

Workshop

# **Cross-linking lab and field measurements and numerical modeling to identify and quantify the mechanisms of air-sea gas transfer**

**5 – 6 September 2022, Heidelberg, Germany**

**Organizers:**

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**Location: Mathematikon, Im Neuenheimer Feld 205, 69120 Heidelberg  
Conference Room 5.104 and Common Room 5.303, 5th floor**

# Program

## Monday 5 September

**12:00 Welcome lunch in Common Room 5.104**

**12:45 – 14:30 First Session in Conference Room 5.303, Status of Field Measurements I**

**12:45** Bernd Jähne, IUP & IWR, Heidelberg University, Germany: *Welcome and introduction to the workshop*

**13:00** Rik Wanninkhof, NOAA AOML, Miami FL, USA: *Knowledge gaps in gas transfer and impact on estimates of global and regional CO<sub>2</sub> fluxes (p. 4)*

**13:30** Yuanxu Dong et al., Centre for Ocean and Atmospheric Sciences, School of Environmental Sciences, University of East Anglia, UK: *Random uncertainties in eddy covariance air-sea CO<sub>2</sub> flux measurements (p. 5)*

**14:00** Minxi Yang et al., Plymouth Marine Laboratory, Plymouth, UK: *How to avoid pitfalls in eddy covariance air-sea gas flux measurements (p. 6)*

**14:30 – 14:45 Coffee break in Common Room 5.104**

**14:45 – 16:15 Second Session in Conference Room 5.303, Status of Field Measurements II**

**14:45** David T. Ho, University of Hawai'i at Mānoa, USA: *<sup>3</sup>He/SF<sub>6</sub> tracer release experiments (p. 7)*

**15:15** David Bastviken, Linköping University, Sweden: *Capabilities and limits of floating chambers to measure the gas transfer velocity (p. 8)*

**15:45** Bernd Jähne et al., IUP & IWR, Heidelberg University: *Thermographic Techniques to Measure the Air-Sea Gas Transfer Velocity and to Explore the Mechanisms: Previous Deficits and New Perspectives (p. 9)*

**16:15 – 16:30 Coffee break in Common Room 5.104**

**16:30 – 18:00 Third Session in Conference Room 5.303 Ongoing Advanced Field Measurements and Status of Laboratory Studies I**

**16:30** Iwona Wrobel-Niedzwiecka et al., Institute of Oceanology, Polish Academy of Sciences, Sopot, Poland: *Effect of organic matter surface layer enrichment on air-sea gas transfer velocity (SURETY) (p. 11)*

**16:50** Christina Braybrook, University of Calgary, Canada: *Eddy covariance measurements in remote sea ice covered marine environments (p. 12)*

**17:10** Zoé Le Bras, Institute of Biogeochemistry and Pollutant Dynamics, ETH Zürich, Switzerland: *Estimation of marine selenium emissions in Baltic Sea using field measurements (p. 13)*

**17:30** Kerstin Krall, IUP, Heidelberg University, Germany: *Strengths and weaknesses of annular wind-wave tanks by the example of the Aeolotron (p. 14)*

**18:00 – 19:00 Visit of Heidelberg Aeolotron including a Poster Session (p. 27-29)**

**19:30 Joint dinner at "Bräustadel" (<https://www.braeustadel.de/>, Berliner Str. 41, Mathematikon B)**

## Tuesday, 6 September

### 9:00 – 10:30 Fourth Session in Conference Room 5.303, Ongoing Advanced Laboratory Studies

**9:00** Pim Bullee et al., Norwegian University of Science and Technology (NTNU), Trondheim, Norway: *The influence of water-side turbulence on the gas transfer rate across an air-water interface (p. 15)*

**9:20** Katherine E. Adler, Cornell University of Civil and Environmental Engineering, Cornell, USA: *Laboratory investigation of significant gas transfer enhancement via capillary-gravity bow waves (p. 17)*

**9:40** Daniel Ruth et al., Institute for Fluid Dynamics, ETH Zürich, Switzerland: *Measurement of entrained air bubble dynamics in a laboratory wind-wave facility (p. 19)*

**10:00** Alexander V. Babanin et al., The University of Melbourne, Australia: *Wave-Coupled Effects in the CO<sub>2</sub> Exchange and Spray Production near the Ocean Interface (p. 20)*

### 10:20 – 10:35 Coffee break in Common Room 5.104

### 10:35 – 12:20 Fifth Session in Conference Room 5.303, Status of Laboratory Studies II and Status Modeling

**10:35** Denis Bourras et al., Institut Pythéas, Aix Marseille Université, France: *The Large Air-Sea Interaction Facility (LASIF) of Luminy-Marseille: overview of past and ongoing activities (p. 21)*

**11:05** Yuliya Troitskaya et al., Institute of Applied Physics, Nizhny Novgorod, Russia: *The fetch dependent sea spray generation function at high winds: theoretical background and laboratory verification (p. 22)*

**11:30** Breanna Vanderplow et al., Nova Southeastern University Oceanographic Center, Dania Beach, FL, USA: *Sea Spray Generation Under Tropical Cyclone Conditions in the Presence of Surfactants and Implications for Air-Sea Gas Exchange (p. 23)*

**11:55** Alexander V. Soloviev et al., Nova Southeastern University Oceanographic Center, Dania Beach, FL, USA: *Air-Sea Gas Transfer in Tropical Cyclones: Multiphase Modeling and Comparison with Laboratory and Field Experiments (p. 25)*

### 12:20 – 13:00 Lunch break in Common Room 5.104

### 13:00 – 14:15 Sixth Session: Split up in working groups on best possible linked research strategies from different perspectives

Working group I Field experiments

Working group II Lab experiments

Working group III Modelling

### 14:15 – 14:30 Coffee break in Common Room 5.104

### 14:30 – 15:00 Wrap up with short reports from the working groups in Conference Room 5.303

## Knowledge gaps in gas transfer and impact on estimates of global and regional CO<sub>2</sub> fluxes

*Rik Wanninkhof<sup>1</sup>*

<sup>1</sup>NOAA AOML, Miami FL

Significant advances have been made over the past decades to constrain regional and global air-sea carbon dioxide (CO<sub>2</sub>) flux estimates. Monthly data-based products at 1-degree resolution are routinely produced, often utilizing machine learning approaches to fill in the sparse surface water CO<sub>2</sub> data. The fluxes are determined by a bulk flux formulation where the flux density is the product of a gas transfer velocity ( $k$ ) and the air-water CO<sub>2</sub> concentration difference ( $\Delta C$ ). A simple quadratic parameterization of gas exchange and windspeed is commonly used. This parameterization shows broad agreement with experimentally determined  $k$ 's in several field studies. The resulting global air-sea CO<sub>2</sub> fluxes agree with global ocean inventory estimates and models. These global constraints are powerful but do not necessarily properly represent local and regional CO<sub>2</sub> fluxes which are gaining increasing societal interest with respect to carbon mitigation efforts and "stocktakes". Moreover, such a simple representation is at odds with our knowledge of processes controlling the exchange. To reconcile this conundrum systematic approaches need to be developed spanning micro to regional scales with robust cross-checks and modeling. In specific, the conventional way of estimating regional and global fluxes with a bulk formulation must be rigorously compared with direct flux estimates and other approaches. Upscaling from experimental facilities to the field must be done with full appreciation of scales of phenomena. All estimates should include a full sensitivity, error, and uncertainty analyses. Forcing of gas transfer needs to be parameterized in a physical framework with remotely sensed information for gap filling of  $k$  values that remains a key uncertainty in flux.

## Random uncertainties in eddy covariance air-sea CO<sub>2</sub> flux measurements

*Yuanxu Dong<sup>1,2</sup>, Mingxi Yang<sup>2</sup>, Thomas G. Bell<sup>2</sup>*

<sup>1</sup>Centre for Ocean and Atmospheric Sciences, School of Environmental Sciences, University of East Anglia, Norwich, UK

<sup>2</sup>Plymouth Marine Laboratory, Prospect Place, Plymouth, UK

Eddy covariance (EC) is the most direct method to measure the air-sea carbon dioxide (CO<sub>2</sub>) flux and offers a powerful way to study relatively small-scale processes that drive gas transfer. Since the late 2000s, the quality of the ship-based EC air-sea CO<sub>2</sub> flux observations has been significantly improved because of instrumentation and data processing advancements. However, inherent random uncertainties in EC CO<sub>2</sub> flux observations have not been well-quantified. Random uncertainty can be reduced by averaging repeated measurements. However, averaging the EC flux for too long will obscure the natural flux variability. So, how large is the random flux uncertainty under different conditions, and what is the optimal flux averaging timescale?

In this talk, I will present:

- The contribution of the instrumental noise and the insufficient sampling to the total random uncertainty.
- Why insufficient sampling is an inherent issue and dominates the random uncertainty in EC flux measurements.
- A practical way to find the optimal averaging timescale (1-3 hours for CO<sub>2</sub>) for analysis of EC flux observations.
- The implications of this study on the EC-derived gas transfer velocity analysis.

# How to avoid pitfalls in eddy covariance air-sea gas flux measurements

*Mingxi Yang<sup>1</sup>, Yuanxu Dong<sup>1,2</sup>, Tom Bell<sup>1</sup>*

<sup>1</sup>Plymouth Marine Laboratory, Prospect Place, Plymouth, UK;

<sup>2</sup>Centre for Ocean and Atmospheric Sciences, School of Environmental Sciences, University of East Anglia, Norwich, UK

Eddy covariance (EC) is the most direct approach to measure gas fluxes over the ocean at time scales of ca. 1 hr. However, due to complexities and nuances in the applications of EC for measuring different gases and on different platforms, it is quite easy to bias the measurement. In this talk, I will touch upon the following considerations in the setup and analysis of EC flux data over the ocean.

- Flow distortion: where should the EC system be set up?
- Gas of interest (CO<sub>2</sub>, DMS, VOCs, and others): open path vs. close path? dryer vs. no dryer?
- When things move on ships: motion correction of winds and motion sensitivity in gas instrument
- Quality control filtering of data: measures of stationarity and homogeneity

Finally, I will talk about how to assess the accuracy of EC flux data.

## **$^3\text{He}/\text{SF}_6$ tracer release experiments**

*David T. Ho<sup>1</sup>*

<sup>1</sup>University of Hawai'i at Mānoa

As requested, I will discuss the capabilities, accuracy and limits of  $^3\text{He}/\text{SF}_6$  dual tracer measurements to determine gas transfer velocities in the coastal and open oceans.

# Capabilities and limits of floating chambers to measure the gas transfer velocity

*David Bastviken*<sup>1</sup>

<sup>1</sup>Department of Thematic Studies – Environmental Change, Linköping University

The gas transfer velocity ( $k$ ) represents a key factor shaping water-air gas exchange rates. Current uncertainties in  $k$  across various aquatic systems translate to large uncertainties in e.g. aquatic greenhouse gas emissions. Making accurate in situ assessments of  $k$  remains challenging. Estimating  $k$  from measurements of concentration gradients across the water-air interface and of gas fluxes using floating chambers are considered one option that is conceptually straightforward, widely transparent and available in terms of practical work and calculations, and possible to perform with simple, inexpensive and mobile field equipment. Such measurements have been discussed and challenged in terms of system disturbance and chamber design. Comparisons with other non-invasive approaches have provided guidance and allowed progress. Increased use of the floating chamber approach has generated additional questions, triggered by e.g. differences in Schmidt number normalized  $k$  among gases. A fundamental question is how empirical assessments of apparent  $k$  from floating chamber measurements, integrating many complex processes and being influenced by various practical constraints, are linked to ideal  $k$  values and general theories and models regarding gas exchange. This presentation summarize two decades of attempts to estimate  $k$  using floating chambers, along with associated findings on pros, cons, and potential improvements.



# Thermographic Techniques to Measure the Air-Sea Gas Transfer Velocity and to Explore the Mechanisms: Previous Deficits and New Perspectives

*Bernd Jähne<sup>1,2</sup>, Lucas Warmuth<sup>1</sup> and Kerstin E. Krall<sup>1</sup>*

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For almost 40 years thermographic techniques have been used to explore air-sea gas transfer (Jähne et al., 1989). Compared to geochemical tracer and eddy covariance techniques, thermography comes with distinct advantages: a) a shorter measuring time, b) a small measuring area at the water surface in the order of just one square meter, c) direct insight into the mechanisms by analyzing the spatio-temporal temperature distribution at the water surface and d) suitability for both laboratory and field experiments.

The application of thermographic techniques in the past decades also revealed their deficits. The three main issues are:

1. The Prandtl number is about 100 times smaller than the Schmidt number in water. This raises the principal question of whether heat transfer across the water-sided boundary layer is governed by the same mechanisms as mass transfer (Asher et al., 2004).
2. Until now, thermographic techniques have been modeled as a simple one-dimensional system with vertical transport only. There is, however, also horizontal transport. Once a parcel at the water surface enters a heated patch, it takes some time for it to come into equilibrium with the applied heat flux.
3. Thermographic techniques cannot be used to quantify gas transfer induced by submerged bubbles. This can be seen as an opportunity to solely estimate the water surface-related gas exchange.

A careful study in the Heidelberg Aeolotron (Nagel et al., 2014) has shown that heat transfer velocities can correctly be scaled to gas transfer velocities, if the Schmidt number exponent is known with sufficient accuracy. But a direct proof that both transfer processes are governed by the same mechanisms can only be given when the gas concentration fields in the mass boundary layer are measured in conjunction with thermographic measurements.

In this contribution we will discuss how the spatio-temporal analysis of the temperature patterns at the water surface in a patch heated by a 10.6  $\mu\text{m}$  carbon dioxide laser could open up new perspectives:

1. Even faster measurements of the transfer velocity with the time constant natural to the transport process across the aqueous heat boundary layer (seconds instead of minutes). The faster measurements are essential for the study of the transport processes under unsteady conditions.
2. The direct identification of the transport mechanisms and the determination of the Schmidt number exponent.

## References

Jähne, B., Libner, P., Fischer, R., Billen, T., and Plate, E. J. (1989), Investigating the transfer process across the free aqueous boundary layer by the controlled flux method, *Tellus B*, 41B, 177–195, doi:10.3402/tellusb.v41i2.15068

Asher, W. E., Jessup, A. T. and Atmane, M. A. (2004), Oceanic application of the active controlled flux technique for measuring air-sea transfer velocities of heat and gases, *J. Geophys. Res.*, 109, C08S12, doi:10.1029/2003JC001862

Nagel, L., Krall, K. E. and Jähne, B. (2014), Comparative heat and gas exchange measurements in the Heidelberg Aeolotron, a large annular wind-wave tank, *Ocean Sci.*, 11, 111–120, doi:10.5194/os-11-111-2015

## **Effect of organic matter surface layer enrichment on air-sea gas transfer velocity (SURETY)**

*Wrobel-Niedzwiecka I.<sup>1</sup>, Drozdowska V.<sup>1</sup>, Kulinski K.<sup>1</sup>, Makuch P.<sup>1</sup>, Piskozub J.<sup>1</sup>*

<sup>1</sup>Institute of Oceanology of the Polish Academy of Sciences, Sopot, Poland

In 2022 at our Institute, we started a new project: Effect of organic matter surface layer enrichment on air-sea gas transfer velocity (SURETY). The aim of this project is to study the effect of surfactants and surface-active organic matter on the CO<sub>2</sub> air-sea gas exchange. For this purpose, measurements are made from r/v Oceania (the ship of IOPAS, Sopot) including: in-water measurements of CO<sub>2</sub> partial pressure using an underwater system equipped with a shower-type equilibrator and a Cavity Ring Down Spectrometer (Picarro G-2101i) and, for cross-checking, also AT, CT, pH; the atmospheric CO<sub>2</sub> concentrations and fluxes using a direct Eddy Covariance method, where we use two sets of equipment: Li-COR 7200 with GiLL WindMaster Pro sonic anemometer and 7500 with GiLL WindMaster sonic anemometer. To obtain the most accurate estimation results, air-se CO<sub>2</sub> flux systems are located at 10 m above sea level (7 m above ship deck) and approximately 3-5 m overboard the ship; fluorescence properties of surfactants are measured using a fluorescence spectrometer that produces excitation-emission matrices in a wide spectral range with the hanging mercury drop method. Our measurements are from the Baltic Sea and North Atlantic Ocean/Arctic Ocean during different sea conditions.

In our project, I am responsible for simultaneous EC measurements, which we want to use to propose a new CO<sub>2</sub> flux parameterization using a fluorescence-based index. During my Ph.D. I studied the influence of different gas transfer parameters on air-sea CO<sub>2</sub> fluxes through the Arctic Ocean surface, effects of different drag coefficient formulas depending on wind stress climatology, and after my Ph.D. I studied the distribution of pCO<sub>2</sub> in the water and air-sea CO<sub>2</sub> fluxes using the feedforward neural network method.

## **Eddy covariance measurements in remote sea ice covered marine environments**

*Christina Braybrook*<sup>1</sup>

<sup>1</sup>University of Calgary, Canada

The global ocean is currently a net sink for atmospheric carbon dioxide (CO<sub>2</sub>), a potent greenhouse gas. Boarding three oceans, Canada's ongoing federal policy regarding the reduction of CO<sub>2</sub> emissions has significant global influence, which ultimately relies on understanding how atmospheric CO<sub>2</sub> is being exchanged at the ocean surface. This has been difficult, particularly in the Arctic Ocean, where there is high spatiotemporal variability and remote, harsh conditions. Specifically, the unreconciled influence of sea ice on CO<sub>2</sub> exchange at the surface contributes to the gap in knowledge of the Arctic marine carbon sink. Broadly, the goals of this research are twofold: (1) design and implement innovative systems and techniques to observe CO<sub>2</sub> gas exchange in remote sea ice covered marine environments, and (2) use observations to contribute to global models that currently do not account for gas exchange from polar oceans during sea ice cover. To achieve this, we have established an eddy covariance weather tower near Cambridge Bay, Nunavut in Dease Strait, where CO<sub>2</sub> exchange in and out of the surface ocean is being measured. Previous research has shown that operating these types of systems in cold, marine regions have been challenging due to power limitations and instrument bias due to water vapour. However, we have developed a remote power grid and an instrument drying system to combat this, and this tower has resulted in the only successful collection of seasonally resolved CO<sub>2</sub> exchange measurements from a sea ice environment. Not only does this shed insight into the role of sea ice on gas exchange in the Arctic Ocean, but this provides a template for how to effectively monitor CO<sub>2</sub> exchange using micrometeorological systems in remote regions that are currently underrepresented in larger scale climate models.

## Estimation of marine selenium emissions in Baltic Sea using field measurements

Zoé Le Bras<sup>1</sup>

<sup>1</sup>Institute of Biogeochemistry and Pollutant Dynamics, ETH Zürich, Switzerland

Selenium (Se) is an essential trace element for humans and animals. The atmosphere represents an important reservoir of Se, which supplies Se to the Earth's surface and thus presents a source of Se to ecosystems and food chains. The main natural atmospheric Se source is derived from marine biogenic emissions with dimethyl selenide (DMSe) the major species followed by dimethyl diselenide (DMDS<sub>e</sub>) and dimethyl selenyl sulphide (DMSeS) present in trace concentrations (fM-pM) in seawater and in the atmosphere. As Se plays an important role for human health, a global atmospheric Se model (Feinberg et al., 2020) has been developed based on Se emissions and Se chemistry. For that, the biogenic Se emissions have been estimated using sea surface DMSe concentration scaled on the DMS concentration (Lana et al., 2011) and wind-driven parametrization (Nightingale et al., 2000). The understanding of the sea-air exchange of DMSe is not only hindered by the scarce amount of data available in marine systems but also by the lack of dedicated laboratory studies focused on the determination of empirical values.

Nevertheless the concentration of these volatile organic Se species are poorly constrained in marine environments due to a lack of adequate analytical methods and large uncertainties remains in the sea-air flux estimations. To improve our understanding of DMSe in marine systems, we recently developed a highly sensitive and selective offline analytical method using a thermal desorption coupled with gas chromatography inductively coupled plasma mass spectrometry (TD-GC-ICP-MS) to quantify volatile Se as well as volatile organic sulfur species in seawater and in the atmosphere. Field campaigns in early and late summer were conducted in the Baltic Sea by simultaneous sampling in the air and in the seawater to further obtain insights into DMSe sea-air exchange.

### References

Feinberg, A.; Stenke, A.; Peter, T.; Winkel, L. H. E. Constraining Atmospheric Selenium Emissions Using Observations, Global Modeling, and Bayesian Inference. *Environ. Sci. Technol.* 2020, 54 (12), 7146–7155. <https://doi.org/10.1021/acs.est.0c01408>.

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Lana, A.; Bell, T. G.; Simó, R.; Vallina, S. M.; Ballabrera-Poy, J.; Kettle, A. J.; Dachs, J.; Bopp, L.; Saltzman, E. S.; Stefels, J.; Johnson, J. E.; Liss, P. S. An Updated Climatology of Surface Dimethylsulfide Concentrations and Emission Fluxes in the Global Ocean. *Glob. Biogeochem. Cycles* 2011, 25 (1). <https://doi.org/10.1029/2010GB003850>.

## Strengths and weaknesses of annular wind-wave tanks by the example of the Aeolotron

*Kerstin E. Krall<sup>1</sup>, Bernd Jähne<sup>1,2</sup>*

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Wind-wave tanks are essential tools to study the small-scale mechanisms governing the exchange of mass, heat and momentum across the air-water interface. In wind-wave tanks, environmental factors such as wind speed, air and water temperatures and surfactant cover of the water surface can be precisely controlled. Two very different shapes of wind-wave tanks are commonly used, the more common linear type and the annular type. The Heidelberg Aeolotron is the world's largest operational annular wind-wave tank. The annular shape of the Aeolotron has some major advantages over the more conventional linear wind-wave tanks:

- homogeneous conditions across the whole water surface allow for comparison between measuring techniques integrating over the whole water surface as well as local techniques
- the virtually infinite interaction time between wind and waves allows waves to come into equilibrium with the wind, wave ages achievable are much higher than in linear tanks
- the whole fetch range can be studied by replacing fetch distance with time

However, also some disadvantages have to be considered:

- centrifugal forces prevent the formation of a logarithmic wind profile and give rise to secondary currents
- the maximum wave age attainable is still limited by the water depth

In this talk, the strengths and weaknesses of the Aeolotron will be discussed and contrasted with linear tanks and open ocean conditions.

# The influence of water-side turbulence on the gas transfer rate across an air-water interface

*Pim Bullee<sup>1</sup>, Astri Nore<sup>1</sup>, R. Jason Hearst<sup>1</sup>*

<sup>1</sup>Department of Energy & Process Engineering Norwegian University of Science & Technology (NTNU)

Understanding and predicting the gas transfer rate across the air-sea interface is a multi-faceted and complex problem. In situ, there are waves at the interface and turbulence in both phases (amongst other factors). In this study, we perform the first steps in decomposing these effects, herein focusing on turbulence in the liquid-phase. We introduce a methodology whereby we can manipulate the intensity and length scales of the turbulence in the water while holding the bulk Reynolds number constant. This allows us to disassociate turbulence effects from bulk Reynolds number effects. Compared to stationary box experiments and DNS, we allow the water to flow, increasing our bulk and turbulence Reynolds numbers by nearly an order of magnitude ( $Re = UH/\nu = 65000$ ,  $Re_T = u'2L/\nu = 7400$ ,  $Re_\lambda = u'\lambda/\nu = 335$ ), and we allow the turbulence to deform the interface.

An active grid is placed at the inlet of a 11.2 m long recirculating water facility. This device is composed of a series of vertical and horizontal bars with paddles attached to them. These bars are actuated in different patterns to manipulate the turbulence properties in the flowing water. A surface plate is used immediately downstream of the grid to establish a consistent initial surface condition, so that the downstream surface topology is a function of the turbulence in the flow, rather than the grid itself. Four test cases were studied, resulting in turbulence intensities ranging from 2% to 7%. All measurements were conducted far downstream of the active grid so that the flow can be considered fully developed. The total volume of water in the facility is roughly 20,000 litres. At the start of each experiment the water is depleted of dissolved oxygen using sodium sulphite, and the re-oxygenation of the water is measured with fine optical probes. Re-oxygenation can take up to 24 hours for full recovery to the oxygen saturation concentration, and this time decreases with increasing turbulence intensity. The flow conditions were quantified using particle image velocimetry and laser-Doppler velocimetry, from which the relevant length scales were derived. The free surface topology was captured with synthetic schlieren and resistive wave gauges.

The oxygen concentration probe was traversed vertically to resolve the concentration boundary layer and its instantaneous position relative to the interface was assessed via simultaneous wave gauge measurements. Both the concentration boundary layer thickness and the gas transfer rate were found to increase with the measured turbulence intensity. Both monotonically increased within the measurement range investigated here, i.e., an asymptote was not achieved within the measurement range, indicating that mapping the effects of turbulence is crucial for the understanding of the mechanisms involved in gas transfer. Moreover, the mean interface area changed only marginally between test cases, suggesting turbulent transport was the primary mechanism for the changes rather than a change to the surface area.

This is a first step in exploring the relationship between turbulence and gas-transfer in flowing system. The facility is equipped with a wavemaker and is presently having airflow installed with adjustable turbulence properties, which will enable the addition of increasing complexity in future investigations.



## Laboratory investigation of significant gas transfer enhancement via capillary-gravity bow waves

*Katherine E. Adler<sup>1</sup>, Edwin A. Cowen<sup>1</sup>*

<sup>1</sup>Cornell University School of Civil and Environmental Engineering

Gas transfer increases more rapidly with wind speed after waves begin to form on the water surface (e.g., Kanwisher, 1963; Broecker et al., 1980). When waves are present, the gas transfer velocity scales with the mean square slope of the waves (e.g., Jähne et al., 1987; Saylor and Handler, 1997). Capillary-gravity waves (~1cm in length) are of particular interest because they are more common and steeper than gravity waves. Saylor and Handler (1997) isolated non-breaking capillary waves and achieved almost two orders-of-magnitude enhancement in gas transfer velocity,  $k$ , compared to the static case, over a 0.016-m<sup>2</sup>-area interface. However, analytical models have yet to explain such a dramatic increase, predicting up to 3- or 3.5-fold enhancement due to the steepest capillary waves which are likely unstable over a large area (Szeri, 1997; MacIntyre, 1971; Hasse and Liss, 1980).

To further investigate the interfacial scalar flux enhancement due to capillary-gravity waves isolated from wind over a large (5.95 m<sup>2</sup> total) area, we conducted several reaeration experiments with bow waves in a straight, open, recirculating flume. Bow waves form upstream of objects disturbing the water surface at sufficient relative flow speed (at least 23 cm/s at standard air-water interface). Based on analytical models relating capillary-gravity bow wave amplitude to peak external pressure at the object and velocity (assuming the object imposes a pressure field in the form of a Dirac delta function or imposes a fixed surface draw-down depth [Raphaël & de Gennes, 1996; Chevy & Raphaël, 2001]), and estimating a peak pressure that scales with the stagnation pressure ( $1/2\rho v^2$ ), it is hypothesized that the mean squared slope ( $(ak)^2$ ) of these waves and the resistance force on the object due to the waves scale approximately with velocity to the fourth power or greater. Such a dependence would suggest that gas transfer rate in the presence of these waves has a similarly strong dependence on flow velocity.

Capillary-gravity bow waves were generated over a 2-m<sup>2</sup> (34% of total area) area using an array of vertical, 3.2-mm-diameter cylinders suspended above the interface to penetrate the water surface by about 1 cm. In some cases, dowels were suspended from a conveyor belt apparatus to isolate the influence of relative dowel speed from that of flow speed, which contributes to other sources of mixing.

Cases with no dowels, stationary dowels, and moving dowels were compared at several relative velocities between 5 and 60 cm/s. The presence of the capillary-gravity bow waves increased gas transfer velocity by at least 20-68% compared to cases with similar background flow speed and no dowels. When the base flow was reduced to about 5 cm/s and the dowels were conveyed against the flow at 60 cm/s, gas transfer was enhanced

607% over the 5 cm/s control case without dowels. It is clear that the enhancement in gas transfer due to these waves increases with velocity but it is not yet clear if that dependence is greater than quadratic as predicted by the analytical slope models mentioned previously

## Measurement of entrained air bubble dynamics in a laboratory wind-wave facility

*Daniel Ruth<sup>1,2</sup>, Baptiste Néel<sup>1</sup>, Martin Erinin<sup>1</sup>, Megan Mazzatenta<sup>1</sup>, Robert Jaquette<sup>3</sup>, Fabrice Veron<sup>3</sup>, and Luc Deike<sup>1,4</sup>*

<sup>1</sup>Department of Mechanical and Aerospace Engineering, Princeton University

<sup>2</sup>Institute for Fluid Dynamics, ETH Zurich

<sup>3</sup>School of Marine Science and Policy, University of Delaware

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Air bubbles that are created at the ocean surface when waves break significantly enhance the transfer of gases between the atmosphere and the ocean, accounting for roughly 40% of the oceanic uptake of anthropogenic carbon dioxide emissions. We use high-resolution photography and high-speed stereo imaging in a laboratory wind-wave channel to measure the statistics of their sizes, depths under the water surface, and dynamics while moving through the water. Along with enabling highly-resolved measurements, the laboratory setting offers the ability to control the characteristics of the breaking wave field and to relate an observation of an entrained bubble to the properties of the wave which broke to entrain it. The wave fields we generate are characterized by  $U_\infty/c_p$  (the ratio between the free-stream wind velocity and the phase speed) from 5.9 to 11.1 and characteristic wave slopes  $a_0k_p$  from 0.15 to 0.36.

Our measurements allow for the characterization of three quantities which are important to air-sea gas exchange. First, for each of the wave fields we generate, we obtain transient and time-averaged bubble depth distributions, which will impact the time available for the gases in the bubbles to diffuse to the water as the bubbles rise back towards the surface. Below the depths of the deepest wave troughs, the concentration of the bubbles decays exponentially, with a length scale set by the characteristic height of the wave field. Second, we find a common shape of the distribution of sizes of entrained between conditions, and we relate the total quantity of air under the surface to characteristics such as the breaking frequency and wave height. Finally, our dynamical measurements, which are comprised of three-dimensional trajectories of entrained bubbles and the local surface elevation, suggest that the bubbles are advected in the direction of the wave faster than the Stokes drift associated with the wave height and phase speed. This is due to the additional Lagrangian transport caused by wave breaking, which has been previously shown to enhance the advection speed of fluid tracers and solid particles.

# Wave-Coupled Effects in the CO<sub>2</sub> Exchange and Spray Production near the Ocean Interface

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Air-sea interaction processes are often facilitated, controlled and therefore essentially coupled with the surface waves, and those include exchange of momentum, energy, heat, moisture, aerosols, gasses. We particularly note the role of wave breaking which locally enhances the intensity of such exchanges by orders of magnitude.

In the presentation, we will look at the role of waves and wave breaking in CO<sub>2</sub> exchange and spray production, across the full range of Metocean conditions from light to strong winds. Proposed parameterisations are based on both laboratory experiments and field measurements, and accommodate dependences on the surface waves.

For CO<sub>2</sub> flux estimates, the gas transfer rate was scaled with wave orbital velocity and laboratory experiments were conducted. Non-dimensional gas transfer velocity was further related with wind-sea Reynolds numbers (composed of wave parameters), wave breaking probability and an enhancement factor due to the wind speed. To stress the role of the waves, we conducted laboratory experiments with waves produced by wavemaker with and without wind in the flume, i.e. including cases when the gas transfer cannot be described by traditional wind-based formulae in principle. The laboratory-developed gas transfer parameterisation was further tested with historic field data. We explicitly parameterised the bubble injection rate through Reynolds number and breaking probability instead of wind speed and the final expression performed well across the data sets both in laboratory and the field.

Sea spray, also, while largely produced by wave breaking (or by interactions between wind and waves), is traditionally parameterized in terms of winds alone. We present in-situ observations of sea spray volume fluxes derived from laser altimeter readings. The measurements cover a broad range of wind and wave properties and are used to develop a novel wind-wave-dependent spray-volume-flux model. While our observations and parameterisation are on average consistent with the classic sea spray volume flux models in magnitude and respective trends, locally it can be distinctly different depending explicitly on the properties of the wave field.

## **The Large Air-Sea Interaction Facility (LASIF) of Luminy-Marseille: overview of past and ongoing activities**

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The LASIF is a 40 m long wind-wave tank built in the early 70's. It is equipped with a programmable wave generation paddle and a controlled fan that blows quasi-laminar air at speeds up to 15 m/s. A wide range of fetches is reachable, from some meters to 30 m. A unique property of the facility is its well-shaped and fully developed turbulent atmospheric boundary layer from the lightest winds (1 m/s). A survey of the available instruments for studying air, wave, and sea properties will be presented, including current sensors, wave gauges, microwave sensors, and PIV and CTA probes. Next, a summary of recent science results obtained at the facility and ongoing activities will be described, they regard both engineering inclined subjects and fundamental wind turbulence-wave studies. The LASIF is fully and easily open to new collaborations, with a specific interest to questions around the reconciliation of open sea data, turbulence theories, and wind-wave tank measurements that account for wind, wave, currents, as well as thermal stratification.

# The fetch dependent sea spray generation function at high winds: theoretical background and laboratory verification

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The "bag breakup" fragmentation is the dominant mechanism for generating spray in hurricane winds, which parameters substantially affect the exchange processes between the ocean and the atmosphere including the dynamics of the development of sea storms and gas exchange. This fast process can only be studied in lab using sophisticated experimental techniques based on high-speed video filming. In such circumstances, the transfer of laboratory data to field conditions requires a kind of theoretical model that describes how the initiation of disturbances occurs, which then lead to fragmentation events, what is the threshold for fragmentation, what is the volume of liquid, which determines the size of spray droplets, that undergoes fragmentation, and how it depends on wind parameters, etc. The conclusions of the model can be first verified in the laboratory experiment and then applied to field conditions.

In the present work, such a model is proposed. First of all, a linear theory of small-scale disturbances on the water surface under the action of a strong wind has been built, which makes it possible to describe their structure, dispersion properties and determine the threshold value of the dynamic air flow velocity at which such disturbances become growing. These disturbances comprise small-scale ripples concentrated within the thin surface layer and growing fast due to shear instability of the wind drift flow in the water. The peculiarity of the structure of these disturbances enables one to consider the nonlinear stage of their evolution within the Riemann simple wave equation modified to describe the increasing disturbances. The analytical solution of the obtained equation suggests the scaling of the volume of liquid undergoing the "bag-breakup" fragmentation, to estimate the scale of the formed droplets and the speed of their injection into the atmosphere. The scaling correctly describes the dependencies of these quantities on the wind friction velocity obtained in laboratory experiments performed at two completely different facilities, Large thermostated tank with high speed wind-wave channel at the Institute of Applied Physics and at the annular wind-wave flume Aeolotron at Heidelberg University.

The obtained results are applied for the construction of the fetch-dependent spray generation function, which is applicable in the field. Within the Lagrangian stochastic model for the inertial droplets in the marine boundary layer, the momentum, heat, moisture and enthalpy exchange coefficients are calculated. One should notice substantial feedback effect on the atmosphere caused by the presence of spray in hurricane conditions.

# Sea Spray Generation Under Tropical Cyclone Conditions in the Presence of Surfactants and Implications for Air-Sea Gas Exchange

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Gas transfer velocity, which is key to gas exchange, depends on a variety of factors including near-surface turbulence, wind speed, wave breaking, the presence of surfactants, bubbles, and sea spray generation. While air-sea gas exchange has been studied for many years, gas exchange under tropical cyclone conditions has been only briefly studied due to obvious difficulties in data collection. Gas transfer velocities increase as wind speeds do, specifically above 33 to 35 m s<sup>-1</sup>. Krall et al. (2019) studied air-sea gas exchange under high winds using wind-wave tanks at Kyoto University and University of Miami and found that above 33 m s<sup>-1</sup>, gas transfer increases dramatically across the air-sea interface, and that bubbles have a low contribution to gas exchange. Their experiments revealed that the gas transfer velocity dependence on solubility is reduced significantly for sea spray under high winds. Spray and spume generation are considerably increased under increasing wind speeds, meaning that the gas flux may also increase substantially. Surfactants, which are produced by marine microorganisms or applied as dispersants, further complicate the understanding of this phenomenon. Surfactants alter surface tension, and thus under tropical cyclone conditions, impact sea spray and spume generation by changing sea spray size distribution (Vanderplow et al., 2020). Surfactants also may suppress gas exchange through their suppression of turbulence and formation of films on the sea surface. Therefore, surfactants can potentially modify air-sea gas exchange.

To better understand the relationship between surfactants, spray generation, and gas exchange, we simulated the spray generation under Category 1, 3, and 5 tropical cyclone conditions using a computational fluid dynamics model. The key feature of our model is ANSYS Fluent's Volume of Fluid to Discrete Phase Model, which converts water parcels to Lagrangian particles, representing sea spray. Utilizing our 528-core supercomputer and Fluent's dynamic mesh adaptation that refines areas of high curvature, we can resolve spray droplets with radii starting from 100- $\mu$ m. Our numerical results were partially validated with experimental data from the University of Miami SUSTAIN facility under Category 1 tropical cyclone conditions including cases with surfactants (Vanderplow et al., 2020). The Category 1 model revealed an approximately three-fold increase in the sea spray generation function due to surfactants. In all three tropical cyclone categories, surfactants caused an increase of spray concentration (from 20-34%) within the spray radii range from 100 to 500  $\mu$ m. Based on these results, we anticipate that surfactants alter air-sea gas exchange through modified size distribution and increased concentration of spray. An increase in sea spray generation should lead to an increase of air-sea gas exchange. At

the same time, surfactant films at the free wavy air-water surface suppress gas exchange (Jähne, 2019). For sea spray droplets, the presence of surfactants can affect fluid motion within spray droplets and the ratio of the spray time scales to the residence time, impeding gas exchange between spray droplets and the air (Andreas et al., 2017). Future computational studies verified with laboratory data are expected to better understand the effect of surfactants on the sea spray generation function.



# Air-Sea Gas Transfer in Tropical Cyclones: Multiphase Modeling and Comparison with Laboratory and Field Experiments

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The influence of tropical cyclones on the air sea gas exchange is characterized by the presence of a two-phase environment comprising air bubbles and sea spray. Under low wind speed conditions, the interfacial component of the air-sea gas exchange dominates while different gases tend to converge when normalized by the Schmidt number. With increasing wind, the bubble-mediated component becomes more important and depends on the gas solubility. Under tropical cyclone conditions, only about 4% of the sea surface is covered by whitecaps (Holthuijsen et al. 2012). The rest of the sea surface is covered by so-called whiteout. Soloviev et al. (2017) identified the main component of the whiteout as the spume generated by the local air-sea interface instability by different mechanisms including Kelvin-Helmholtz instability. Tennekes and Lumley (1972) and Andreas et al. (2017) found that fluid motion within spray droplets significantly increases the effective gas diffusivity. As a result, spray (spume) can provide a large contribution to the air-sea gas transport under tropical cyclone conditions. Furthermore, in these conditions, the gas flux is no longer expected to depend on gas solubility (Andreas et al. 2017). Remarkably, in a series of laboratory experiments conducted in two high-speed wind-wave tanks (Kyoto University and the SUSTAIN facility, RSMAS, University of Miami) with 12 tracer gases, Krall et al. (2019) and Jähne (2019) have found that under tropical cyclone conditions, the dependence of the air-sea gas transfer velocity on gas solubility is significantly reduced or eliminated. To make a computational insight into the process of the spume generation, we have implemented a multiphase computational fluid dynamics model, ANSYS Fluent's Volume of Fluid to Discrete Phase Model (VOF to DPM) with dynamic remeshing. VOF to DPM generates liquid spray particles and converts those that are close to being in a spherical shape into Lagrangian particles. Lagrangian particles participate in a two-way interaction with the airflow. Non-spherical spray particles travel as liquid particles. The VOF to DPM model computes the sea spray generation function (SSGF) directly at the air-sea interface. The dimethyl sulfide (DMS) gas transfer velocities estimated from this model for Category 1, 3, and 5 tropical cyclones are close to the Krall et al. (2019) laboratory observations. Also, the model estimates for air-sea oxygen transfer velocities are close to those inferred from the field measurements of McNeil and D'Asaro (2007) beneath Hurricane Frances 2004.

## Poster: 3-D Imaging of Air-Water Gas Exchange

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In 2014 Kräuter et al. demonstrated 2-D fluorescence imaging of the mass boundary layer thickness at a wind-driven wavy water surface. This technique was later used in the large Heidelberg Aeolotron to study the effect of microscale wave breaking (Kräuter 2015) and the fetch dependency of gas transfer (Klein 2019).

Because of the chemical setup used to image the boundary layer, it is not suitable to infer the gas transfer velocity directly from the images. In this contribution a modified setup is discussed which allows for local gas transfer measurements from the images. This technique is based on a concept worked out almost 40 years ago (Jähne, 1985, Chapter 6). Because of technical limitations at that time, only a few single images could be taken back then. The recording of high frame rate image sequences requires dedicated high intensity light sources and sensitive high-speed cameras and could only be realized recently in a small linear wind-wave tank (Papst 2019).

In the Aeolotron, the fluorescence in the vicinity of the water surface will now be acquired with cameras from below the facility and not from above as in previous investigations (Kräuter 2015, Klein 2019). This arrangement makes 3-D imaging possible, because concentration fields below the water surfaces, which have detached from the mass boundary layer are not distorted by refraction at the wavy water surface. Taking up to nine cameras running with more than 600 fps, we hope to perform a 3-D reconstruction of the shape of the water surface and to separate the mass boundary layer at the water surface from structures swept down into the bulk water by microscale wave breaking or other surface renewal events using light field imaging techniques (Wanner and Goldlücke 2013).

The fluorescence imaging technique can be used simultaneously with thermographic measurements. In this way it is possible to measure both the mass and heat transfer velocity across the aqueous boundary layer at the same small footprint. If this approach is successful, the Schmidt number exponent can be measured within a few seconds locally also under non-stationary conditions.

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## Poster: High Resolution Vertical SO<sub>2</sub> Profiles in the Vicinity of a Wavy Wind-driven Water Surface Measured via LIF

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In a small linear wind-wave facility, Friman and Jähne (2019) demonstrated that it is possible to measure vertical air-side sulfur dioxide concentration profiles with laser induced fluorescence (LIF) with sufficient resolution to resolve the mass boundary layer, even though a suboptimal fixed excitation wavelength of 223.7 nm was available.

In this contribution an optimized system is introduced that will be used in the large Annular Heidelberg Air-Sea Interaction Facility, the Aeolotron. For fluorescence excitation an InnoLas SpitLight Compact OPO-355 with UV extension is used to tune the excitation wavelength between 220 and 280 nm with a pulse energy of about 5 mJ at 20 Hz. In a first round of experiments, the optimum excitation wavelength was selected in order to achieve maximum fluorescence at the minimum possible absorbance.

Sulfur dioxide is an ideal tracer not only to study the transfer of highly soluble gases which are controlled by the air-side transfer resistance, but also to which extent the transfer velocity fluctuates and changes with the phase of the wind waves. If the pH value is lowered to values of about 3, the effective solubility of sulfur dioxide reduces as well and reaches values where the transfer is partitioned between air and water. The partitioning ratio can be measured directly from the concentration sulfur dioxide reaches at the water surface.

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## Poster A Novel Active Thermographic Technique to Investigate the Water-sided Shear Layer at a Wind-driven Wavy Water Surface

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Several attempts have been made to measure the shear at a wind-driven water surface by thermographic techniques. The basic idea is to heat a thin line perpendicularly to the wind direction by a short pulse of infrared radiation and to observe how the line is getting wider with time. This happens by an interplay between heat diffusion and the shear current. This technique does not work well, if the heat flux is applied at the water surface, e. g., by a 10.6  $\mu\text{m}$  carbon dioxide laser with a penetration depth of only about 12  $\mu\text{m}$ . The best choice is a heat source which penetrates into the water with a depth of about the thickness of the viscous boundary layer.

In this contribution an optimized system is introduced that has been set up at the Heidelberg Aeolotron. It consists of a diffraction-limited 100 W Erbium fiber laser. Its wavelength of 1550 nm penetrates about 1.5 mm into the water. A beam expander is used to form a thin 100 to 200 mm wide line at the water surface. Infrared image sequences at 100 fps were taken with an Infratec ImageIR 9420 hp MF cooled thermal camera.

First test measurements were performed at non-steady conditions with quickly switching on and off the wind. The image sequences reveal a complex and intermittent velocity field and shear stress at the water surface, which entirely changes within seconds when the wind is turned off. Surface renewal events wipe out the heated line in a very short time span.

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