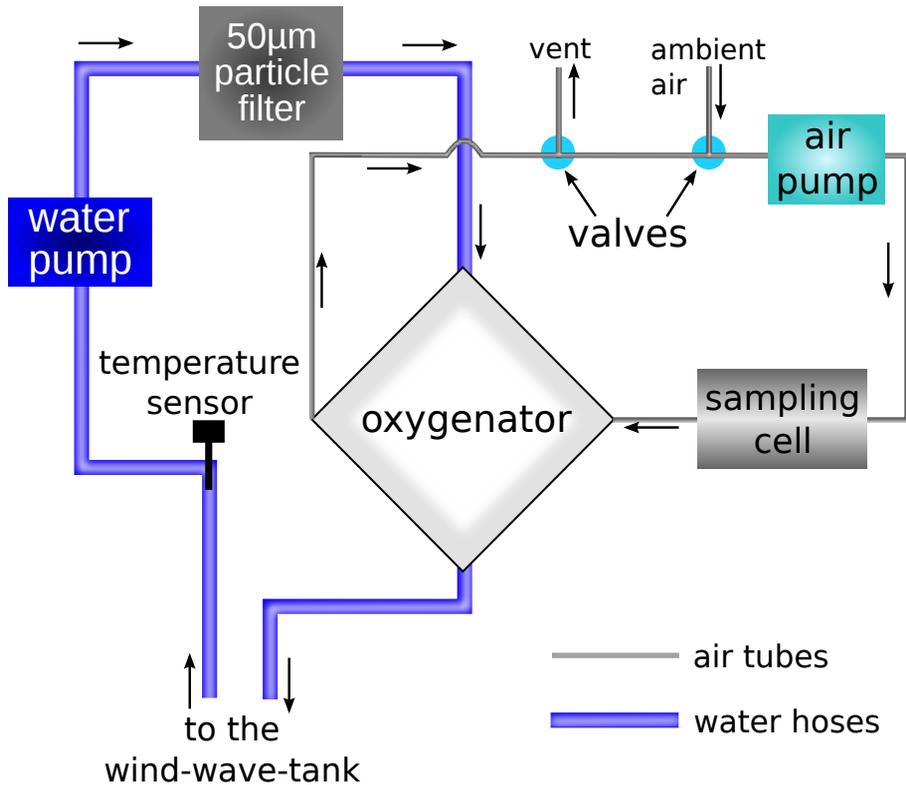
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**Fig. 2.** Schematic view of the flume section of the Kyoto High-Speed Wind-Wave Tank. Not shown is the radial fan producing the wind (left side). The red cross marks the approximate sampling position.

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**Fig. 3.** Gas extraction setup. Water pumped from the wind-wave tank is equilibrated with air using an oxygenator. The air is continuously cycled between the oxygenator and the UV-spectroscopic measuring cell. The valves allow background sampling and are closed during measurements.

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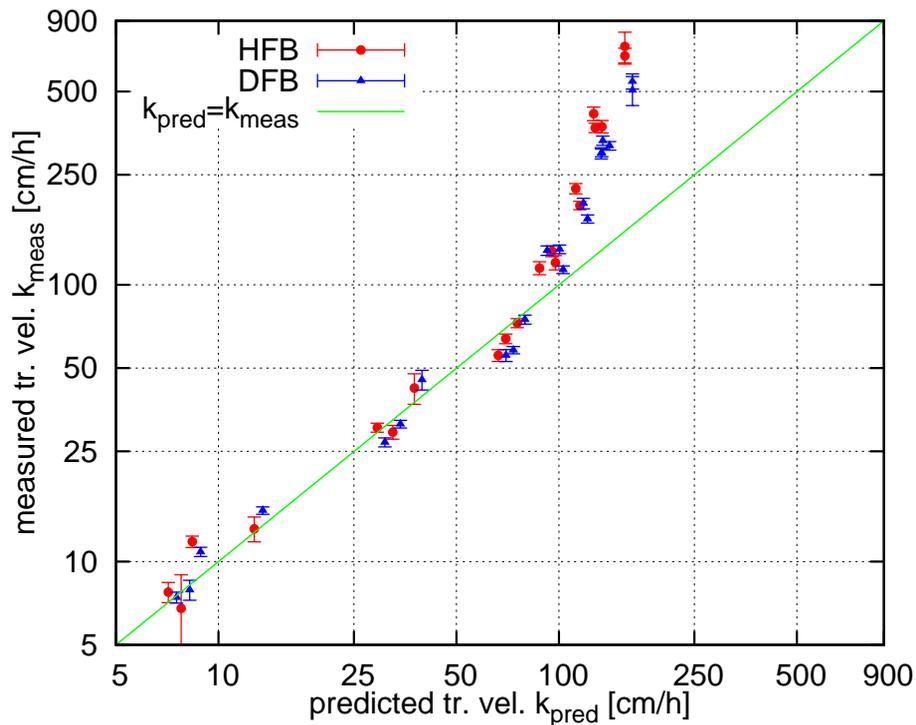
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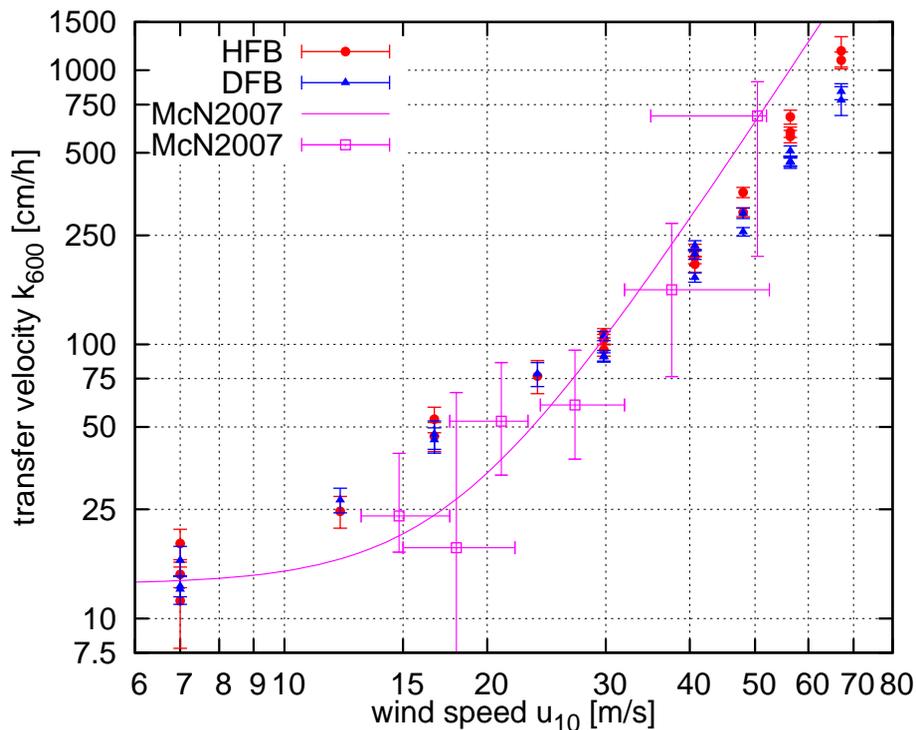


**Fig. 4.** Measured transfer velocities  $k_{\text{meas}}$  for HFB and DFB as well as the transfer velocities  $k_{\text{pred}}$  predicted by Eq. (2) in a double logarithmic plot.

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**Fig. 5.** Measured transfer velocities for HFB and DFB, compared to the data and parameterization by McNeil and D’Asaro (2007), all scaled to a Schmidt number of 600, in a double logarithmic plot.

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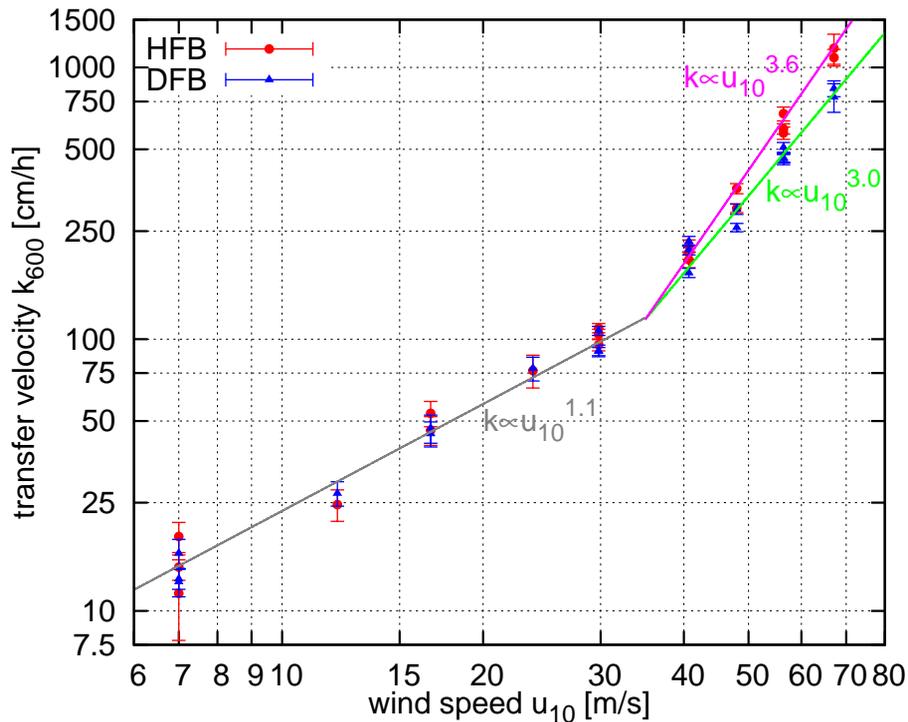
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**Fig. 6.** Measured transfer velocities for HFB and DFB scaled to a Schmidt number of 600. Also shown are lines indicating proportionalities of the transfer velocities  $k_{600}$  to  $u_{10}^x$  with  $x$  depending on the wind speed range and the tracer.

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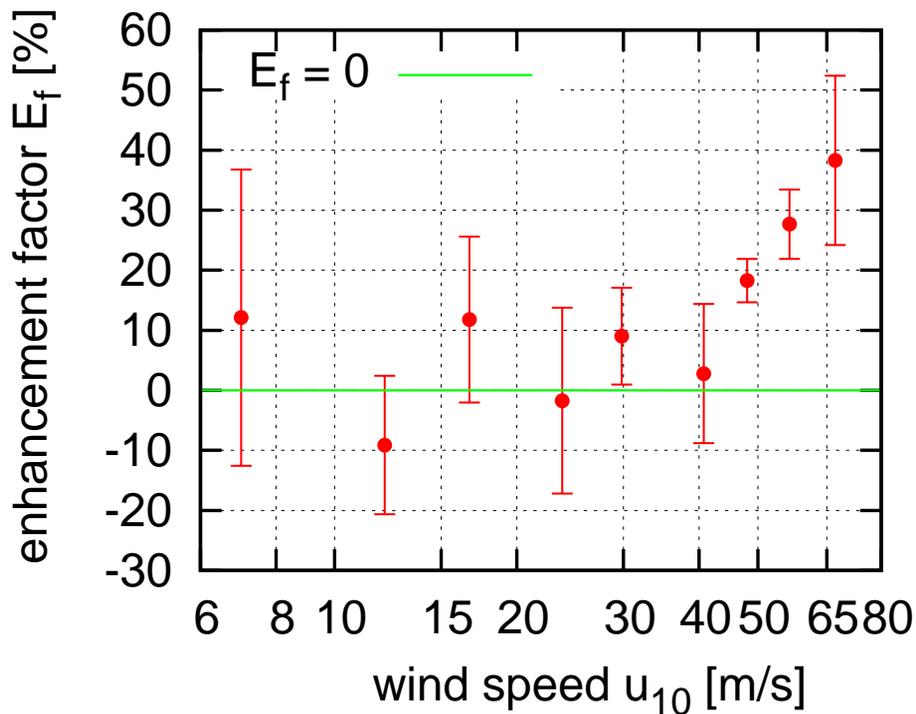
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**Fig. 7.** Mean enhancement of the transfer velocity of HFB over that of DFB, both scaled to a Schmidt number of 600. An  $E_f$  of 0 means no enhancement.

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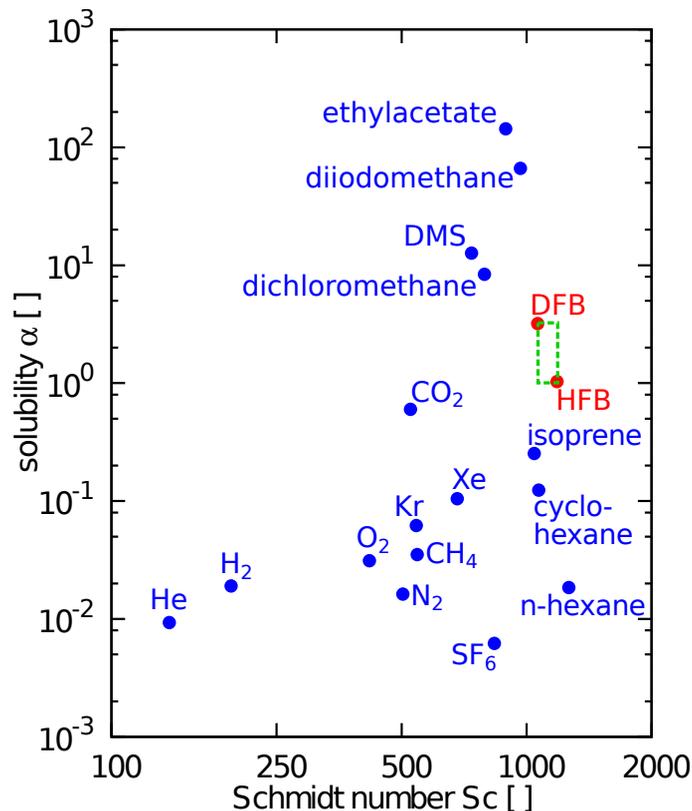
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**Fig. 8.** Double logarithmic Schmidt number – solubility diagram of some environmentally important tracers for a temperature of 25°C. The tracers used in this study, hexafluorobenzene (HFB) and 1,4-difluorobenzene (DFB), cover a very limited parameter range, marked by the dashed green rectangle.

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