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Contents

Abstracts are listed in alphabetical order by author surname. Click on the abstract title/author to navigate to the corresponding page to read the full abstract.

Influence of carbonate chemistry on mangrove-dominated estuarine system and carbon dioxide fluxes in Indian Sundarban Avanti Acharya	1
Laboratory investigation of significant gas transfer enhancement via capillary-gravity bow waves Katherine Adler	2
Comparing in-situ and earth-observation derived CO ₂ fluxes to assess uncertainties in global estimates Ian Ashton	3
Seasonal and Diurnal Variations in Organic Matter Composition Influence the Biogenic Surfactant Pool in the Coastal Baltic Sea Theresa Barthelmeß	4
Relationships between CO ₂ gas transfer velocity, radar backscatter and wave properties Thomas Bell	5
Greenhouse gases in the urban Clyde estuary: Physical estuarine processes and nutrient loading impact greenhouse gas generation Alison Brown	6
Energy dissipation-based estimates of whitecap coverage and air entrainment rates in whitecaps Adrian Callaghan	7
Observations of breaking wave bubbles and air entrainment in varying wave and wind conditions Rui Cao	8
Towards estimating the air-sea gas exchange velocity from a statistical reconstruction of observations of ocean turbulence Giulia Carella	9
The role of chemistry in air-sea fluxes Lucy Carpenter	10
Bubble size distributions in spilling breakers with different phase shifts Konstantinos Chasapis	11
The Thermal Signature of the Residual Foam in Breaking Waves Chris Chickadel	12
Jungle BOOGIE: Investigating the impact of terrestrial organic matter on surfactant control of air-water gas exchange along a land-ocean tropical river transect Beth Cowling	13
Bubble size distributions measured in high wind conditions, and their potential contribution to oxygen uptake during winter storms Helen Czerski	14
Understanding and modeling bubble mediated gas transfer by breaking waves Luc Deike	15

Air-sea gas exchange in a seagrass ecosystem Ryo Dobashi	16
Near-surface stratification biases the Arctic and global air-sea CO ₂ flux estimates Yuanxu Dong	17
Uncertainties in eddy covariance air-sea CO ₂ fluxes and implications for gas transfer velocity parameterisations Yuanxu Dong	18
The effect of non-local processes on eddy covariance air-lake gas fluxes Leonie Esters	19
Turbulence-based air-sea CO ₂ exchange in the Jade Bay Leonie Esters	20
The effect of biogenic surfactants on the spatial variability of surface water temperature under low-wind conditions Mehrshad Foroughan	21
Air-water momentum exchange in Lake Geneva under light wind conditions: the effect of natural surfactants Mehrshad Foroughan	22
Multiscale temporal variability of the global air-sea CO ₂ flux anomaly Yuanyuan Gu	23
Controls of air-sea CO ₂ exchange under high and low wind-speed conditions Lucia Gutierrez-Loza	24
Air-sea scalar transfer – effects of wind and waves on equivalent roughness length Tetsu Hara	25
Laboratory measurements of size-dependent spray distributions above both fresh and seawater. Brian Haus	26
Simulation of high-intensity isotropic turbulence driven gas transfer Herlina Herlina	27
Relationship between wind speed and gas exchange in the coastal Baltic Sea David Ho	28
What Do Flux Chambers Really Measure? A Proposal for Comparative Measurements at the Heidelberg Aeolotron Bernd Jähne	29
On the Limitations of Current Field Measuring Techniques and Measurements for Air-Sea Gas Exchange Bernd Jähne	30
IRISS, an IR Radiometer System for Measurement of Skin Temperature from USVs and Buoys Andrew Jessup	31
Nineteen years of surface ocean nitrous oxide along the Atlantic Meridional Transect Jan Kaiser	32
Gas transfer at high wind speeds: extrapolating concurrent CO ₂ /DMS field measurements to SF ₆	

Kerstin Krall	33
How to cross-link lab and field measurements	
Kerstin Krall	34
Sufficiently Realistic Simulation of Oceanic Conditions for Air-Sea Gas Exchange at the Re-Engineered Heidelberg Aeolotron	
Kerstin Krall	36
The impact of rain on ocean surface waves and currents	
Nathan Laxague	38
Observations of mean and wave orbital flows in the upper centimeters of the ocean surface layer	
Nathan Laxague	39
CLAW: Dead or Alive?	
Peter Liss	40
Working Towards Improved Gas Transfer Prediction by Understanding the Impact of Gustiness on Momentum Fluxes	
Meng Lyu	41
Near-surface Turbulence in Arctic, Temperate, and Tropical Inland Waters: Implications for Gas Fluxes	
Sally MacIntyre	42
Concurrent, open ocean eddy covariance flux measurements of dimethylsulfide and carbon dioxide: What have they taught us about gas transfer and what should we do next?	
Christa Marandino	43
Airborne observations over the North Atlantic Ocean reveal urea is a missing component of atmospheric reduced nitrogen	
Emily Matthews	44
Rethinking Arctic Ocean CO ₂ Fluxes	
Lisa A. Miller	45
Measurements of surface-cooling induced gas-transfer using fluorescence-lifetime imaging (FLI) technique	
Erni Murniati	46
Constraining the role of the surface micro layer in tropical riverine headwaters of Amazonia	
Sevda Norouzi Alibabalou	47
The CO ₂ fluxes at the ocean-atmosphere interface on the Brazilian continental shelf: a review of its behavior as a source or sink of atmospheric CO ₂	
Raquel Oliveira	48
Statistical distributions of whitecap variables using a novel remote sensing technique to detect and track individual whitecaps in digital sea surface images	
Joe Peach	50
Breathing Oceans: understanding the role of surface-active organic matter composition in the ocean skin layer to modulate gas exchange between the atmosphere and ocean	
Ryan Pereira	51
Air-sea exchange of acetaldehyde, acetone and DMS at a UK coastal site.	

Daniel Phillips	52
Shipboard Infrared and Visible Remote Sensing of Whitecaps	
Henry Potter	53
A Field Experiment to Determine the Impact of Nearshore Processes on Air-Sea Mass, Momentum, and Heat Fluxes	
Henry Potter	54
Wind and fetch dependent gas transfer velocity in an Arctic sea-ice lead determined from eddy covariance CO ₂ flux measurements	
John Prytherch	55
The role of sea ice in CH ₄ and CO ₂ air-sea gas transfer in the central Arctic Ocean	
John Prytherch	56
pCO ₂ gradient in the near surface ocean	
Mariana Ribas Ribas	57
Surfactant control on air-water gas exchange in freshwater lakes	
Philippa Rickard	58
Should we account for the skin temperature effect in model simulations?	
Andrea Rochner	59
Gas transfer velocities for greenhouse gases (CO ₂ , CH ₄ , N ₂ O) along the river-estuary continuum	
Judith Rosentreter	60
Using land-based stations for air-sea interaction studies, issues with land influence and non-stationarity	
Anna Rutgersson	61
Bubble break-up and the formation of sub-Hinze scale bubbles in turbulence	
Daniel Ruth	62
Testing and application of a diffusion-based method for sampling DMS in the Sea Surface Microlayer	
Alexia Saint-Macary	63
Evidence that differences between the dominant drivers of surface air-sea exchange and those of surface cross-shelf transport are controlling continental shelf-sea carbon sinks	
Jamie Shutler	64
Reassessing the 4.33% / °C constant used for temperature partitioning of the inorganic carbonate system, how do derived constants impact the air-sea CO ₂ flux?	
Richard Sims	66
Direct flux measurements of carbon dioxide and methane in the Canadian Archipelago in variable sea ice conditions	
Richard Sims	67
Quantifying the decadal and global scale impact of tropical cyclones on the ocean carbon sink using remote sensing, in situ and models	
Richard Sims	68
On the parameterisation of air-sea gas transfer of CO ₂ via wave breaking energy dissipation rate	
Andrew Smith	69

Modeling Air-Sea Gas Transfer Under Tropical Cyclone Conditions Alexander Soloviev	70
Air-sea gas exchange and connections to bubble fluxes at high wind speeds, as revealed by noble gases Rachel Stanley	71
Air-sea gas exchange fluxes and steady state saturation anomalies at very high wind speeds, as revealed by noble gases Rachel Stanley	72
Greenhouse gas fluxes over a boreal river measured with eddy covariance Aki Vähä	73
Greenhouse gases (CO ₂ , CH ₄ and N ₂ O) emissions from a tropical micro- tidal estuary (Cochin, India) Sudheesh Valliyodan	74
A thermographic approach to measure the wind shear stress at the water surface Philipp Immanuel Voigt	75
Diurnal BOOGIE: An investigation into spatiotemporal and climate change effects on organic matter in the sea surface microlayer and its movement between marine and atmospheric environments Katrina Walker	76
Global estimates of air-sea CO ₂ fluxes: Contributions of Wallace Broecker and Taro Takahashi Richard Wanninkhof	77
New, substantially larger, estimates of global air-sea CO ₂ flux from surface data Andrew Watson	78
The effects of surfactants on air-water gas transfer. Jan Wissink	79
Underway seawater and atmospheric measurements of volatile organic compounds in the Southern Ocean Charel Wohl	80
Sea ice concentration impacts dissolved organic gases in the Canadian Arctic Charel Wohl	81
The peculiar characteristics of air-water gas transfer across a broken surface David Woolf	82
Autonomous eddy covariance system for long-term shipboard measurements of CO ₂ flux - setup, data processing, and uncertainty analysis Mingxi Yang	83
Natural variability in air-sea gas transfer efficiency of CO ₂ Mingxi Yang	84
Global synthesis of air-sea CO ₂ transfer velocity estimates from ship-based eddy covariance measurements Mingxi Yang	85
Whitecaps Identification and Extraction in Infrared Imagery Using Machine Learning Xin Yang	86

The Impact of Wind Gusts on the Ocean Skin Layer
Christopher J Zappa 87

Using Ship-Deployed High-Endurance Unmanned Aerial Vehicles for the Study of Ocean Surface and
Atmospheric Boundary Layer Processes
Christopher J Zappa 88

Influence of carbonate chemistry on mangrove-dominated estuarine system and carbon dioxide fluxes in Indian Sundarban

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Abstract

We investigated the response of estuarine system in Indian Sundarban to varying carbonate system parameters like total alkalinity (TA) and dissolved inorganic carbon (DIC), pH and $p\text{CO}_2$ along with salinity, water temperature, chlorophyll-a and dissolved oxygen (DO) from rivers Saptamukhi, Thakuran, and Matla from October 2012 to August 2019. Using TA-DIC couple, the carbonate system parameters like carbon dioxide partial pressure ($p\text{CO}_2$), bicarbonate (HCO_3^-), carbonate (CO_3^{2-}), saturation states of calcite and aragonite (ΩC and ΩA) and Revelle Factor (RF) were calculated along with CO_2 flux (FCO_2). Results show significant negative correlations of DIC/TA ratio with oxygen saturation (DO%) and pH. Furthermore, DO% shows an exponential negative correlation with $p\text{CO}_2$. These results indicate that the estuarine DO% and pH are sensitive to increase in DIC and TA levels which results in an exponential rise in $p\text{CO}_2$ in the system. Dissolved $p\text{CO}_2$ rises to a maximum of 914 μatm in the waters during monsoon consequent to possible conversion of incoming DIC from land run-off. Increasing DIC/TA ratio also corresponds with High Revelle Factor (12-16) meaning decrease in buffering capacity of the waters. However with DIC/TA ratio beyond 1, the Revelle Factor falls sharply. The rising ratios did not affect calcite and aragonite saturation states which were found well above 1 although a sharp seasonal drop was observed from pre-monsoon to monsoon. The carbon dioxide fluxes also remained feeble between 0.67 ± 1.47 and 1.96 ± 3.63 $\text{mmol m}^{-2} \text{h}^{-1}$. Further, DO% remains 90-96% indicating the surface depletion is not severe. This indicates that seasonal productivity (average chl-a at 3.6 ± 1.8 $\mu\text{g l}^{-1}$) and marine water intrusion into the waters optimise the elevated dissolved CO_2 levels through consumption and dilution processes respectively thereby modulating the negative impacts of increasing DIC/TA ratio by consuming CO_2 . But the significant fall in buffering capacity (high Revelle Factor) is a cause of concern and might indicate the need to regulate anthropogenic carbon footprints in the mangrove-dominated areas to retain this natural compensation.

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

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Abstract

Air-water gas exchange increases significantly at the onset of wind waves (e.g., Kanwisher, 1963; Broecker et al., 1978), and this gas transfer rate, k , scales with the waves' mean square slope (Jähne et al., 1987). Capillary-gravity waves are of particular interest because they are ubiquitous on water surfaces exposed to wind and are steeper and shorter than gravity waves. Saylor and Handler (1997) isolated non-breaking capillary waves and achieved almost two orders-of-magnitude enhancement in k compared to the static case, over a 0.016-m² cross-sectional surface area. Analytical models have yet to explain this dramatic increase, predicting up to only 3.5-fold enhancement due to the steepest capillary waves (Szeri, 1997; MacIntyre, 1971). Isolating the effects of these waves from the effects of wind-driven shear is difficult over a large area.

To further investigate the interfacial scalar flux enhancement due to capillary-gravity waves over a large area, several reaeration experiments were conducted in a straight, open channel, recirculating flume. Capillary-gravity bow waves were generated over a 2.7-m² area using an array of hanging 3.2-mm-diameter cylinders mounted on a motor-driven conveyor belt. Cases with no cylinders, stationary cylinders, and moving cylinders were compared at several relative (water to cylinder) velocities. So far, the presence of the capillary-gravity bow waves increases gas transfer velocity by at least ~40% at modest relative velocities of about 30 cm/s. Analytical capillary-gravity wave dynamics models suggest that the maximum wave slope (ak) of these waves increase with relative velocity, but the function describing this dependence varies with which assumptions about the applied external pressure field or force are made (Chevy & Raphaël, 2003; Raphaël & de Gennes, 1996). Experiments with higher relative velocities are in progress. The results of these experiments will be used to derive an empirical model relating gas transfer enhancement due to these waves and relative velocity, and thus wave steepness.

Note that the impact of these waves in salty water should be greater than that observed in our experiments, which are thus far conducted in fresh water, because reducing surface tension would increase the steepness of these waves. These results further support the significant role that capillary-gravity waves play in the transportation of greenhouse gases between the atmosphere and water bodies, even in the presence of other mixing-driving mechanisms, such as boundary layer shear. These results also provide a path to engineered solutions for carbon capture by using shallow ponds as large-scale air contactors.

Oral presentation

Comparing in-situ and earth-observation derived CO₂ fluxes to assess uncertainties in global estimates

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Abstract

Methods for estimating global air-sea CO₂ flux have evolved rapidly due to research into the environmental controls on air-sea gas transfer, advances in Earth-observation (EO) data and novel in situ technologies for direct surface exchange measurements. However, uncertainties in calculations and global estimates remain, predominately relating to the gas transfer velocity or the interfacial gas concentration gradient. This study uses concurrent air-sea CO₂ flux and gas transfer velocity estimates from eddy-covariance techniques, and indirect techniques based on wind-speed parameterisations from both in-situ oceanographic data and EO measurements in the South Atlantic. All three methods show good general agreement and implicate the subtropical South Atlantic as a significant atmospheric CO₂ sink. The largest differences in net flux between the three estimates result from variation in waterside pCO₂, sea surface temperature and wind speed data. Eddy-covariance estimates were consistently higher than those produced using indirect techniques, which may indicate surface microlayer processes have a strong influence on flux dynamics, as these are not detected using other techniques. Continued work on these data will improve analysis of the accuracy of wind speed-based parameterisations to calculate gas transfer velocities. These results indicate that the focus should be on precisely quantifying waterside pCO₂, sea surface temperature and wind-speed to further reduce uncertainties in global air-sea CO₂ flux estimates.

Seasonal and Diurnal Variations in Organic Matter Composition Influence the Biogenic Surfactant Pool in the Coastal Baltic Sea

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Abstract

Biogenic surfactants can hamper gas exchange by up to 50% in coastal seas, however, their small-scale temporal and spatial dynamics are poorly constrained. Classically, it is assumed that surfactants origin from primary production. Alternative hypotheses suggest that microbial or photochemical degradation of organic matter replenishes the surfactant pool. This study investigated possible biogenic sources of surfactants in the sea surface microlayer (SML) and the underlying water at a coastal Baltic Sea site. The focus was set on amino acids and carbohydrates as the main components of phytoplankton-derived organic matter. We aim to resolve similarities and dissimilarities between two seasons (early summer and autumn 2018). The composition of the biochemicals provided further insights into microbial degradation dynamics and was complemented by flow-cytometry-based community analysis. In total, 76 samples were collected within a radius of less than ~8 NM allowing for high spatial resolution. Moreover, morning and afternoon sampling enabled us to investigate diurnal cycles. In summer, surfactant concentrations were generally lower than in autumn. In summer, surfactant concentration was best explained by the combined effect of the particulate fraction of the non-essential amino acid serine, particulate combined carbohydrates (PCHO), and dissolved organic carbon (DOC). Surfactant and PCHO concentrations were significantly enriched in the SML and followed a pronounced diurnal cycle. In contrast, the surfactant pool in autumn corresponded to a diverse mixture of semi-labile organic matter components, represented best by the dissolved fractions of glucose and the essential amino acid isoleucine. Surfactant concentration correlated significantly with the abundance of nano-phytoplankton cells. Therefore, we hypothesize that the surfactant pool is mainly composed of recalcitrant organic matter components that resist rapid microbial degradation. Elevated surfactant concentrations, on the other hand, are triggered by the release of fresh organic matter. While the effect of the resistant but less surface-active stock is potentially longer-lasting, the effect of labile, highly surface-active agents on gas exchange may diminish within days.

Oral presentation

Relationships between CO₂ gas transfer velocity, radar backscatter and wave properties

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Abstract

The gas transfer velocity of carbon dioxide (K_{CO_2}) is influenced by surface turbulence or roughness, but in practice is typically parameterized using wind speed. Satellite microwave backscatter/roughness measurements are typically used to infer sea surface wind stress/speed. Wind speeds determined from satellite measurements of roughness are used to estimate the global CO₂ air/sea flux. CO₂ air/sea fluxes could be estimated more directly using K_{CO_2} parameterized from satellite observations of sea surface backscatter.

We have made the first concurrent observations of K_{CO_2} and high resolution, shipborne C-band synthetic aperture radar (SAR) backscatter during an Atlantic Meridional Transect cruise (AMT-28, Oct. 2018). K_{CO_2} data were derived from air/sea CO₂ concentration differences and eddy covariance flux observations ($K_{CO_2} = F_{CO_2}/\Delta CO_2$). Data were collected at wind speeds between 4 m/s and 14 m/s, which encompasses the wind speed range where waves break and bubbles are formed. In situ wind speed explains approximately half of the variance in the AMT-28 K_{CO_2} data. Polarized SAR data are used to assess the different contributions to surface roughness (e.g. from non-breaking and breaking waves). Different polarizations and incidence angles were investigated, with the horizontal-vertical polarization at 40° explaining more of the variance in K_{CO_2} than the wind speed.

K_{CO_2} -wind speed relationships from previous studies diverge at intermediate-high wind speeds. Recent work suggests that some of the variability in K_{CO_2} may be explained by accounting for wave field properties. We will discuss whether a combination of wave information from the ECMWF wave model (ECWAM) and satellite backscatter retrievals can be used to predict the wind-wave dependence of K_{CO_2} and thus estimate global CO₂ fluxes.

Greenhouse gases in the urban Clyde estuary: Physical estuarine processes and nutrient loading impact greenhouse gas generation

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Abstract

Biologically productive regions such as estuaries, even though they only cover a small percentage of the world's oceans, contribute significantly to methane and nitrous oxide emissions. This paper synthesises greenhouse gas (GHG) and nutrient data measured in the Clyde estuary; including data measured at the near-surface and near-bed, through the tidal cycle and longitudinally through the estuary to determine the main physical and biogeochemical mechanisms that influence GHG sources and sinks, and ultimately lead to high GHG evasion. The Clyde estuary, an urban mesotidal system, is often highly stratified with reduced mixing and a high loading of nutrients both from agricultural and urban wastewater sources. The physical processes within the estuary are strongly influenced by river flow and tide, which significantly impact the saline extent and flushing times. These, together with nutrient loading, impact the physical water properties and the amount and location of GHG generation. Nitrous oxide (N_2O) concentrations can be predicted in both the upper fresh and lower saline layers throughout the inner estuary primarily by consideration of: the total dissolved nitrogen (TDN) concentration, oxygen saturation and conductivity. The linear increase in the percentage of TDN converted to N_2O with decreasing oxygen saturation implies that denitrification is triggered in lower oxygen conditions. The consistency of the prediction in both layers suggests there is limited diffusion of N_2O through the pycnocline until mixing occurs. The apparent rapid response of N_2O production to increases in TDN concentration in the surface layer implies that most nitrogen processing is occurring in the water column despite the low turbidity. Methane (CH_4) concentrations are more variable and show clear differences between the upper fresh and lower saline layers. Methane in the surface layer, above the saline intrusion, typically increases and peaks at the start of the surface saline transition. This increase within the estuary before subsequent dilution confirms that CH_4 generation occurs within the estuary rather than only being passed from the river. Methane concentrations within the lower saline layer are significantly higher than the surface layer suggesting generation within the bed and for a specific salinity regime they increase in low oxygen conditions. The prolonged low river levels that occurred between June and September 2021, and caused high salinity throughout the inner estuary, appear to prevent CH_4 generation. Understanding these dynamics helps to improve our knowledge of estuarine environments and their potential for GHG release to atmosphere.

Energy dissipation-based estimates of whitecap coverage and air entrainment rates in whitecaps

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Abstract

Direct estimates of bubble-mediated gas exchange are not well constrained, in part, due to a lack of in-situ data related to air entrainment rates and bubble size distributions in oceanic whitecaps. This has led to the development of some parameterisations of bubble-mediated gas exchange in terms of the coverage of the ocean surface in whitecap foam. These parameterisations in turn rely on parameterisations of either total whitecap coverage (W), or the whitecap coverage associated with actively breaking waves when air is entrained below the sea surface. The majority of W parameterisations are driven by estimates of the 10m (neutral) wind speed. However, wind-speed only parameterisations of W have inherent uncertainty which ultimately feeds into estimates of bubble-mediated gas exchange models. More recently, several authors have progressed the field and developed sea-state dependent W parameterisations which recognise the fact that myriad different sea states can exist at a given wind speed.

Here, we present a model of W that is forced by the energy dissipation rate of the surface wave field, thus allowing W to be easily estimated from modern 3rd generation spectral wave models. The European Centre for Medium Range Weather Forecasts (ECMWF) spectral wave model (ecWAM) was operated at a spatial resolution of 11 km and 1-hour temporal resolution to estimate W . These model W estimates are compared to photographic-based W measurements made in the North Atlantic during the 2006 MAP cruise and show good agreement. The energy dissipation-based W model is then developed and extended to estimate energy-dissipation based air entrainment rates in whitecaps. These model values also show good agreement with more recent sea-state dependent parameterisations of air entrainment rates and some existing laboratory data.

Observations of breaking wave bubbles and air entrainment in varying wave and wind conditions

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Abstract

Bubble clouds within the two-phase flow as a result of white-capping is critical to the enhancement of the exchange of mass and gas between the ocean and atmosphere. Upon breaking, bubbles of radius ranging from order tens of microns to centimetres are produced. Therefore, more accurate models of bubble-mediated gas exchange and aerosol production flux require accurate determinations of air entrainment rates and bubble size distributions.

We report experimental measurements of time- and space-evolving bubble size distribution in 2-D breaking waves. The bubbles were measured with high resolution digital images using a range of novel image processing and object detection techniques. A wide range of breaking waves were considered by altering the underlying scale, nonlinearity and spectral bandwidth of the dispersively-focused wave groups. The experiments were initially conducted in the absence of wind, and again under influence of direct wind of varying wind speeds. This is to replicate the effects of different wave age on the breaking process, air entrainment and resulting bubble size distribution.

The experimental results demonstrate that underlying wave scale, non-linearity, spectral bandwidth and wind speed (wave age) all have a measurable influence on the evolution of the two-phase flow and bubble size distributions within the breaking waves studied here, highlighting the complexity of the air entrainment over the breaking process. The relative magnitude and importance of these influences will be discussed in detail in the present study. For instance, compared to breaking waves without wind stress, waves in the presence of wind tend to break at lower wave steepness, resulting in a reduction of total air entrainment and significantly different spatial distribution of bubbles.

Towards estimating the air-sea gas exchange velocity from a statistical reconstruction of observations of ocean turbulence

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Abstract

Although the air-sea gas transfer velocity is usually parameterized with wind speed, there are attempts to relate it to oceanic turbulence. For example, the so-called small-eddy model suggests a relationship between the gas transfer velocity and the ocean surface turbulence in the form of the dissipation rate of turbulent kinetic energy ϵ .

Available observations of ϵ are spatially and temporally sparse. In this study, we use observations of ϵ and co-located atmospheric and oceanic fields from the ERA5 reanalysis. We apply a Gaussian Process model to this combined data set to investigate the relationship between the observed profiles of ϵ and the ERA5 fields. Using the model, we construct monthly maps of ϵ and estimate the climatological air-sea gas transfer velocity from existing parametrizations based on the small-eddy model. The resulting air-sea gas transfer velocities are validated with independent measurements that again are co-located with fields from the ERA5 re-analysis.

In order to test the impact of the limited sample on the model results, we used simulations from an ocean-based model to perform a perfect-model experiment where a statistical model was fitted only using data limited to the profiles' sampling sites and times. The simulations are monthly runs from NEMO forced with ERA5.

Our results provide a new look at the interaction between wind-induced turbulence, sea state and heat fluxes in driving gas transfer velocity, and point to data sparseness as a major source of uncertainty for large-scale estimation of gas exchange.

Oral presentation

The role of chemistry in air-sea fluxes

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Abstract

The exchange of chemical species across the air-sea interface is a key component of Earth's biogeochemical cycling and exerts a profound influence on the chemistry of the atmosphere with impacts on climate and regional air quality. The sea surface microlayer represents a particularly reactive region that can lead to the production of chemicals and particles and/or modify air-sea exchange rates. For example, deposition of ozone and subsequent reactions at the sea surface is an important global loss term for tropospheric ozone and a dominant production pathway of volatile ozone-destroying halogens. This presentation discusses advances made in understanding sea surface microlayer chemical reactions with ozone and iodine, the exchange of these gases across the air-sea interface, and the subsequent impacts on the atmosphere.

Bubble size distributions in spilling breakers with different phase shifts

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Abstract

The bubbles generated by breaking waves in the open ocean are an important feature of the ocean surface. They affect optical and acoustical properties of the top few meters of the ocean, influence surfactant scavenging, aerosol production and air-sea gas transfer. The bubble populations formed by breaking waves are complex, but can broadly be split into two categories: short-lived larger bubbles which resurface and burst and longer-lasting smaller bubbles which form deep diffuse plumes. The first category appears to dominate the transfer of less soluble gases (such as carbon dioxide). However, our understanding of the mechanistic link between the type of breaking wave and the consequent bubble behaviour is limited. Breaking waves may vary in terms of their steepness, phase and spectrum, and we do not know how these parameters affect bubble populations, their penetration depth and their residence time underwater. The aim of this study is to address this gap in understanding, by examining the breaking process and bubbles generated for a range of different wave types.

Breaking waves in the open ocean are most commonly of the spilling type. In this paper we calculate characteristic bubble size distributions for spilling breakers generated in the laboratory with dispersive focusing. The waves have a prescribed linear target spectrum (a Gaussian spectrum). The method ensures control over the focus location of the breaking wave group and a high level of repeatability of experiments. Experimental runs with different phase shifts of the same amplitude spectrum show that when a peak-focussed wave (zero phase shift) breaks, then wave groups with other added phase shifts break as well. A laser induced fluorescence (LIF) technique is used to capture wave crests. Images from high speed cameras are analysed with an algorithm that extracts the moving shape of the breaking crest. An algorithm processes high speed images of bubbles and detects with a modified Hough transform their circular shapes for radii $r \geq 0.1$ mm. The bubble number per radius is identified in successive snap shots, initiating at wave breaking.

The results from the proposed framework indicate that phase shifted breakers demonstrate different behaviour, that affects distribution of bubbles. Our approach is to parametrise the wave crest shapes and to examine the causal links to derived typical bubble distributions. The observed differences in the latter, suggest that we should further employ our experimental methods to investigate formulation of wave properties in existing bubble generation models.

Oral presentation

The Thermal Signature of the Residual Foam in Breaking Waves

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Abstract

Quantifying energy dissipation due to wave breaking remains an essential but elusive goal for studying and modeling air-sea fluxes of heat, gas, and momentum. Previous observations have shown that lifetimes of bubble plumes and surface foam are directly related to the dissipated energy. Specifically, the foam decay time can be used to estimate the timescale of the subsurface bubble plume and the energy dissipated in the breaking process. A mitigating factor is that the foam decay time can be significantly affected by the surfactant concentration. We present an experimental investigation of a new technique that exploits the thermal signature of cooling foam to infer wave breaking dynamics. The experiments were conducted in a laboratory wave tank using artificial seawater with and without the addition of a surfactant. We show that the time from the start of the breaking process to the onset of cooling scales with the bubble plume decay time and the dissipated energy, and is not significantly affected by the presence of additional surfactants. We confirm observations from the field of the spatial variability of the temperature of foam generated by an individual breaking event, which has implications for inferring the spatial variability of bubble plume depth.

Jungle BOOGIE: Investigating the impact of terrestrial organic matter on surfactant control of air-water gas exchange along a land-ocean tropical river transect

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Abstract

Understanding how the ocean's organic skin layer modulates gas exchange is critical to estimating how the intrinsic oceanic sinks and sources of carbon dioxide (CO₂) and other greenhouse gases are changing, both now and in the future. Organic substances in the skin layer, known as surfactants, are known to influence this gas exchange. Reduced exchange by the suppression of the gas transfer velocity (k_w) from surfactants, known as the 'surfactant suppression effect' (SSE), has been shown to reduce the amount of CO₂ stored annually in the Atlantic Ocean. Surfactants are difficult to characterise through traditional definitions but are known to be derived from multiple organic matter sources undergoing biogeochemical transformations along the land-ocean continuum. The BOOGIE project (Breathing Oceans: understanding the organic skin that modulates the exchange of greenhouse gases between the atmosphere and the ocean) investigates organic matter control of air-water gas exchange along a land-ocean transect across the tropical Atlantic from South America to the African continent. Contributing to this wider research, the Jungle BOOGIE project specifically explores the fate of terrestrially derived organic material transported along the Essequibo River in Guyana, focusing on the connectivity between river and ocean. Previous research has identified the presence of invisible dissolved organic matter (iDOM) during high discharge events, potentially influencing surface enrichment of DOM in the lower river and estuary. We propose to use a combination of nested techniques to analyse DOM compositions and concentrations using fluorescence indexes and stable carbon isotope signatures. This will determine how DOM varies in relation to hydrological changes and whether iDOM is enhancing surfactant suppression of air-water gas exchange in both the river and ocean surface microlayer (SML). Sampling will take place during wet and dry seasons in intensive periods at four key locations, targeting the upper and lower river, estuary and coastal regions. Lab experiments will also be conducted to assess the degradation potential of DOM under different scenarios, including the effects of salinity and photo-oxidation, and the resulting effect on surfactant suppression. We anticipate presenting the first results and interpretation in May 2022.

Bubble size distributions measured in high wind conditions, and their potential contribution to oxygen uptake during winter storms

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Abstract

The bubble-mediated gas transfer caused by breaking waves in the open ocean is a complex and poorly-understood process. We still lack a detailed mechanistic understanding of the formation and evolution of bubble plumes beneath breaking waves and also of the consequent contribution that these plumes make to gas transfer. The major focus has traditionally been on the larger short-lived bubbles which exist in the top metre of the ocean in the few seconds after a wave breaks, because these are thought to make a significant contribution to carbon dioxide transfer. However, a very recent study (by other authors) has suggested that bubble-mediated gas transfer could also be important for oxygen uptake during winter in the open ocean, and that this could explain some inconsistencies in current model predictions. The bubbles most likely to be significant for oxygen transfer are the deeper longer-lasting plumes of very small bubbles.

During the HiWINGS (High Wind speed Gas exchange Study) expedition in the North Atlantic in autumn 2013, bubble size distributions and plume spatial distributions were measured in detail in the top eight metres of the water. During the four major storms when we had the opportunity to conduct bubble measurements, the wind speed varied from 10 m/s to 28 m/s, providing an ideal situation to study deep bubble plumes in detail. Bubble size distributions were measured simultaneously by a specialised bubble camera at 2m depth, and an acoustical resonator at 4m depth (both making continuous measurements at 1 Hz). An upward-pointing sonar at 8m depth provided information about plume morphology. The results suggest that bubbles at 2m depth and below are not rising back to the surface, but remain trapped in the ocean. There are clear patterns in bubble size distributions and void fraction distributions over time. This study uses those data to estimate the bubble-mediated oxygen uptake associated with the observed plumes, and to compare it to the numbers predicted by current models. This preliminary work allows an assessment of the contribution of these bubbles to overall oxygen transfer in high wind conditions, based on direct bubble measurements made at sea.

Oral presentation

Understanding and modeling bubble mediated gas transfer by breaking waves

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Abstract

Physical processes at the ocean-atmosphere interface have a large effect on climate and weather by controlling the transfer of momentum and mass. Without wave breaking, transport between the ocean and the atmosphere is through slow conduction and molecular diffusion, while wave breaking is a transitional process from laminar to turbulent flow. When waves are breaking, the surface experiences dramatic changes, with sea spray ejection in the atmosphere and air entrainment into the ocean water. The complex dynamics and statistics of wave breaking in a particular ocean location depends mainly on the local wave state not the wind velocity, while current parameterizations for ocean-atmosphere interactions are based almost exclusively on the wind speed.

In this talk I will discuss recent efforts towards improving parameterizations of gas transfer through a multi-scale approach. We present a general theoretical framework to account for the complex nature of wave breaking and air entrainment, a two-phase turbulent process, and the very large range of scales involved in the process, from wave statistics scales of order of km, $O(1m-1km)$, to wave breaking dynamics, $O(1-10m)$, air bubble entrainment, bubble dynamics in turbulence and finally bubble bursting at the first surface, $O(\text{microns to mm})$.

The wave and wave breaking statistics are described through spectral representation, through the wave spectrum and the distribution of length of breaking crest, while breaking waves energetics, air entrainment and spray production are investigated by direct numerical simulations and laboratory experiments. We describe the statistics of air bubbles under breaking waves and due to turbulent break-up, as well as the residence time in turbulence and combine these data with a chemical model to estimate the gas transfer of various species.

This theoretical framework can then be implemented in regional and global wave simulations with spectral wave models such as WavewatchIII. We show that bubble mediated gas transfer accounts for about 40% of the total CO_2 flux, with significant seasonal and regional variability. The role of such sea-state dependent variability in global geochemical cycle remains to be tested, which could be done by implementing our formulation in ocean and climate models, and should reduce the uncertainties at moderate to high wind speeds.

Oral presentation

Air-sea gas exchange in a seagrass ecosystem

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Abstract

Seagrass meadows are one of the most productive ecosystems in the world, and could play a role in mitigating the increase of atmospheric CO₂ from human activities. However, to understand their role in the global carbon cycle requires knowledge of air-water CO₂ fluxes, which in turn require knowledge of the gas transfer velocity. Gas transfer velocity was determined using the 3He and SF₆ dual tracer technique in Florida Bay near Bob Allen Keys (25.027°N, -80.681°W) between April 3 and 8, 2015. The average gas transfer velocity, $k(600)$, was 4.67 ± 1.76 cm hour⁻¹. When correlated with wind speed, the results show that gas transfer velocities were lower than previous experiments in the coastal and open oceans, and that using published wind speed/gas exchange parameterizations would overpredict gas transfer velocities in this area. The deviation from other settings was examined from tidal velocity and air-sea temperature difference; tidal amplitude was small implying tidal velocity was weak, and the relationship between the deviation and air-sea temperature difference was weak. It was clear that wind remained the dominant factor driving gas exchange, and the decrease in gas transfer velocity was due to wave attenuation by sea grass and limited wind fetch in this area. A new wind speed/gas exchange parameterization for wind fetch limited environments was produced, which can be applied to other seagrass ecosystems.

Near-surface stratification biases the Arctic and global air-sea CO₂ flux estimates

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Abstract

The global oceans are considered to be a major sink for atmospheric carbon dioxide (CO₂). Air-sea CO₂ flux is generally estimated by the bulk method using a parameterised gas transfer velocity and upper ocean CO₂ fugacity (fCO_{2w}) measurements. The fCO_{2w} and seawater temperature are often taken from a ship's seawater inlet at ~5 m depth (fCO_{2w_bulk} and T_{bulk}) by assuming that the upper ocean seawater is well-mixed. However, in the summertime Arctic, sea-ice melt results in shallow stratification (top ~10 m), which can bias bulk CO₂ flux estimates when the fCO_{2w} measured at ~5 m depth is used. The micrometeorological eddy covariance flux technique is not affected by stratification. Here for the first time, we employ eddy covariance air-sea CO₂ flux measurements during two Arctic cruises to assess the impact of sea-ice melt on Arctic Ocean CO₂ uptake estimates. In sea-ice melt regions, fCO_{2w_surface} values are consistently lower than fCO_{2w_bulk} by an average of 39 μatm. Lower fCO_{2w_surface} can be partially accounted for by fresher (≥27%) and colder (17%) meltwaters. A back-of-the-envelope calculation shows that neglecting the summertime sea-ice melt could lead to a ~10% underestimate of the annual Arctic Ocean CO₂ uptake.

Sea surface temperature (SST) is key for the global air-sea CO₂ flux estimates. Any bias in in-situ SST and/or any upper ocean vertical temperature change such as the cool skin effect can generate a bias in air-sea CO₂ flux estimates. We assess a slight warm bias in the in-situ SST dataset using an accurate drifting buoy SST dataset. We propose that the cool skin correction should vary with latitude. The adjusted estimate of the ocean CO₂ uptake derived from an ensemble of upscaled estimates is able to fill the gap between surface fluxes and the ocean carbon inventory.

Uncertainties in eddy covariance air-sea CO₂ fluxes and implications for gas transfer velocity parameterisations

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Abstract

Air-sea carbon dioxide (CO₂) flux is often indirectly estimated by the bulk method using the air-sea difference in CO₂ fugacity ($\Delta f\text{CO}_2$) and a parameterisation of the gas transfer velocity (K). Direct flux measurements by eddy covariance (EC) provide an independent reference for bulk flux estimates and are often used to study processes that drive K. However, inherent uncertainties in EC air-sea CO₂ flux measurements from ships have not been well quantified and may confound analyses of K. This talk will present a thorough analysis of the uncertainties in EC CO₂ fluxes from four cruises measured with two state-of-the-art closed-path CO₂ analysers on two ships. The mean bias in the EC CO₂ flux is low but the random error is relatively large over short time scales. The relative uncertainty in hourly averaged EC air-sea CO₂ fluxes (cruise-mean) was ~20% during two Arctic cruises that observed large CO₂ flux magnitude. The relative uncertainty was ~50% when the CO₂ flux magnitude was small during two Atlantic cruises. Auto-covariance analysis of CO₂ fluxes suggests that the optimal timescale for averaging EC CO₂ flux measurements ranges from 1–3 hours, which increases the mean signal-to-noise ratio of the four cruises to higher than 3. Applying an appropriate averaging timescale and suitable $\Delta f\text{CO}_2$ threshold (20 μatm) to EC flux data enables an optimal analysis of K.

Oral presentation

The effect of non-local processes on eddy covariance air-lake gas fluxes

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Abstract

Inland freshwater bodies such as lakes provide the largest contribution of natural carbon to the atmosphere. To address this carbon contribution to the atmosphere, eddy covariance flux measurements at lake sites have received increasing attention. The eddy covariance method is formulated for air-water gas fluxes, which are driven by local surface processes. Large-scale non-local processes such as advection or entrainment can potentially add erroneous contributions to the eddy covariance flux estimations. Scalar characteristics from the lakes' topographic surroundings can be horizontally advected to the measurement site. Also, scalar characteristics from the above-lying free atmosphere can be entrained into the boundary layer. The entrained signal can be strong enough to affect the characteristics down to the surface. Both processes will be erroneously captured by an installed eddy covariance instrumentation as local fluxes.

During four years of eddy covariance measurements at Lake Erken, a Swedish freshwater lake, we found that upward carbon dioxide fluxes were observed even when the lake was entirely covered with ice. In order to investigate these unexpected fluxes, we used a statistical approach, which uses only on surface-layer data (van de Boer et al., 2014). This analysis revealed that non-local processes caused the fluxes. The strength and onset of these non-local processes depend on a combination of wind speed and fetch from the measurement tower. The shorter the fetch and the faster the wind, the higher is the contribution of the non-local processes to the measured eddy covariance flux estimates. We corrected the contribution of the non-local processes on the fluxes during the ice-covered and open-water periods. Using the corrected fluxes to calculate the air-lake gas transfer velocity, revealed values that fall closer to frequently used wind-speed based parametrizations than the ones based on the uncorrected fluxes.

Here, we propose analysis methods to correct for eddy covariance measurements at lake sites by the example of Lake Erken. These methods are potentially of great importance for better quantification of the carbon exchange between lakes and other freshwater bodies to the atmosphere.

Turbulence-based air-sea CO₂ exchange in the Jade Bay

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Abstract

An accurate description of the air-sea gas exchange is crucial e.g. for climate predictions. The gas transfer velocity describes the efficiency of the gas fluxes. It is challenging to measure the gas transfer velocity in the field. Therefore, it is commonly parameterized as a function of wind speed, which is a more accessible parameter. However, the gas transfer velocity is known to be actually driven by surface ocean turbulence. Existing uncertainties in the attempts to parameterize the gas transfer velocity with turbulence are caused by the lack of accurate observations of oceanic turbulence. Conventional methods to measure oceanic turbulence are temporally and spatially restricted. In order to overcome the restrictions and gain observational information on turbulence, we combine oceanic turbulence measurements with measurements of the carbon dioxide (CO₂) flux in the same measurement platform: the unique Sniffle buoy. The turbulence is measured with a Nortek Signature1000 Acoustic Doppler Current Profiler (ADCP) as well as two Acoustic Doppler Velocimeters (ADV). The CO₂ flux and air/water pCO₂ concentrations are measured with a SubCtech surface buoy and floating chamber technique. The measurements are conducted in the coastal Jade Bay of the North Sea. Existing uncertainties are particularly high in coastal regions, which feature higher dynamic variability than the open ocean. Our final aim is to reduce uncertainties in the air-sea gas exchange descriptions in coastal areas and better understand the specific processes that control the exchange. Based on the observations, the Jade Bay in autumn 2021 and winter 2022 is a sink of CO₂. This study highlights the importance of multidisciplinary research within the SOLAS (Surface Ocean Lower Atmosphere Study) community, with the need to merge biogeochemistry and physics on individual projects.

The effect of biogenic surfactants on the spatial variability of surface water temperature under low-wind conditions

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Abstract

Natural slicks are manifested by changing the reflective property of water surfaces through inhibition of wave-growth and damping short gravity-capillary waves. Such influence on near-surface hydrodynamics can affect the air-water exchange of momentum, heat, and gas. Here, we present observations from a field campaign carried out in Lake Geneva in the Spring of 2019, which show the response of Lake Surface Water Temperature (LSWT) to transient light winds. For this research, we used an autonomous catamaran equipped with near-surface water temperature sensors, a weather station, and an RGB camera taking slanted-view images of the water surface. The second imagery package included an LWIR camera carried by a balloon, providing thermal images with a resolution of $O(1\text{m})$ from an altitude of about 500m. As our result demonstrates, shortly after wind speed exceeded a threshold ($U_{10} > 2\text{m/s}$), smooth and rippled patches appeared on the surface, resembling slick/non-slick regions commonly observed on water bodies. Such patterns have an exact correspondence with surface temperature contrasts of up to 2°C . To associate these surface patches with the wave-damping effect of biogenic surfactants, the enrichment factor of Fluorescence Dissolved Organic Matter is measured as a proxy from samples collected during other field campaigns. The results point to the spatial variability of LSWT at the sub-pixel satellite scale. It is shown that such warm and cool patches can persist on the water surface after waves disappeared in very low-wind conditions ($U_{10} < 2\text{m/s}$). Hence, such interaction between transient wind and random-like distribution of natural slicks leads to heterogeneous near-surface mixing. This can, in turn, play a role in the generation of intermittency of air-water fluxes, particularly during strong lake stratification and low-wind conditions.

Air-water momentum exchange in Lake Geneva under light wind conditions: the effect of natural surfactants

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Abstract

Randomly distributed patches of smooth surfaces are readily observed on most water bodies. They are called natural slicks when biogenic surfactants in the surface microlayer accumulate above a certain threshold. Slicks typically form under low wind conditions ($< 6 \text{ m s}^{-1}$), having spatial scales from tens of meters to kilometers. They suppress the formation of wind-induced Gravity-Capillary Waves (GCW), leading to altered surface reflectance of light and microwaves, and can also affect near-surface convective motions. Therefore, it is of interest to understand how slicks affect the air-water exchange of momentum, heat, and gas, which can influence the biogeochemical dynamics in the near-surface layer of lakes and oceans. We examined the spatiotemporal variability of momentum flux caused by slicks in Lake Geneva using eddy covariance instrument setups mounted on an autonomous catamaran during several field campaigns. These measurements were combined with aerial and shore-based imagery (both RGB and thermal). We also sampled surface microlayers in an accompanying boat to determine whether visually-identified smooth patches had higher concentrations of fluorescent dissolved organic matter, a proxy for natural surfactants. Using wavelet analysis, we investigated short-time $O(1 \text{ min})$ averaged air-water momentum flux variations associated with the transition from smooth slicks to rough surface areas, which could not be captured by conventional eddy covariance analysis. Results suggest that under light wind conditions and in the absence of short GCW, wind stress cannot effectively be transferred to the water leading to a reduction of momentum exchange within slicks in comparison to the surrounding non-slick areas. The resulting slick-induced horizontal gradients in vertical mixing can contribute to spatial variability in surface temperature and near-surface heat content, which in turn affects air-water exchange processes.

Multiscale temporal variability of the global air-sea CO₂ flux anomaly

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Abstract

The global air-sea CO₂ flux (F) impacts and is impacted by a plethora of climate-related processes operating at multiple time scales. In bulk mass transfer formulations, F is driven by the partial pressure difference between air and water ($\Delta p\text{CO}_2$), and by physico- and bio-chemical factors such as the gas transfer velocity, sea surface temperature and salinity—all varying at multiple time scales. To deconvolve the impact of these factors on F variability at different time scales, time-resolved estimates of F were computed using a global data set assembled between 1988 and 2015. F anomalies were defined as temporal deviations from the 28-year averaged value. Spectral analysis revealed four dominant time scales of variability in F – subseasonal, seasonal, interannual, and decadal with relative amplitude differences varying across regions. A second order Taylor series expansion was then conducted along these four timescales to separate drivers across differing regions in space. The analysis showed that on subseasonal time scales, wind speed variability explains some 65% of the global F anomaly and is the dominant driver. On longer timescales (i.e., seasonal, interannual and decadal timescales), the $\Delta p\text{CO}_2$ effect, controlled by the $\Delta p\text{CO}_2$ anomaly, explained much of the F anomaly. On decadal timescales, the F anomaly was almost entirely governed by the $\Delta p\text{CO}_2$ effect with large contributions from high latitudes. Finally, the drivers of the $\Delta p\text{CO}_2$ effect was shown to be closely connected with the atmospheric $p\text{CO}_2$ anomaly and nonthermal component of oceanic $p\text{CO}_2$ anomaly conjectured to be associated with dissolved inorganic carbon and alkalinity.

Oral presentation

Controls of air-sea CO₂ exchange under high and low wind-speed conditions

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Abstract

Coastal and marginal seas play a relevant role in the global carbon system. In terms of air-sea CO₂ fluxes, these regions represent a disproportionately large contribution compared to the open ocean. The spatial and temporal variability of the fluxes in these regions is often attributed to the heterogeneity of the physical and biogeochemical processes. In the Baltic Sea, the pCO₂ in the seawater presents large spatial and seasonal variability caused by different physical forcings and biogeochemical processes. Such mechanisms also play a key role on modulating the efficiency of the transfer across the air-sea interface. Further understanding of the relevant driving mechanisms in these areas is essential to address the variability of the CO₂ fluxes and their contribution to the regional budgets.

In this study, we evaluate the effect of forcing mechanism on the air-sea CO₂ flux under different wind-speed regimes. The fluxes and associated gas transfer velocities (k_{660}) are calculated from eight years (2013-2021) of eddy covariance data and sea surface pCO₂ measurements from the station Östergarnsholm in the Baltic Sea. The resulting k_{660} values show a good agreement with commonly used wind-based parameterizations, but only under particular conditions on both sides of the interface. The mechanisms that seem to explain the remaining observed variability in k_{660} vary significantly depending on the wind-speed regime. At high wind speeds ($U_{10} > 8$ m/s), both atmospheric and water-side controls are necessary to explain the rapid increase of k_{660} . Water-side processes are well known to be of relevance in the transfer of slightly soluble gases. However, at high wind speeds, atmospheric stability conditions, relative humidity and the enthalpy fluxes seem to play a relevant role in modulating the effect of the water-side processes on the gas exchange. On the contrary, at wind speeds lower than 6 m/s, atmospheric controls are not relevant for the exchange, and processes such as water-side convection seem to explain most of the variability.

Oral presentation

Air-sea scalar transfer – effects of wind and waves on equivalent roughness length

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Abstract

The air-sea transfer coefficients of heat, humidity, and soluble gases (air-side controlled fluxes) are normally assumed to be independent of wind speed or sea states. However, the scalar transfer coefficient depends on both the equivalent roughness of the mean wind profile (z_o) and the equivalent roughness of the mean scalar concentration profile (z_s). Since z_o is known to increase significantly as wind speed increases and/or the sea surface becomes rougher, z_s must simultaneously decrease so that the transfer coefficient remains unchanged. In this study we employ large eddy simulations to investigate how z_o and z_s are modified by surface waves of different wavelength, amplitude, and direction. The results confirm that z_s generally decreases as z_o increases. However, the mechanism of z_o increase is strongly dependent on strength and frequency of airflow separation events that determine the pressure form drag. Whereas, the decrease of z_s is mostly related to the decrease in the frictional stress along the air-water interface. Therefore, the cancellation of z_o increase and z_s decrease is not always perfect, that is, the scalar transfer coefficient can depend on sea states in a complex manner. Since existing studies suggest that the sea state dependence of z_o is particularly important in high wind speeds (storms) and in coastal (shallow water) regions, it is possible that z_s also becomes more sea state dependent in such conditions.

Laboratory measurements of size-dependent spray distributions above both fresh and seawater.

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Abstract

The size-dependent production of spume particles in high wind conditions and their transport away from the air-water interface is necessary to understand the interfacial fluxes of heat, momentum and gases. Because of the inherent difficulties of directly measuring spray exiting the ocean surface in highly energetic conditions, investigators have instead typically observed the size-dependent spray concentration just above the surface and have estimated the spray production based on empirical functions. This laboratory study follows this strategy but extends the range of observed winds to hurricane strength. Previous work has also focused predominantly on the marine environment, with the assumption that the spray production is primarily a mechanical process. Spray dynamics in non-seawater bodies have not been extensively studied, and any significant differences between sea and freshwater remain unquantified. To address this gap, we have conducted the first laboratory experiments directly comparing spume concentrations above fresh and real seawater for 10-m equivalent wind speeds of 36-54 m/s. Droplets in the air above the intensely breaking wind-waves were directly observed and their distribution as functions of wind speed, height, and droplet radius was compared between the two water types. From these unique measurements, parameterizations of the spray generation source functions have been developed, which can be incorporated into numerical models. Substantially higher concentrations of seawater spume were observed as compared to freshwater across all particle sizes and wind speeds. The seawater particles' vertical distribution was concentrated near the surface, whereas the freshwater droplets were more uniformly distributed. Seawater and freshwater height-dependent distributions also exhibited different wind-speed dependences. Incorporating these new concentration dependencies in the interfacial transfer models may enable improved representation of spray mediated fluxes. These findings were generally unexpected and point to an unanticipated role of physiochemical processes in the spume generation mechanism which may impact spray-mediated flux parameterization over water bodies of different salinities. This may also be critical to future lab-based spray studies which up to this point assumed droplets produced from fresh and sea water were essentially interchangeable.

Simulation of high-intensity isotropic turbulence driven gas transfer

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Abstract

It is known that the atmospheric gas (high-Schmidt number) transfer process is characterised by a concentration boundary layer thickness that is smaller than the Kolmogorov length scale of the turbulent flow. This fact has posed a challenge for numerical, laboratory and field experiments, as an extremely fine resolution is required to fully resolve the gas transfer dynamics. Consequently, previous direct numerical simulations (DNS) were limited to low turbulent Reynolds and/or Schmidt numbers. The present work was motivated by the need for highly accurate unbiased data in the high turbulent Reynolds number (R_t) regime. Here, we present the results of a large-scale DNS of gas transfer driven by isotropic turbulence, with an intensity that is significantly higher (R_t up to 1856) than the critical turbulent Reynolds number ($R_t = 500$). To fully resolve all scales, we employed a dual-mesh strategy, where the gas concentration field with Schmidt number $Sc = 500$ was resolved on a finer mesh (65.5×10^9 grid-points) than the flow field (524×10^6 grid-points).

Compared to the data from our previous DNS performed at low turbulent Reynolds numbers, spectral analysis shows that with increasing turbulent intensity the contribution of small-eddies to the turbulent mass flux increases significantly. Furthermore, by comparing snapshots of the surface divergence and the instantaneous transfer velocity, it was observed that footprints of small-scale structures are more pronounced in the gas transfer velocity contour maps than in the surface divergence contour maps. Therefore, the time-averaged spatial correlation between the transfer velocity and surface divergence was found to decrease with increasing turbulent Reynolds number. This indicates that the surface divergence model may not be applicable for high turbulent Reynolds numbers. Also, with the new high-turbulent Reynolds number data, for the first time the existence of the small- and large-eddy regimes, at least in the case of isotropic turbulence driven gas transfer, could be clearly identified numerically.

In addition, the correlation between transfer velocity and near-surface vortical structures was investigated. Because of the flat, free-slip surface boundary condition employed, the turbulent flow was forced to become more and more two-dimensional when approaching the surface causing the vortical structures to become orientated either parallel or normal to the surface. The contribution of both parallel and normal vortical structures to the gas transfer will be discussed.

Oral presentation

Relationship between wind speed and gas exchange in the coastal Baltic Sea

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Abstract

$^3\text{He}/\text{SF}_6$ experiments conducted over the past several decades in the coastal and open oceans show similar results, and indicate that existing wind speed/gas exchange parameterizations are applicable to the coastal and open oceans at moderate wind speeds. Despite these findings, there are still topics that remain underexplored, including whether these parameterizations could be applied to inland seas such as the Baltic. $^3\text{He}/\text{SF}_6$ experiments were conducted in the coastal Baltic Sea, and results show that commonly use wind speed/gas exchange parameterizations for the coastal and open oceans might over-predict gas transfer velocities in that region.

What Do Flux Chambers Really Measure? A Proposal for Comparative Measurements at the Heidelberg Aeolotron

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Abstract

The flux chamber, floating box or direct flux measurement is one of the oldest techniques to measure the gas flux between the ocean and the atmosphere. But its usefulness is highly controversial. Liss and Merlivat [1] wrote in a review paper 1986: “Normal interaction between wind and water is severely inhibited by the presence of the box. Since wind stress is, from the results of wind tunnel experiments, an important control on k_w , there are large doubts as to what the method actually measures, and therefore about its usefulness.” The flux chamber technique is still being used extensively in field measurements, predominately in lakes and estuaries, but also in coastal waters. It has been augmented recently [2] by turbulence measurements to correct for the turbulence distortion introduced by it. But it is still an open question whether these corrections result in correct transfer velocities. Accurate comparisons with other techniques in the field are hardly possible because of the different temporal scales and foot prints of the different techniques.

Accurate comparative measurements are, however, possible in the annular Heidelberg Air-Sea Interaction Facility, the Aeolotron, with a diameter of 10 m and a water channel 0.6 m wide. Because of its geometry, the conditions are homogeneous all over the water interface, in contrast to linear facilities, and a floating chamber can drift freely with the water flow. Therefore a small floating chamber with a diameter of 0.2 m and a Styrodur foam ring was designed. A new technique is proposed to perform fast flux measurements within the chamber by flushing the chamber continuously with nitrogen. In this way transfer velocities can be measured with a time constant better than one minute, well matching the fast gas transfer velocity measurements that are possible in the Aeolotron. Possible tracers for the measurements are oxygen, carbon dioxide and nitrous oxide. Input from the community for the optimal setup and operation conditions of this new approach is very welcome.

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On the Limitations of Current Field Measuring Techniques and Measurements for Air-Sea Gas Exchange

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Abstract

Despite many field measurements of the gas transfer velocity, the measurements cover only a limited wind speed range from about 4 to 20 m/s. Why are no measurements available for lower wind speeds? An analysis shows that none of the currently used field measuring techniques is really suitable for low-wind speeds. Mass balance methods suffer from large time constants and eddy covariance measurements from too low fluxes. Active thermography does not work either, because of the need to heat a too large patch at the water surface. Therefore the community is in need to develop improved or even better novel field measuring techniques.

Secondly, it is investigated what can be learned from the collection of all available oceanic field measurements. Because only the wind speed is given as a parameter for all measurements, only the relation between the gas transfer velocity corrected to a constant Schmidt number and the wind speed can be analyzed. A general regression is applied with an unknown offset at zero wind speed and an increase with any combination from linear to cubic. Using discrete inverse theory, the uncertainty of the estimated transfer velocity is computed as a function of the wind speed. This analysis is based on the hypothesis that the measurements are not biased by any systematic errors and is performed separately for dual tracer and eddy covariance measurements. The main findings are:

- In a narrow wind speed range (for dual tracer measurements between 8 and 16 m/s) the one-sigma uncertainty of the transfer velocity is less than 10%.
- For wind speeds lower than 3 m/s the estimates are too uncertain. It can only be said that the transfer velocity is lower than about 14 cm/h.
- The uncertainties of all regression coefficients are larger than the values themselves. It is not possible to distinguish between a quadratic or cubic increase with the wind speed.

Given the fact that in reality the gas transfer velocity depends on other parameters than the wind speed, it is obvious that it will hardly be possible to infer more complex relations based on field data only. This emphasizes the importance of systematic wind-wave tank experiments to resolve the basic mechanisms under conditions which are as close to oceanic conditions as possible. The predictions gained from such investigations can then be tested with a small number of field experiments under well chosen environmental conditions.

Oral presentation

IRISS, an IR Radiometer System for Measurement of Skin Temperature from USVs and Buoys

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Abstract

The turbulent exchange of heat across the air-sea interface occurs in the topmost layer, making the accurate parameterization of sea surface temperature (SST) of fundamental importance for air-sea process studies. At the surface of the ocean, the typically net upward heat flux results in a cool skin layer of $O(1 \text{ mm})$ in thickness. The temperature at the top of this layer, known as the “skin” temperature SST_{skin} , can be up to 0.5 K less than the temperature directly below.

The impact of the cool skin on gas exchange was first estimated to be significant by Robertson and Watson (1992). Although McGillis and Wanninkhof (2006) later reported its effect is minimal, Woolf et al. (2016) recently revisited its impact and concluded the effect of the cool skin on CO_2 fluxes is large and globally significant. Woolf et al. also noted that warm layers can result in a locally large reduction in gas exchange. Most recently, Watson et al. (2020) applied the corrections for the cool skin and warm layer effects and found an increase in the CO_2 sink by up to 0.9 PgC yr⁻¹.

The advent of reliable uncrewed surface vehicles (USVs) has fostered a growing community consensus for the need to make accurate flux measurements autonomously. Here we report on the development of IRISS (InfraRed In situ Skin System), an infrared radiometer system to remotely measure SST_{skin} under all weather conditions for routine deployment on USVs and buoys. IRISS is designed to be compact, low-power, and cost effective by exploiting the increased stability of commercially-available sensors and is intended to provide measurements with accuracy comparable to current systems.

The S-MODE pilot cruise on the R/V Oceanus took place off the US West Coast in Fall 2021. An IRISS prototype was deployed side-by-side with two ROSR instruments, which are established ship-based systems measuring SST_{skin} with demonstrated state-of-the-art accuracy. The cruise included five Saildrone USVs, three of which included IR radiometer configurations based on the IRISS design. During the course of the 3-week experiment, the Oceanus shadowed the saildrones for extended periods. Here we report on the comparison of the ship- and saildrone-based IRISS sensors to the ROSR measurements. We also compare the measured bulk-skin temperature difference to the COARE model.

Nineteen years of surface ocean nitrous oxide along the Atlantic Meridional Transect

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Abstract

Nitrous oxide is the third most important anthropogenic greenhouse gas and the most important stratospheric ozone depleting substance in terms of current emissions. Approximately 25 % of global nitrous oxide emissions originate from the oceans, coasts and estuaries, produced during microbial nitrification and denitrification. However, the uncertainty around the relative contribution from the ocean is large and our current best estimates range from 11 to 69 % of total emissions.

Only very few open ocean regions have been occupied by sufficiently frequent repeat transects to provide robust constraints on seasonal and internannual variations in nitrous oxide emissions, which would help reduce budget uncertainties, offer potential mechanistic insights into the biogeochemical and physical processes responsible for oceanic nitrous oxide cycling and study the effects of climate change.

Here, we present eight boreal autumn and austral spring ocean surface water datasets for nitrous oxide spanning a 19-year period from 1996 to 2014, acquired during Atlantic Meridional Transect (AMT) research cruises AMT3 to AMT24. These are used to consider whether a change in the surface ocean nitrous oxide budget has occurred during this period. The datasets combine high-resolution (equilibrator-based) and discrete (CTD rosette) measurements, analysed by different instruments including gas-chromatographic separation and electron capture detection (GC-ECD) and as well as integrated cavity output laser spectrometry (ICOS).

Perhaps surprisingly, all eight datasets displayed regions of nitrous oxide undersaturations in contrasting hemispheres and seasons in the Atlantic Ocean. This was more pronounced in the recent datasets (AMT20, 22, 23 and 24), where most of the Atlantic Ocean acted as a small nitrous oxide sink. Seasonal variations between the two hemispheres were found, with autumn cooling producing surface waters more undersaturated for nitrous oxide in the northern than the southern hemisphere. The surface concentration of nitrous oxide between 10 to 30° S appeared to be influenced by increases in the atmospheric concentration of nitrous oxide, with the surface waters remaining at around saturation (98 to 102 %). Recommendations for future oceanic nitrous oxide observation programmes include interlaboratory calibration against reference standards and maintaining repeat transects for longer durations and along the same geographic coordinates.

Gas transfer at high wind speeds: extrapolating concurrent CO₂/DMS field measurements to SF₆

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Abstract

In several field studies (e.g. [1,2]), the transfer velocities of DMS and CO₂ were measured simultaneously using eddy covariance techniques. In most of these studies, a difference between DMS and CO₂ transfer was found, which was attributed to bubble mediated gas transfer. Several models predict the magnitude of this bubble mediated gas transfer [3,4]. However, most of them require too many tuning parameters, such that it is very difficult to extrapolate the gas transfer velocities measured for CO₂ and DMS to gases with much lower solubility such as SF₆ and He. A recently developed bubble model [4] only requires one tunable parameter, the transition solubility, which parameterizes the relative importance of the bubble surface area and the bubble volume for bubble mediated transfer, is used to estimate the transfer velocity of SF₆ from concurrent DMS and CO₂ measurements in the field. The required parameter, the transition solubility, was measured in a recent wind-wave tank study, where transfer velocities a total of 12 trace gases with solubilities between 0.005 and 15 were studied at wind speeds between 7 and 85m/s in two wind-wave tanks using fresh and salt water [4]. Using this method, SF₆ transfer velocities extrapolated from measured DMS and CO₂ transfer velocities under the assumption, that all differences found between DMS and CO₂ are due to bubbles, are found to be much higher than those measured by the dual-tracer method (e.g. [1,2]). Possible reasons and explanations for this discrepancy will be discussed.

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How to cross-link lab and field measurements

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Abstract

Field measurements showed to be useful for finding empirical gas transfer velocity - wind speed relations. However, identifying and quantifying the mechanisms of the small-scale mass exchange processes from field measurements alone remains challenging. This is due to many factors influencing the gas transfer velocity besides the wind speed, including buoyancy effects at low wind speeds, the state of the wind wave field (wave age, swell), rain, bubbles and spray and last but not least surface films. In addition, measurements both at very low wind speeds and high wind speeds are fundamentally difficult. Thus, fully resolving the mechanisms of air-sea gas exchange even with a significant effort towards more and larger field campaigns only is not expected to be successful in the near future.

In contrast, systematic studies isolating individual influencing factors are easy to perform in wind-wave tank facilities. However, the question remains how to simulate realistic oceanic conditions. In linear facilities measurements can be conducted at very low fetch conditions only, and even in annular facilities high wave ages are not possible because of the limited water depth. No lab facility is currently available to perform measurements with large oceanic gravity waves. Still, in recent years, major advances in understanding the small scale physical processes governing air-sea interactions have been made in wind-wave tank studies, e. g. the equivalency of heat and gas transfer (Nagel et al. 2015), the structure of the airflow above water waves (Buckley and Veron, 2016), the identification of the dominant mechanism of spray production at high wind speeds (Troitskaya, 2017), the enhancement of heat transfer at low fetch conditions (Kunz and Jähne, 2018) and bubble mediated gas transfer at extremely high wind speeds up to 85 m/s (Krall et al. 2019)

The fundamental question is what we can achieve by systematically combining knowledge from laboratory and field experiments. More specifically:

Which measuring conditions in laboratory facilities are most useful for field conditions?

What kind of field measurements could and should be performed to verify that lab results can be transferred to oceanic conditions?

How can lab studies help with development of measurement techniques to be used in the field and with the verification of currently used techniques?

Should a workshop be organized with this topic, e. g. within the SOLAS initiative? Who could contribute what?

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Sufficiently Realistic Simulation of Oceanic Conditions for Air-Sea Gas Exchange at the Re-Engineered Heidelberg Aeolotron

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Abstract

While many gas exchange measurements in wind-wave tanks have been made decades ago, almost none are performed nowadays. One reason is certainly the growing knowledge that with the available facilities oceanic conditions cannot be simulated adequately. Linear facilities offer only short-fetch wind conditions with very young wave fields, leaving a “fetch gap” to long fetch open ocean conditions. In an annular tank, such as the Heidelberg Air-Sea Interaction Facility, the Aeolotron [1], the wave field can come into equilibrium with the wind, because of the virtually unlimited interaction time. But because of the limited water depth (a problem of all lab facilities), the wave field has a limited phase speed and therefore high wave ages cannot be reached (“wave age gap”).

Here we propose a novel approach to air-sea gas transfer measurements in the lab, which allow to cover a much wider range of conditions than in any previous lab study, using the recently remodeled Aeolotron and novel measuring techniques. Now, sufficiently realistic simulation of oceanic conditions with respect to air-sea gas exchange are possible. These systematic measurements can lead to a prediction of oceanic gas exchange rates from lab measurements.

The new approach includes four key elements: (1) Measurements are performed under non-stationary wind conditions. Within a few seconds, it is possible to switch on the wind, so that measurements over the whole fetch range are possible. To this end, three techniques for fast measurements of the gas transfer velocity with a temporal resolution of only several seconds to minutes have been developed. (2) The wave age effect can be studied by changing the water depth in the facility from 15 to 90 cm. In this way the limiting phase speed (and thus the wave age) of the wind speed can be lowered by more than a factor of two. Using a heavier gas than air as an atmosphere in the Aeolotron, it is possible to reach even higher wave ages. (3) Experiments at different water temperatures will be performed to investigate if other effects than just the change in the Schmidt number influences the gas transfer velocity. (4) The walls of the Aeolotron have been covered by a Teflon layer so that more reliable measurements with different types of surfactants are possible.

In this presentation, first proof of concept measurements will be shown.

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Poster presentation

The impact of rain on ocean surface waves and currents

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Abstract

Precipitation is an important component of the interaction between earth's atmosphere and oceans, modifying air-sea fluxes of momentum, heat, and gas. It has been hypothesized that rain's suppression of ocean surface gravity waves and centimeter-scale wave enhancement should alter the nature of air-sea momentum flux, resulting in increased near-surface current. Here, we use field observations to describe this impact and measure the very near-surface current response to rainfall. During heavy rain, surface-roughening ring waves were generated and longer gravity waves were suppressed; immediately following, the magnitude of the near-surface current increased in response to wind forcing, but died as the rain subsided and long waves recovered. These first-of-their-kind field observations indicate that rain reduces ocean wave form drag in favor of tangential stress, resulting in the acceleration of current near the sea surface.

Oral presentation

Observations of mean and wave orbital flows in the upper centimeters of the ocean surface layer

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Abstract

The near-surface current response to wind forcing and wave-induced motions is a topic of fundamental interest within air-sea interaction. However, obtaining measurements of fluid velocity close to an undulating boundary presents a unique observational challenge. Here we describe first-of-their-kind thermal and polarimetric camera-based observations of wave orbital velocities and mean shear flows in the upper centimeters of the ocean surface layer, gathered aboard the "laboratory at sea" R/P FLIP. Measurements reveal a well defined logarithmic layer as seen in laboratory measurements and described by classical surface layer theory. Measured orbital velocity magnitudes are generally found to agree well with the prescriptions of linear wave theory, with departures at high levels of wind forcing attributed to the effects of microscale wave breaking. Measurements of wave characteristics and near-surface current during heavy rain indicate an acceleration of current near the sea surface. These phenomena are broadly important to the processes of upper-ocean mixing and global ocean-atmosphere interaction.

Oral presentation

CLAW: Dead or Alive?

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Abstract

A quarter of a century ago a paper was published that would have a profound influence on ocean biogeochemistry, atmospheric chemistry/physics and also possibly climate (Charlson, Lovelock, Andreae, Warren, aka CLAW; 1987, 'Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate,' *Nature*, 326: 655-661). In the paper the idea was proposed that the gas dimethylsulphide (DMS) emitted by marine phytoplankton could, after passage across the air-sea interface, form particles on oxidation in the atmosphere which could affect cloud albedo and climate.

When it was published the idea was novel and far reaching in its implications for atmospheric properties. But even more so for the suggestion that biological processes in the ocean could play an important role in controlling those properties. Because the idea was quite radical it led to much discussion as well as laboratory, field and modelling studies to assess its relevance and quantitative importance.

In this talk I will review the evidence both for and against and how the discussion has moved over time. Further I will discuss how the CLAW idea has affected how we view the Earth as a biogeochemical system and how it has also led to studies of the role of other volatile biogenic gases,, as well as DMS in the atmosphere.

Oral presentation

Working Towards Improved Gas Transfer Prediction by Understanding the Impact of Gustiness on Momentum Fluxes

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Abstract

Momentum transfer is important to wave growth and breaking, ocean currents and global circulation, and ocean mixing, all of which, to a greater or lesser extent, impact air-sea gas fluxes by altering conditions at the air-sea interface. Hence, a robust parameterization of the momentum flux is necessary to accurately predict air-sea gas exchange. However, while significant improvements have been made to momentum flux parameterization in recent decades, the impact of gustiness remains underexplored. Using direct flux measurements captured at sea, we investigate the influence of gustiness, i.e., rapid fluctuations in wind speed or direction, on the momentum flux. We will show that gustiness has a profound impact on the marine boundary layer by reducing air-to-sea momentum transfer. We will also explore the interaction between waves and gustiness which may have important implications for gas transfer prediction.

Near-surface Turbulence in Arctic, Temperate, and Tropical Inland Waters: Implications for Gas Fluxes

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Abstract

Near-surface turbulence is a key determinant of gas exchange velocities (k) used to compute fluxes of climate forcing trace gases under light to moderate winds. Scaling approaches to accurately predict turbulence would enable modeling of fluxes from diverse water bodies over large spatial scales. While wind based models have been used frequently, heating (buoyancy flux, $\beta +$) or cooling ($\beta -$) in the upper water column are likely to moderate turbulence relative to predictions from wind. Monin-Obukhov similarity theory (MOST) estimates turbulence, as rate of dissipation of turbulent kinetic energy (ϵ), taking into account the relative contributions of wind and β . With MOST, ϵ can be computed from time series meteorological and temperature data. The accuracy of MOST was evaluated in tropical floodplains and adjacent lakes and rivers, tropical reservoirs, temperate, boreal and Arctic lakes, and in Arctic ponds and rivers using measurements of ϵ from temperature-gradient microstructure profilers and acoustic Doppler velocimeters. Within sheltered flooded forests where cooling predominated and wind was negligible, measured dissipation rates were $10^{-8} \text{ m}^2 \text{ s}^{-3}$, similar to predictions from buoyancy flux under cooling based on MOST. With increased flow during falling water, dissipation rates were two orders of magnitude higher. This change effectively doubled predicted gas transfer velocities to 6 cm hr^{-1} . Diel cycles of stratification and mixing are accentuated in tropical water bodies with their intense heating during the day. In open waters under heating, winds up to 3.5 m s^{-1} , and waves which varied from ripples on the surface to 10 cm , maximum likelihood estimates of near-surface ϵ were independent of wind speed and high, $\sim 5 \times 10^{-6} \text{ m}^2 \text{ s}^{-3}$, one to three orders of magnitude higher than predictions from wind shear, and increased with heating. The accentuation of dissipation rates from $\beta +$ followed MOST with the accentuation higher than in atmospheric boundary layers as the mixing efficiency was low near the air-water interface. Thus, turbulence production was nearly balanced by dissipation. ϵ was ~ 10 times higher during heating than cooling. Gas transfer velocities for CO_2 at 20°C , $k \sim 600$, estimated using measured ϵ , were $\sim 10 \text{ cm hr}^{-1}$, validated with k obtained from chamber measurements, and 2.5 to 5 times higher than computed from wind-based models. Our predicted dissipation rates, from which k was computed, were validated with chamber measurements of CO_2 and CH_4 at these sites. Under cooling conditions with moderate winds, measured dissipation rates tended to be less than predictions from MOST, similar to our observations from a temperate lake. There, the discrepancy increased as wind speeds increased. We found similar patterns of enhanced dissipation rate under heating in the Arctic lake. Merging variable mixing efficiency with MOST will lead to improved time series estimates of k needed for modeling fluxes of dissolved gases over regional scales.

Oral presentation

Concurrent, open ocean eddy covariance flux measurements of dimethylsulfide and carbon dioxide: What have they taught us about gas transfer and what should we do next?

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Abstract

Direct flux observations using the eddy covariance (EC) technique can help us understand and quantify the global biogeochemical cycles of important elements, such as sulfur and carbon, as well as unravel the multiple physical forcings on gas transfer at the air-sea interface. By measuring compounds with different biogeochemical and physical properties simultaneously, we can gain insight into interfacial gas transfer, bubble-mediated gas transfer, and influence of efflux vs. influx on gas exchange, among other processes. Here I will present simultaneous DMS and CO₂ EC datasets measured to date and examine what they have taught us about gas exchange in the open ocean. Finally, I will present a research cruise we are planning to address these issues in the Labrador Sea with a focus on oxygen (Bubble mediated exchange in the Labrador Sea, BELS).

Oral presentation

Airborne observations over the North Atlantic Ocean reveal urea is a missing component of atmospheric reduced nitrogen

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Abstract

Despite the reduced nitrogen (N) cycle being central to global biogeochemistry, there are large uncertainties surrounding its sources and rate of cycling. Here, we present the first observations of gas-phase urea ($\text{CO}(\text{NH}_2)_2$) in the atmosphere from airborne high-resolution mass spectrometer measurements over the North Atlantic Ocean. We show that urea is ubiquitous in the marine lower troposphere and find that the ocean surface is the primary emission source. Urea is also frequently observed aloft due to long-range transport of biomass-burning plumes, and in instances where the air mass has been subjected to rapid frontal uplift. These observations alongside global model simulations point to urea being an important, and as yet unaccounted for, component of reduced-N to the remote marine environment. Since we show it readily partitions between gas and particle phases, airborne transfer of urea between nutrient rich and poor parts of the ocean can occur readily and could impact ecosystems and oceanic uptake of CO_2 , with potentially important atmospheric implications.

Rethinking Arctic Ocean CO₂ Fluxes

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Abstract

The paradigm that the Arctic Ocean is generally a sink for atmospheric CO₂ is based primarily on bulk flux calculations using underway shipboard data collected in late summer, when sub-surface chlorophyll maxima dramatically reduce pCO₂ at the base of the winter mixed layer. Regions of CO₂ outgassing in the Arctic are generally thought to be very localized and specific, e.g., near river mouths and in ice-edge associated upwelling events. However, extreme surface stratification coupled with variable bio-/geochemistry of sea-ice melt and river waters may result in actual fluxes that are very different from what's implied by waters drawn from 5-10 m below the surface. Our work in regions heavily influenced by river run-off and glacial melt, as well as sea-ice melt, has shown that in early spring (when sea-ice melt dominates the surface stratification) the ship-board underway system underestimates CO₂ drawdown, whereas in late summer (when river waters and warming control stratification), the underway system overestimates drawdown. We have also found that high pCO₂ from remineralized organic matter in river waters can extend over very large distances under the sea ice, across the entire Arctic; high-pCO₂ waters observed in a main outlet from the central Arctic Basin into the eastern Canadian Arctic carried a strong tracer signal from Russian rivers. Our observations of the large temporal, vertical, and horizontal variability in surface pCO₂ in the Arctic Ocean emphasize the importance of developing robust methods for directly measuring, and more effective model parameterizations of, air-sea CO₂ fluxes in polar waters.

Oral presentation

Measurements of surface-cooling induced gas-transfer using fluorescence-lifetime imaging (FLI) technique

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Abstract

Previous studies have shown the significant role of buoyant-convective instability in enhancing the transfer rate of heat and greenhouse gases across the air-water interface. To study the dynamics of such transport processes without disturbing the thin dissolved gas concentration boundary layer, we employed a fluorescence-lifetime imaging (FLI) technique to measure the oxygen concentration distribution. Platinum octaethylphorphyrin (PtOEP) embedded in nanoparticles were introduced into the water as tracer. The measurement area was illuminated by a laser light sheet with a wavelength of 405 nm. The experiments were performed in a 30 cm × 30 cm × 43 cm tank filled with warm (25°C) deionized-water to a depth of 40 cm. Before each experiment, oxygen was purged out of the water. The buoyant convective instability was promoted by exposing the surface of warm water to the cool ambient air. The temperature difference between the air measured at 30 cm above the water surface and the water in the bulk was varied from 5 to 20 °C. The relative humidity in all experiments was about 70-80%.

Sequences of oxygen concentration images showed that shortly after the water surface was exposed to the cool air, at some locations the concentration boundary layer thickened forming relatively large plumes. While sinking, these cool and oxygen-rich plumes deformed into anchor-like structures. The penetration depth of these structures often exceeded the field of view (> 5cm), showing the effective transport mechanism of oxygen-rich fluid from the top into the oxygen-poor bulk region. It was observed that the plumes appeared more frequently and progressed faster with increasing temperature difference, which is positively correlated with the transfer rate. In addition, the setup allowed optical access from the top enabling the coupling of the FLI with an infrared-thermal imaging technique. Simultaneous measurements of both techniques showed the correlation between the development of convection cells at the water surface and the movement of sinking oxygen-rich plumes.

Constraining the role of the surface micro layer in tropical riverine headwaters of Amazonia

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Abstract

The interface between the world's atmosphere and hydrosphere, known as the surface microlayer (SML) exhibits many unique properties in comparison to subsurface water (SSW) (Wu et al., 2017). It is subjected to greater environmental and climatic variation than the underlying water column (Hardy, 1982) and is known to be enriched, to varying degrees, in many chemical compounds (Elzerman and Armstrong, 1979; Pellenburg and Church, 1979; Wurl et al., 2011). The SML is a fundamental control of climate active gas cycling through regulation of the gas transfer velocity (k_w). While organic matter (OM) in the SML of marine environments has received attention demonstrating up to a 50% reduction in k_w (Pereira et al., 2016; 2018), the role of the SML of inland waters is poorly constrained (Raymond et al., 2013; Regnier et al., 2013).

Inland waters are terrestrial integrators, regulators of climate change (Kokic et al., 2015), and a net source of over 3.9 Pg C year⁻¹ in the form of CO₂, of which 1.8 Pg C year⁻¹ are emitted from streams and rivers (Marcé et al., 2018; Raymond et al., 2013). Headwaters comprise over 70% of worldwide river networks (Gomi et al., 2002); however, the sparsity of data in temporal regions causes a large uncertainty in outgassing estimates (Drake et al., 2018). Where estimates exist, they typically rely on proxies to infer gas concentration (pH and alkalinity) and k_w (river gradient) (Raymond et al., 2013). These estimates are likely compromised by significant secondary factors such as bubbles and OM enrichments in the SML with temporal and spatial variability that will cause variable k_w . To address this, we combine high-end analytical dissolved OM (DOM) analysis with home-built CO₂ samplers specifically designed to estimate k_w during variable SML OM compositions.

Our results from Amazonian headwaters of the Essequibo River in 2019 show that while dissolved organic carbon is not enriched in the SML there are compositional changes in OM. Specific UV absorbance at wavelength 254nm (SUVA₂₅₄) enrichments increase over time indicating an increase in aromatic moieties. This is confirmed by a pioneering liquid chromatography (LC) technique (Huber et al., 2011) which demonstrates increasing aromaticity in the humic fraction of OM. Initial observations suggest that increasing aromaticity likely results in fluctuations of k_w with flux estimates impacted by variable SML OM composition changes over time.

The CO₂ fluxes at the ocean-atmosphere interface on the Brazilian continental shelf: a review of its behavior as a source or sink of atmospheric CO₂

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Abstract

Most studies on the global carbon cycle have neglected the Southern Hemisphere oceans, due to the limitations of spatiotemporal sampling, especially in continental shelves and coastal zones. Although Brazil's continental shelf has continental size, it remains poorly studied in terms of the carbonate system and the CO₂ fluxes between ocean and atmosphere, even in national level. In this study, we prepared a literature review on papers about the ocean-atmosphere CO₂ fluxes along the Brazilian shelf, including nearshore areas, to characterize the current knowledge about the carbonate system, and to assess the Brazilian coastal zones behavior as a source or sink of CO₂. We found around 110 articles, published between the years 2000 and 2021, at the Web of Science and Scopus repositories. For methodological purposes, the Brazilian shelf was divided into the regions South-Southeast (around -34°S to -18°S), North-Northeast (around -18°S to 5°N), and we further considered the whole Brazilian continental shelf in the global context. The large-scale studies show that the continental shelf presents latitudinal range of CO₂ fluxes, where the North region acts as a source of CO₂ to the atmosphere, the Southeast shows the neutral condition, and the South region acts as a sink of CO₂. For instance, the mean CO₂ fluxes in these large areas during the boreal autumn between 2000 and 2008 were, respectively, 1.6 ± 0.6 mol CO₂ m⁻² year⁻¹ (between 1°N–15°S), 0.5 ± 0.7 mol CO₂ m⁻² year⁻¹ (between 15°S–31°S) and -3.2 ± 2.7 mol CO₂ m⁻² year⁻¹ (between 31°S–40°S) (Padin et al., 2010). However, locally, the CO₂ fluxes are widely influenced by seasonality, mesoscale, and synoptic oceanographic and meteorological processes, also by the large plumes from Da Plata and Amazonas rivers, hence the same area can be a source or sink of CO₂ depending on these different conditions. We highlight the lack of regional information especially between Bahia and Ceara states, and the absence of a consistent temporal data series product for the entire continental shelf. Finally, this study emphasizes the need for long-term observations and monitoring

initiatives alongshore to better understand the Brazilian continental shelf role on the global carbon budget.

Oral presentation

Statistical distributions of whitecap variables using a novel remote sensing technique to detect and track individual whitecaps in digital sea surface images

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Abstract

Quantification of air-sea bubble-mediated gas exchange can be enhanced through a better understanding of whitecap air entrainment rates, bubble size distribution and bubble residence times. Sophisticated digital image-based remote sensing techniques of the sea surface are an invaluable tool in providing better descriptions of wave breaking activity on a whitecap-by-whitecap basis. Further, the use of these techniques allows for improved knowledge on the variability of populations of individual whitecaps across a multitude of sea states, of which little is known.

This work uses a stereographic video system mounted on the Aqua Alta oceanographic Tower (AAT) in the Adriatic Sea east of the Venice Lagoon to gather digital images of the sea surface. Using these images, we have developed a novel remote sensing technique that detects and tracks individual whitecaps. To date, data have been acquired in wind speeds ranging from 6 to 16 m/s. The whitecaps are first detected in digital grayscale images with a brightness thresholding technique that utilizes the image pixel intensity histogram. The movement of individual whitecaps is estimated with optical flow and is used to track whitecaps between consecutive frames. After tracking the whitecap throughout its life cycle, various geometric, kinematic and dynamic properties such as the time-evolving foam area [m²], breaking speed [m/s], average crest length [m] and foam area growth and decay timescales [s] are extracted from the corresponding frames of the 3D reconstructed sea surface. These extracted quantities are subsequently aggregated and presented as statistical distributions of whitecap variables such as the maximum area, growth timescales and decay timescales.

Distributions of these whitecap variables will be presented and discussed in the context of current understanding of wave breaking activity at the sea surface and relevance to bubble-mediated gas transfer.

Breathing Oceans: understanding the role of surface-active organic matter composition in the ocean skin layer to modulate gas exchange between the atmosphere and ocean

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Abstract

Oceans are a global reservoir of greenhouse gases estimated to account for 20–40% of the post-industrial sink for anthropogenic carbon dioxide (CO₂). However, quantifying the exchange of key greenhouse gases such as CO₂, methane (CH₄), and nitrous oxide (N₂O) across the air-water interface of the ocean is a major challenge.

The oceanic uptake of greenhouse gases is determined by relative differences in gas concentrations of water and air and its transfer velocity (k_w), which is controlled by variability of near surface turbulence in the sea surface microlayer (SML). The SML (depth < 400 μ m) is a physically and biogeochemically distinct ocean-atmosphere interface covering the entire ocean surface, containing enrichments in surface-active organic matter (surfactants). Gas exchange suppression by surfactants in the SML, has been shown to reduce the amount of CO₂ annually stored by ~9% in the Atlantic Ocean (Pereira et al., 2018). Wind speed is a fundamental control of near surface turbulence but known to be a weak k_w predictor with other key controlling variables. These uncertainties impede our ability to model and predict the role of the oceans in modulating climate.

Surfactants are derived from multiple organic matter (OM) sources along the land-ocean continuum include in-situ primary production, allochthonous inputs of terrestrial material of either natural or anthropogenic origin, and the photochemical and/or microbial reworking of higher molecular weight material. However, the role of surfactants in the SML is obscured by our current (in)ability to characterise OM. Pereira et al. (2016) demonstrated differences in k_w suppression along a terrestrial-marine gradient that is concomitant with a changing OM composition in the North Sea and that OM source and associated composition may have a large control on the supply of surfactants to the SML, which in turn reduces k_w . Pereira et al. (2018) further demonstrated up to 50% k_w suppression by surfactants was related to sea surface temperature in 'hotspots' of the Atlantic Ocean.

Here we outline a new 5-year European Research Council project (BOOGIE) that builds on our previous work and aims to further understand how the dynamic OM composition in the SML impacts gas exchange over space and through time to improve estimates of oceanic sinks and sources of key greenhouse gases.

Air-sea exchange of acetaldehyde, acetone and DMS at a UK coastal site.

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Abstract

Volatile organic compounds (VOC) are important for atmospheric chemistry because they influence the oxidative capacity of the atmosphere by acting as hydroxyl radical sinks. Their subsequent lower-volatility oxidation products can also condense to form particulates leading to cloud condensation nuclei. Large discrepancy remains in the role of the ocean towards the global VOC budget due to incorrectly understood, missing or poorly scaled sources and sinks.

We present air–sea fluxes of acetaldehyde, acetone and dimethylsulfide, quantified simultaneously by eddy covariance using a proton transfer reaction quadrupole mass spectrometer, at a coastal observatory in the south-west UK during the spring phytoplankton bloom (Apr–May 2018). Comparisons are made between an open-ocean (North Atlantic Ocean) and urban-dominated (Plymouth Sound) wind sector. We demonstrate the quadrupole mass spectrometer can be used to resolve eddy-covariance fluxes of acetaldehyde, acetone and dimethylsulfide in a highly variable coastal environment. On the other hand, we show isoprene is below our limit of detection using two separate analyte masses and cannot be resolved in this setting.

Mean (± 1 standard error) fluxes of acetaldehyde, acetone and dimethylsulfide from the open-ocean wind sector were 1.21 ± 0.69 , 7.94 ± 0.50 and 2.53 ± 0.27 $\mu\text{mol m}^{-2} \text{d}^{-1}$ respectively (+ sign indicates sea-to-air). Good agreement was shown with other Atlantic Ocean flux studies at the same latitude. In comparison, the urban-dominated wind sector showed fluxes of -5.22 ± 1.14 , -12.66 ± 1.32 and 1.19 ± 0.39 $\mu\text{mol m}^{-2} \text{d}^{-1}$. The greater influxes of acetaldehyde and acetone and the lower efflux of DMS from the Plymouth Sound wind sector were likely driven by higher atmospheric concentrations from the urban environment and reduced wind speed respectively.

Further comparisons are made to fluxes calculated from the two-layer model, using atmospheric and seawater concentrations from the observatory and off-shore L4 marine station respectively. The model results supported our eddy covariance measurements with agreement in expected surface water saturation and flux direction. Generally, modelled values predicted an average flux $\approx 25\%$ lower than the average direct measurements.

Shipboard Infrared and Visible Remote Sensing of Whitecaps

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Abstract

Under influence from the wind, waves grow until they become unstable and break. Breaking entrains air, creating plumes of bubbles below the surface. Smaller bubbles dissolve into the water, larger bubbles rise and form whitecaps. Whitecaps can be distinguished as either actively generated (stage A), or decaying (stage B). Stage A marks an acoustic period with turbulence, energy dissipation, ocean mixing, increased surface roughness, and bubble-driven gas diffusion. Buoyant bubbles drag water upward bringing surface active material and creating regions of divergence which enhance air-sea gas transfer. Stage A generation also enhances spray through the tearing of wave crests which significantly enhance sensible and latent heat fluxes. At stage B the bursting of bubbles produces small droplets which reside in the air long enough to reach moisture equilibrium and transform into sea salt aerosols which through several pathways impact the climate. Whitecap coverage is used expansively to estimate these processes yet the routinely employed wind speed dependence has an order of magnitude uncertainty that, in large part, is due to significant variations in stage B lifetime. Here we present initial results from two research cruises in the Gulf of Mexico to understanding the factors which influence stage B lifetime and whitecap fraction. Whitecaps were observed using visible and infrared remote sensing which provides clear, unambiguous separation of whitecap stages. Concurrent measurements were made of mean atmospheric and oceanic variables as well as momentum and enthalpy fluxes, surface tension, and subsurface bubble plume characteristics and supplemented with wave information taken from existing buoys networks.

Oral presentation

A Field Experiment to Determine the Impact of Nearshore Processes on Air-Sea Mass, Momentum, and Heat Fluxes

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Abstract

The accuracy of weather and climate models depends on reliable quantification of air-sea energy transfer. While decades of research and technological advances have led to significant improvements to momentum, mass, and heat flux parameterization, even the most robust models are developed from open-ocean measurements where conditions are spatially uniform and similarity theory generally applies. Nearshore, wave shoaling and breaking, varying wind-swell incidence angles, complex currents patterns, rapid bathymetric changes, and shore-side topographic features all contrast open-ocean homogeneity meaning flux parameterizations are less effective. Therefore, there is a critical need to identify and systematically quantify the impact of coastal processes and features on air-sea exchange over a wide range of wind speeds. The US SOLAS Science Plan (2021) lists Conducting coastal gas exchange experiments to test existing parameterizations and develop new parameterizations in various shallow water environments as an important priority. To this end, we deployed flux packages on the pier and the beach tower at the Army Corp Field Research Facility (FRF) in Duck, North Carolina, as part of DUNEX (DUring Nearshore EXperiment). During the months-long campaign we recorded momentum, heat, H₂O, and CO₂ fluxes in the nearshore, which are complimented by FRF's extensive metocean observational network. We will present an overview of the experiment and selected results.

Oral presentation

Wind and fetch dependent gas transfer velocity in an Arctic sea-ice lead determined from eddy covariance CO₂ flux measurements

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Abstract

Gas transfer velocity of poorly soluble trace gases is driven by near-surface ocean turbulence, which may be enhanced or suppressed by the presence of sea ice. There are few measurements of gas transfer from sea-ice regions, increasing uncertainty in the magnitude of the polar ocean CO₂ sink.

Here, gas transfer velocity is directly determined from eddy covariance CO₂ flux measurements at a sea-ice lead in the Arctic Ocean close to the North Pole during the transition between summer-melt and autumn-freeze-up seasons. The flux measurements were made with relatively high precision due to low humidity flux and high $\Delta p\text{CO}_2$ conditions. Measurements were made of both lead water-atmosphere and ice/snow-atmosphere fluxes. The ice/snow-atmosphere fluxes are the first direct flux measurements reported for pack sea ice in the summer-autumn season. A flux footprint analysis was used to determine the flux into the lead surface from which the gas transfer velocity was derived.

Gas transfer velocities were determined for wind speeds up to 13.1 m s⁻¹. It is shown that the wind-speed dependent gas transfer rate in the lead is approximately quadratic and suppressed by 25% relative to commonly used open-ocean parameterisations. The gas transfer measurements exhibit a dependence on both wind speed and on fetch, demonstrating the importance of lead dimensions and the resulting lead wave characteristics on gas exchange. Conversely, the measurements exhibit little dependence on convection-driven turbulence resulting from buoyancy flux, suggesting this is not a significant driver of gas exchange for this location and season. These results show that current estimates of polar ocean carbon uptake, using open-ocean gas transfer parameterisations scaled linearly by open-water fraction, likely overestimate gas exchange rates in summertime.

The role of sea ice in CH₄ and CO₂ air-sea gas transfer in the central Arctic Ocean

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Abstract

The role of sea ice in suppressing or enhancing the air-sea exchange of trace gases is poorly known. Sea ice may enhance near-surface turbulence through ice-edge form drag and shear from ice movement. At the same time sea ice suppresses surface waves through fetch limitation. In addition, sea ice itself plays a complex and important role in gas cycling, with for example brine channels acting as an additional pathway for gases to reach the atmosphere, brine release during freeze up inducing haline convection, potentially enhancing gas transfer, and freshwater release from melting sea ice acting to stratify surface waters, suppressing gas transfer. Here we present recent results from several central Arctic Ocean expeditions onboard the Swedish icebreaker Oden.

In summer 2021 during the Synoptic Arctic Survey 2021 (SAS2021) expedition, a floating chamber system built on a Los Gatos Research (LGR) Cavity Enhanced Laser Spectrometer was used to measure air-sea fluxes of CH₄ and CO₂. To determine gas transfer velocities, flux measurements were combined with atmospheric concentration measurements from an LGR spectrometer-based profiling system on Oden's foremast, and water concentration measurements from GC and membrane-based systems sampling from Oden's CTD, underway line (8 m depth), and near-surface profiles from a Ruttner sampler deployed from the sea ice. Fluxes were measured from leads and melt ponds in pack ice and the marginal ice zone, accessed both from sea ice and from Oden.

Additional measurements of surface-atmosphere CH₄ and CO₂ exchange were made using an eddy covariance (EC) system onboard Oden, also based on an LGR spectrometer, mounted at the ship's foremast and with winds corrected for both platform motion and flow distortion using a CFD model of the airflow over Oden. This EC system was also deployed on previous summertime central Arctic Ocean expeditions in 2014, 2016 and in 2018. In 2018 during a five-week ice camp, an additional EC system was also deployed on sea ice adjacent to an open lead system.

Initial analysis from SAS2021 indicates that CH₄ formed through biological processes under ice can be released to the atmosphere when sufficient mixing mechanisms (i.e. wind and ice drift) are present. The role of sea ice in gas transfer is further examined using results from earlier expeditions, demonstrating reduced gas transfer rates for CO₂ in a sea ice lead, and determining a relationship between gas transfer and sea-ice concentration.

Oral presentation

pCO₂ gradient in the near surface ocean

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Abstract

Around half of all carbon dioxide (CO₂) produced by humans since the Industrial Revolution has dissolved into the ocean. Therefore, understanding how the ocean exchanges CO₂ with the atmosphere is critical for the prediction of climate change. The global assessment of the air–sea CO₂ exchange is based on atmospheric and oceanic measurements. For the latter, ship data are generally collected at an approximate water depth of 3–5 m, e.g., the inlet depth of ship-based pipelines or the CTD depth closest to the water surface. The rationale behind this is the assumption that no gas gradient exists in the upper 0–5 m of the surface layer (Figure 1). However, ignoring this surface layer, including the sea surface microlayer (SML) directly at the air–water interface, can cause biases of 20–50% in estimating the exchange rate of CO₂ (Broecker et al., 1978; Salter et al., 2011; Pereira et al., 2018; Mustafa et al., under revision). This may lead to strong uncertainties for global and regional CO₂ flux calculations.

Calleja et al. (2013) reported for the first time CO₂ and O₂ gradients in the upper 5 m of the surface layer. They concluded that temperature differences accounted for only 11% of the observed partial pressure of CO₂ (pCO₂) gradients. Other processes might generate disequilibria in the CO₂ between the ocean and the atmosphere, leading to a gradient-driven CO₂ exchange. However, the upper surface layer (0–5 m) is poorly characterized in terms of biogeochemistry. The overall objective is to elucidate the importance of the surface layer in the air–sea exchange of climate-relevant gases by comparing air–sea CO₂ fluxes using observed gradients and the assumption that no gradients exist. We propose high-resolution measurements, both in space and time, using a state-of-the-art research catamaran and an autonomous buoy to investigate the short-term variability in the direction and slopes of gradients and air–sea CO₂ fluxes. Fieldwork have been conducted during a research cruise on the RV Falkor in November-December 2019.

Surfactant control on air-water gas exchange in freshwater lakes

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Abstract

Suppression of the gas transfer velocity (k_w) of CO_2 by natural biological surfactants at the ocean basin scale is well established but its importance in freshwater systems remains open to question. This is a major issue given the likely freshwater contribution to the global cycles of CO_2 and other trace gases. For example, it is estimated that the total freshwater emission of gaseous carbon (CO_2 and CH_4) might balance its net uptake by the combined marine and terrestrial biospheres. Large variations in local and continental scale emissions are indicated. Small high productivity lakes may play a disproportionality large role, and rapid lake warming in climate sensitive regions is likely to cause accelerated carbon mobilisation. However, our ability to accurately quantify this is seriously limited by a scarcity of data and large measurement uncertainties.

To address this, in an ongoing three-year project funded by the UK Leverhulme Trust we are making the first assessment of seasonal k_w control by natural surfactant in a freshwater lake. Our study site is Uppsala University's permanent research station at Lake Erken (Sweden), established in 1946. The site is fully instrumented: a suite of biogeochemical variables is routinely monitored in situ and at high frequency. During 2021-2022 we have quantified total surfactant activity (SA) seasonally on-site, in the microlayer (Garrett screen sampler: uppermost $\sim 400 \mu\text{m}$) and in subsurface water, with the addition of dissolved organic matter composition analysis. k_w for CO_2 is derived concurrently from Eddy Covariance fluxes measured at a permanently stationed flux tower on the lake ($\sim 6 \text{ km}$ fetch), and CO_2 concentrations in surface water. An ADCP mounted in the flux footprint of the tower allows for studies of SA on the surface water turbulence.

Our results should be of value to future estimates of the freshwater-atmosphere exchange rates of CO_2 and other climate-relevant trace gases.

Should we account for the skin temperature effect in model simulations?

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Abstract

Watson et al. (2020) showed that accounting for the cool skin temperature effect in observations increases the ocean's CO₂ uptake by 0.8-0.9 PgC/yr. We investigate if this effect should be included in ocean models as well. We use two simulations from 1980 to 2017 with an ocean model consisting of NEMO (ocean physics), MEDUSA (marine biogeochemistry) and CICE (sea ice), which is forced with the ERA-Interim atmospheric reanalysis. Physical and biogeochemical fields are initialised from reanalysis or climatology in 1980. The first simulation uses the model's top layer temperature for the CO₂ flux calculation (hereafter BASE). The second simulation instead uses the skin temperature, which is derived from the upper ocean temperature at each time step (hereafter SKIN). This modification only impacts the biogeochemistry simulation because NEMO and MEDUSA are coupled one-way. Initially, the ocean CO₂ uptake in SKIN is about 15% larger than in BASE in response to the skin SST effect, which increases the CO₂ solubility. This difference decreases to less than 3% within the simulation period. The cause is a build-up of DIC in the surface ocean, reducing the chemical gradient across the air-sea interface and counteracting the skin temperature effect. DIC builds up because the skin temperature effect induces additional CO₂ entering at the surface but does not increase the export to the deep ocean at the same rate. The DIC build-up probably also causes an enhanced seasonal amplitude of the CO₂ flux in SKIN compared to BASE, by reducing the ocean's capacity to buffer pCO₂ changes. These results suggest that the skin temperature effect should be included in models since it affects the CO₂ flux variability, in particular on seasonal timescales. However, to improve the representation of long-term CO₂ uptake and storage it is more important to accurately simulate processes driving the export of carbon to the deep ocean.

Oral presentation

Gas transfer velocities for greenhouse gases (CO₂, CH₄, N₂O) along the river-estuary continuum

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Abstract

The exchange of greenhouse gases, such as carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O) between the water and atmosphere is of great scientific interest because of its importance in global biogeochemical cycling. However, the physical transfer of gases across the water-atmosphere interface driven by water-side turbulences is system-specific in aquatic ecosystems and to date remains the largest uncertainty in the flux computation. Furthermore, gas transfer velocities (*k*) of CO₂, CH₄ and N₂O are usually estimated from empirical models based on *k*CO₂, however, the three gases can behave quite differently at the water boundary layer.

In this study, we compare CO₂, CH₄ and N₂O gas transfer velocities along the river-estuary continuum, to evaluate the accuracy of using uniform *k* values for estimating water-atmosphere fluxes of CO₂, CH₄, and N₂O, and to identify when an 'estuary' becomes a 'river' for the purposes of evaluating greenhouse gas fluxes.

The *k* values of CO₂, CH₄ and N₂O were estimated from 77 individual 'flying' chamber deployments in the Maroochy river-estuary in Australia in June 2019. Incubations were conducted during spatial surveys across and along the channel, and four time series in different sections from the estuary mouth to the upstream tidal river. Incubations were undertaken over large gradients of water current velocity, wind speed, water depth, temperature and weather conditions. We aim to develop gas-specific *k* parameterizations for the three greenhouse gases based on physical parameters in the Maroochy river and estuary than can be applied in other river-estuaries in the world.

Using land-based stations for air-sea interaction studies, issues with land influence and non-stationarity

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Abstract

In-situ measurements representing the marine atmosphere are taken at ships, buoys or stationary moorings, or on land-based towers. By using fixed towers motion correction can be avoided and measurements can be taken over extended periods of time. One needs to make sure the measurements represents the sea area and evaluate the land influence at different scales on the fluxes, in addition there are indications that non-stationarity of the wind field over the sea significantly disrupts the equilibrium between the wind, stress, and wave fields, which potentially can alter the surface drag as well as heat and scalar fluxes.

Measured gas fluxes and turbulence properties from the land-based marine ICOS station Östergarnsholm have shown to well represent open sea marine conditions for specific wind direction intervals. Data from other sectors are usually discarded as they are disturbed by coastal zone. Data is defined according to the following categories:

- 1) Marine data representing open sea
- 2) Disturbed wave field resulting in physical properties different from open sea conditions and heterogeneity of water properties in the foot-print of the flux tower.
- 3) Mixed land/sea footprint of the tower, very heterogeneous conditions and a very active carbon production/consumption.

There are differences between the data for the different categories, and coastal processes influences carbon and heat fluxes (Rutgersson et al., 2020). Limited fetch conditions have an impact on the surface stress and the impact of non-stationarity on the stress and drag coefficient becomes important for wind speeds less than about 6 m s⁻¹ (Mahrt et al., 2020) even for open sea conditions.

Mahrt, L., E. Nilsson, H. Petersson and A. Rutgersson (2020) Sea-surface stress driven by small-scale non-stationary winds. In revision.

Rutgersson, A., Heidi Pettersson, Erik Nilsson, Hans Bergström, Marcus B.E. Wallin, E. Douglas Nilsson, Erik Sahlée, Lichuan E. Wu & E. Monica Mårtensson (2020) Using land-based stations for air-sea interaction studies, *Tellus A: Dynamic Meteorology and Oceanography*, 72:1, 1-23, DOI: 10.1080/16000870.2019.1697601

Bubble break-up and the formation of sub-Hinze scale bubbles in turbulence

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Abstract

The exchange of gases between the ocean and the atmosphere is mediated by dissolution through the interfaces of bubbles which are entrained by breaking waves. Turbulence in the water induced by wave breaking breaks these bubbles apart, affecting their lifetimes underwater and the amount of surface area available for gas exchange.

Laboratory and numerical experiments have described the size distribution of bubbles under a breaking wave. For bubbles above a critical length scale, the Hinze scale of about 1.5 mm, the size distribution follows a turbulent cascade scaling $N(r) \sim r^{-10/3}$, while the distribution below the Hinze scale remains an open question, with large scatter in reported data sets and little theoretical understanding. These small bubbles are particularly important in the transfer of low solubility gases such as N₂ and O₂.

To understand the formation of sub-Hinze scale bubbles and better understand the physics driving the breakup, we study bubble break-up in an idealized turbulent flow created in the laboratory by the convergence of turbulent water jets. A controlled distribution of bubble sizes is injected into a quiescent region through a needle, and the bubbles are left to rise into the turbulence. Tracking the bubbles in three dimensions with an array of high-speed cameras, an ensemble of breakup events is accumulated, each event consisting of the sizes and trajectories of the parent and its children.

The turbulence is characterized by planar particle image velocimetry performed in a set of planes spanning the turbulent region, which allows the bubbles' trajectories to be used to quantify the turbulence they encounter before and during break-up. Sweeping the sizes of the bubbles injected and the intensity of the turbulence imposed, we find a broad distribution of child bubble sizes created over a range of breakup Weber number. Finally, we apply the measured breakup rates and child size distributions to the prediction of bubble size distributions in an ensemble of bubbles broken apart by turbulence, making comparisons to experimental data.

Testing and application of a diffusion-based method for sampling DMS in the Sea Surface Microlayer

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Abstract

Sampling of trace gases in the sea surface microlayer (SML) is a major challenge as existing sampling methods are not adapted for dissolved gas sample collection and may lead to underestimation of gas concentration. In this study, a method that uses gas-permeable tubing to sample dimethyl sulfide (DMS) in the SML was developed. A silicon tube (diameter 2.41 mm) is filled with Milli-Q[®] water and left in contact with seawater, during which the DMS diffuses into the tube across the concentration gradient. The gas-permeable tubing approach was deployed in semi-controlled conditions using coastal water to determine reproducibility, accuracy and diffusion efficiency. For a 10-minute deployment, DMS concentration in the gas-permeable tube was 61% (10% S.D) of the external seawater concentration, with a reproducibility of 13% (\pm 9% S.D, n=9). DMS diffusion efficiency was influenced by water temperature, and consequently it is necessary to calibrate the DMS diffusion efficiency under the respective sampling conditions.

The performance of the gas-permeable tube method was compared to that of the plate and the screen, during a time-series study over three months of the SML at 3 stations with differing degrees of coastal and open water influence around Wellington, New Zealand. DMS concentrations were consistently higher with the gas-permeable tubing relative to both the plate and screen, at 41% and 21% respectively, reflecting that DMS loss to the atmosphere is minimized with the tube. The reproducibility and accuracy, combined with the higher concentrations compared to other techniques, confirms the potential of this novel technique for trace gas measurement in the SML.

During the time-series, biological, biogeochemical and physical properties of the SML and subsurface water were also determined to explain the variability in dimethylsulfoniopropionate (DMSP) and DMS. DMSP was significantly enriched in the SML in most sampling events, with an enrichment factor ranging from 0.69 to 1.69. DMS enrichment in the SML was 1.18 (0.63 - 1.80) at the shallow littoral site and decreased to 1.06 (0.63 - 1.50) at the offshore site. Overall, there were no temporal trends or coastal-offshore gradient in DMS or related biogeochemical parameters in the SML. However, DMS concentration, and also DMS to DMSP ratio, were significantly correlated with solar radiation, indicating a role for light as a primary determinant of DMSP and DMS in the SML.

Evidence that differences between the dominant drivers of surface air-sea exchange and those of surface cross-shelf transport are controlling continental shelf-sea carbon sinks

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Abstract

The highly heterogeneous and biologically active continental shelf-seas are important components of the oceanic carbon cycle. Recent work has identified that these seas are now acting as increasing global sinks of carbon dioxide, but variations exist both within the same seas and across different shelf systems (Laruelle et al., 2018). Imbalances between surface air-sea exchange of carbon dioxide (CO_2) and carbon export at depth is one proposed explanation for the evolution of these sinks. Recent findings show that the air-sea exchange of CO_2 can be the dominating term in a shelf-sea carbon budget and that wintertime conditions control the strength of the sink (Kitidis et al., 2019). Here two 21 year re-analysis datasets are used to identify that geostrophic, wind, and wave driven currents are all important for the transport of water onto many of these shelf seas at the surface, which in turn drives the off-shelf flow of carbon rich water at depth. The importance of each current component appears to vary within seas, across seasons and different shelf systems. The same datasets are used to characterise the wind driven air-sea gas exchange within these shelf seas. The wintertime cross-shelf transport and gas exchange are then placed into context for fourteen continental shelf-seas that are exhibiting differing rates of change in surface water partial pressure of CO_2 , $p\text{CO}_2$. Generally, shelf-seas with high rates of change in $p\text{CO}_2$ are experiencing medium to high air-sea exchange, but weak to medium cross-shelf surface transport, consistent with a bottleneck in the offshore transport of carbon. Whereas, shelf-seas with low rates of change in $p\text{CO}_2$ are experiencing medium to high air-sea exchange and weak to high cross-shelf surface transport, consistent with no bottleneck. Collectively this work supports the hypothesis that imbalances between air-sea exchange and cross-shelf transport, caused by differences in the dominant process driving surface transport or exchange, are likely determining the change in shelf-sea CO_2 sinks. If true, future changes in large-scale geostrophic ocean currents, and wind and wave climate combined with atmospheric CO_2 concentrations will control the strength of the continental shelf-sea sinks and their acidification rates.

References

Kitidis et al. (2019) Winter weather controls net influx of atmospheric CO₂ on the north-west European shelf, Scientific Reports, doi: 10.1038/s41598-019-56363-5

Laruelle et al., (2018) Continental shelves as a variable but increasing global sink for atmospheric carbon dioxide, Nature Communications, doi:10.1038/s41467-017-02738-z

Reassessing the 4.33% / °C constant used for temperature partitioning of the inorganic carbonate system, how do derived constants impact the air-sea CO₂ flux?

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Abstract

Temperature effects the partitioning of the three inorganic carbonate species that constitute the dissolved inorganic carbon pool of the ocean. For accurate surface seawater air-sea CO₂ flux calculations, it is necessary to correct the CO₂ measurement for temperature differences between the in situ sampling depth, the seawater equilibrator and the actual air-sea interface where exchange occurs. To make these corrections the CO₂ measurements are often corrected twice using a constant of 4.33% / °C, firstly from the equilibrator temperature back to the in situ sampling depth temperature and secondly from the sampling depth temperature to the surface interface temperature. This widely used correction was derived during a single cruise in the North Atlantic Ocean and does not account for regional changes in carbonate chemistry or nonlinearities in the carbonate system, yet is used for calculating global fluxes of CO₂. Previous analysis has shown that in the ocean where the temperature can range from 0–30 °C, the change in pCO₂ with temperature can range from 3.7 to 5.3 % / °C. We use the SOCAT and GLODAP databases with the CO₂SYS carbonate package and the FluxEngine toolbox to derive new temperature constants and to then recalculate global air-sea flux of CO₂. We compare this newly computed air-sea flux with the equivalent flux estimate using the fixed correction of 4.33% / °C. Spatial differences in the flux and their implications on regional and global scales will be presented and discussed. We will comment on the limitations of using GLODAP data as the input data for these calculations and the large effect that the choice of carbonate dissociation constants can have on determining the derived temperature corrections.

Poster presentation

Direct flux measurements of carbon dioxide and methane in the Canadian Archipelago in variable sea ice conditions

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Abstract

Remoteness and tough conditions have made the Arctic Ocean historically difficult to access; until recently this has resulted in an undersampling of trace gas and gas exchange measurements. The seasonal cycle of sea ice completely transforms the air sea interface and the dynamics of gas exchange. To make estimates of gas exchange in the presence of sea ice, sea ice fraction is frequently used to scale gas transfer parametrisations that were derived in the open ocean. It remains unclear whether this scaling is appropriate for all regions where there is sea ice and whether it is necessary to characterise gas exchange under different sea ice regimes. Ship based eddy covariance measurements were made in the Canadian Archipelago during the summers of 2017 and 2018 from the icebreaker CCGS Amundsen. We will present fluxes of carbon dioxide (CO₂) and methane (CH₄) and will show how they change across the archipelago and under different sea ice conditions. We will explore how the flux changes with wind speed and sea ice fraction. These measurements will be compared with other recent measurements made in the presence of sea ice from the Arctic and Antarctica. We will address the suitability of scaling the open ocean flux by sea ice fraction in the Archipelago.

Quantifying the decadal and global scale impact of tropical cyclones on the ocean carbon sink using remote sensing, in situ and models

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Abstract

Global air–sea carbon dioxide (CO₂) flux and carbon sink estimates often use monthly mean data. This means that the impact of temporally short extreme events, including tropical cyclones or polar lows, are averaged out and thus not well represented within the calculated ocean carbon sink. Tropical cyclones can impact air–sea gas exchange by changing the local wind field and by altering the sea surface temperature. Temperature driven changes in CO₂, and the mixing of nutrients and dissolved inorganic carbon from the deepening of the mixed layer must also be considered. To date, one of the biggest challenges to studying the impact of these storms on the CO₂ gas fluxes has been the absence of suitable high-resolution datasets. Here we provide a first approximation of the impact of tropical cyclones on the air sea CO₂ flux using high resolution Earth observation data of detailed storm tracks provided by the European Space Agency Marine Atmosphere eXtreme Satellite Synergy (MAXSS) project. The MAXSS dataset, along with gridded Surface Ocean Carbon Dioxide Atlas data and the FluxEngine gas flux toolbox allow us to calculate CO₂ fluxes for all individual tropical cyclones between 2010 and 2020. This analysis includes a simple box model to simulate the mixing of dissolved inorganic carbon due each storm deepening the mixed layer. The individual and net impact of the tropical cyclones are determined by comparing the storm driven flux against a reference baseline flux data (calculated using hourly Cross-Calibrated Multi-Platform surface wind data and daily sea surface temperature data). This analysis identifies which processes have the biggest impact on the overall flux (and integrated sink) for each individual storm, and which processes have the largest cumulative impact in specific regions and over time. For example early results using a monthly analysis in the North Atlantic suggest that changes to CO₂ due to mixed layer deepening had a nominal change on the net flux, cooler sea surface temperatures increased the net ocean uptake by $\sim -0.01 \text{ Pg C y}^{-1}$ and wind speed changes were highly variable altering the magnitude and direction of the net flux by between $-0.015 \text{ Pg C y}^{-1}$ and $+0.003 \text{ Pg C y}^{-1}$, where the direction of the flux is primarily governed by the carbonate conditions that exist within the underlying water before the storm passes through.

Oral presentation

On the parameterisation of air-sea gas transfer of CO₂ via wave breaking energy dissipation rate

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Abstract

Air-sea gas exchange of carbon dioxide is a crucial part of both the global climate and ocean biogeochemistry, however a complete characterization of the physical processes needed to model this in all relevant conditions remains elusive. The gas transfer velocity (k) is required to quantify the fluxes and budgets of several important trace gases (e.g., CO₂, DMS, and CH₄). Parameterisation of k must account for both diffusive and bubble-mediated components, and despite consensus that diffusive transfer velocity, k_s , can be modeled as a power law using wind speed and Schmidt number Sc , substantial scatter exists in relationships invoked for the bubble-mediated gas transfer velocity, k_b . Since k_b is driven primarily by entrainment of gases through wave breaking, the uncertainty is acutely problematic at high winds where gas flux measurements are scarce. To address the paucity of such data, the High Wind Gas Exchange Study (HiWinGS) directly calculated gas transfer velocity of CO₂ (k_{CO_2}) from carbon dioxide flux and concentration gradient measurements taken in the Labrador Sea from October 9 – November 13, 2013, where 10-meter neutral wind speeds were between 1.8 – 25.2 m s⁻¹. We use these data to validate a novel gas transfer velocity parameterization constructed using output from a wave hindcast obtained with the spectral wave model (ecWAM) forced with the European Centre for Medium-Range Weather Forecasts (ECMWF) 5th Generation Reanalysis (ERA5). Our parameterisation uses both a diffusive transfer term based on wind speed and Sc , and a bubble-mediated term based on gas solubility, wave age, and wave breaking energy dissipation rate to determine the gas transfer velocity. We compare our results to common wind-speed-only parameterisations and more recent sea-state based relationships.

Modeling Air-Sea Gas Transfer Under Tropical Cyclone Conditions

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Abstract

The gas transfer velocities for different gases, except for relatively soluble gases like methyl acetate, converge under low wind speed conditions when normalized with the Schmidt number. Substantial differences between various gases exist for higher wind speeds due to the contribution of bubble-mediated gas transfer, which depends not only on wind speed but also on gas solubility.

Nevertheless, 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) have shown that during hurricane force winds the dependence of the “surface” (including spray) gas transfer velocity on gas solubility is practically eliminated or significantly reduced. Under hurricane force winds, only 4-5% 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. Andreas (2017) found that spray (spume) can provide a large contribution to the air-sea gas transport under hurricane conditions. In these conditions, the gas flux no longer depends on gas solubility and is proportional to the total surface area or volume of the spray depending on its size and environmental conditions. To gain an insight into the process of the spume generation, we have implemented a multi-phase computational fluid dynamics model, ANSYS Fluent’s Volume of Fluid to Discrete Phase Model (VOF to DPM), which converts water parcels to Lagrangian particles representing sea spray and spume. The model also provides spray size distribution. The total amount of spray dramatically increases with wind. The gas exchange flux due to spume appears to be proportional to its abundance. The spray size distribution gives an estimate of the gas exchange enhancement due to spume. Surfactants also play a role in air sea gas exchange. Our laboratory results from an experiment conducted at the University of Miami found that surfactants alter sea spray generation by forming branch-like rather than finger-like structures, which fragment into differing sizes of sea spray or spume. The VOF to DPM model incorporates the effect of surfactants as well. The inclusion of surfactants increases the overall abundance of spray under tropical cyclone winds. This multi-phase model is consistent with Krall et al. (2019) observations.

Air-sea gas exchange and connections to bubble fluxes at high wind speeds, as revealed by noble gases

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Abstract

Air-sea gas exchange is a crucial part of the biogeochemical cycles of many climatically important gases. Air-sea gas exchange parameterizations differ from each other widely at high wind speeds. In addition, bubbles become increasingly important as wind speed increases, but the gas flux due to bubbles is not explicitly represented in many commonly-used air-sea gas exchange parameterizations. We sought to learn more about gas exchange at high wind speeds and the direct connection between bubbles and overall gas transfer by analyzing a suite of noble gases in a wind wave tank at wind speeds between 20 and 50 m s⁻¹.

Noble gases are ideal tools for studying air-sea gas exchange since they are biologically and chemically inert and hence changes in noble gas concentrations are due primarily to air-sea gas exchange. Thus, in order to improve understanding of air-sea gas exchange at high wind speeds, we measured the gas flux of five noble gases (He, Ne, Ar, Kr and Xe) and oxygen, as well as the steady-state saturation anomalies of these gases, at wind speeds of 20 to 50 m s⁻¹ in the University of Miami's SURge STRUCTure Atmosphere InteractioN (SUSTAIN) salt-water wind-wave tank. Experiments were conducted with monochromatic and with spectral waves, and at water temperatures warmer, equivalent to, and colder than atmospheric temperatures in order to examine different atmospheric stability regimes. We observed an expected increase in noble gas fluxes and noble gas steady state saturation anomalies as wind speeds initially increased but then we found an intriguing and surprising flattening of air injection and steady state saturation anomalies at wind speeds greater than 40 m s⁻¹. We relate this flattening, as well as the gas fluxes in general, to observations of bubble size distribution, turbulent kinetic energy, and wave characteristics (significant wave height, steepness, etc.) that were measured concurrently with the noble gases. We interpret the observed fluxes and steady state saturation anomalies of all five noble gases and oxygen through the use of a simple bubble model, which is based on observed bubble distributions.

Oral presentation

Air-sea gas exchange fluxes and steady state saturation anomalies at very high wind speeds, as revealed by noble gases

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Abstract

Air-sea gas exchange is a crucial part of the biogeochemical cycles of many climatically important gases. Air-sea gas exchange parameterizations differ from each other widely at high wind speeds. In addition, bubbles become increasingly important as wind speed increases, but the gas flux due to bubbles is not explicitly represented in many commonly-used air-sea gas exchange parameterizations. We sought to learn more about gas exchange at high wind speeds and the direct connection between bubbles and overall gas transfer by analyzing a suite of noble gases in a wind wave tank at wind speeds between 20 and 50 m s⁻¹. Noble gases are ideal tools for studying air-sea gas exchange since they are biologically and chemically inert and hence changes in noble gas concentrations are due primarily to air-sea gas exchange. Thus, in order to improve understanding of air-sea gas exchange at high wind speeds, we measured the gas flux of five noble gases (He, Ne, Ar, Kr and Xe) and oxygen, as well as the steady-state saturation anomalies of these gases, at wind speeds of 20 to 50 m s⁻¹ in the University of Miami's SURge STRUCTure Atmosphere InteractioN (SUSTAIN) salt-water wind-wave tank using both discrete measurements of noble gases and a continuous noble gas equilibrator mass spectrometer. Experiments were conducted with monochromatic and with spectral (i.e. JONSWAP) waves, and at water temperatures warmer, equivalent to, and colder than atmospheric temperatures in order to examine different atmospheric stability regimes. We observed an expected increase in noble gas fluxes and noble gas steady state saturation anomalies as wind speeds initially increased but then we found an intriguing and surprising flattening of air injection and steady state saturation anomalies at wind speeds greater than 40 m s⁻¹. Noble gas fluxes and steady state saturation anomalies are correlated most strongly with bubble volumes for the less soluble noble gases (He, Ne and Ar) and with wind speed and wave Reynolds number for the more soluble noble gases. In the JONSWAP experiments, significant wave height was also highly correlated with gas saturation anomalies ($R > 0.92$, $P < 0.05$). Moreover, invasion fluxes (i.e. gas fluxes into the water) were larger than evasion fluxes when other experimental conditions were similar. Taken together, these lab-based experiments suggest more attention should be paid to parameterizations based on wave characteristics and bubbles and that current wind-speed based gas exchange parameterizations should not be applied to conditions with very high wind speeds.

Greenhouse gas fluxes over a boreal river measured with eddy covariance

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Abstract

There is a considerable knowledge gap regarding direct greenhouse gas flux measurements from boreal rivers. To reduce this gap, the Kitinen Experiment (KITEX) was set up in northern Finland in May-October 2018. The goal of the experiment was to use the eddy covariance technique to measure and quantify greenhouse gas fluxes on a boreal river throughout the entire growing period, and to measure the physical drivers of the fluxes in order to gain more knowledge on the physics of the riverine gas transfer.

The river Kitinen runs approximately 260 km in northern Finland. Its catchment area consists mainly of northern boreal forest and wetlands. Although the catchment area is sparsely populated, the river is heavily built with altogether seven hydropower plants and at the experiment site, the flow velocity is almost completely controlled by dam operations nearby. Fluxes and gas concentrations over Kitinen were measured on a floating platform, anchored in the middle of the river. Eddy covariance measurements took place on 15/06-02/10. After data quality and wind direction screening, the temporal coverage was 26% for the carbon dioxide flux and 33% for the methane flux. The mean carbon dioxide flux from the river was $0.7 \mu\text{mol m}^{-2} \text{s}^{-1}$ and the mean methane flux was $3 \text{ nmol m}^{-2} \text{s}^{-1}$. The fluxes were highest in August but the differences between months were small.

The gas exchange coefficient k and its normalised value $k600$ were calculated from the carbon dioxide flux and concentrations in water and above the surface. The mean $k600$ over the entire campaign was 20 cm h^{-1} , which is of the same order of magnitude as estimates from earlier river flux studies. The data analysis will continue with parameterising the calculated fluxes and the gas exchange coefficient with the measured physical quantities, such as total heat flux and water surface turbulence.

Greenhouse gases (CO₂, CH₄ and N₂O) emissions from a tropical micro-tidal estuary (Cochin, India)

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Abstract

Greenhouse gases (GHGs) emissions from human activities are an important trigger of observed climate change since the mid-20th century. Carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O) are the three primary GHGs and the inland aquatic systems also contribute significantly to global GHGs emissions. Estuaries, particularly those in the tropical zones act as an important source of GHGs, attributed to anthropogenic activities apart from receiving large terrestrial inputs. However,

processes regulating annual GHGs emissions from these systems remain poorly constrained. The present study was conducted in a tropical Cochin estuary (CE) located on the southwest coast of India (Ramsar site:1204). Apart from the annual monsoonal input of terrestrial organic matter, large-scale

developmental activities along the banks of this system induced eutrophication. Surveys during early monsoon (April), monsoon (September) and post-monsoon (December) of 2012 showed pCO₂ supersaturation in the upstream but gradually decreased towards the sea. The strong negative correlations with pH ($r=0.78$) and salinity ($r=0.57$) pointed out riverine inputs as the main source of CO₂ in the estuary. The significant seasonal variation in pCO₂ ($p=0.005$) could be attributed to substantial heterotrophic activities and the riverine inputs. On the other hand, the weak negative correlations for CH₄ with salinity ($r=-0.43$) and pH ($r=-0.43$) seemed to suggest that the riverine inputs were only a mild source. A higher concentration of dissolved CH₄ in bottom waters as compared to that in surface waters and its significant positive correlation ($r=0.65$) with ammonia indicates that sediment methanogenesis could be the major contributor of CH₄ in the estuary. N₂O recorded higher

values during the monsoon season. The positive correlation of N₂O with NO₃⁻ + NO₂⁻ during the monsoon season indicates that it originates from the nitrification process. The annual flux was estimated to be 1.01, 0.62 and 0.03 Gg y⁻¹ for respective CO₂, CH₄ and N₂O during the study period shows CE is a significant source of GHGs to the atmosphere. These gaseous export fluxes from CE to the adjacent coastal waters were significant mainly during the monsoon probably as a result of the

increased heterotrophic activities fuelled by the input of a large amount of allochthonous organic matter.

A thermographic approach to measure the wind shear stress at the water surface

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Abstract

Under a wide range of conditions the wind blowing over the water surface is the driving force for the exchange of volatile chemical species and heat across the air-water interface. Considering only the wind speed is not sufficient to understand the mechanisms of mass transfer at the air-sea interface, because the wind stress is partitioned into the viscous shear stress, which determines the thickness of the viscous boundary layers on both sides of the interface, wave-induced shear stress and pressure forces. The two latter are feeding the waves, resulting in a second source of near-surface turbulence by turbulent dissipation of wind waves.

Measuring the viscous shear stress by velocity profiles within the viscous boundary layer at a wavy surface using particle imaging velocimetry or related techniques is very demanding, and there are only a few wind-wave tunnel studies available [1]. Here, a new non-invasive technique which does not need any seeding with particles but tags the water-sided flow with a heat profile induced by a laser line, is reported. The temporal development of the heat profile is measured using a 512x640 pixels, 100-200fps thermal camera. In pilot setups at the Heidelberg Aeolotron, 1450nm laser diodes illuminated one to three narrow, 20-35mm long lines on the water surface perpendicular to the wind direction, with laser power per length of line of approximately 1W/cm. A short heating pulse of 10 to 15ms resulted in a maximum temperature increase of 0.4K, penetrating 320 microns into the water. A velocity gradient at the water surface leads to an enhanced broadening of the line due to Taylor dispersion. Hence the velocity gradient and consequently the viscous shear stress can be determined. First results with a flat water surface as well as in the presence of wind generated waves will be presented, and show a good agreement with direct measurements of the velocity gradient in the water-sided viscous boundary layer using particle streak velocimetry [2, 3]. Following this initial success, systematic measurements at the Aeolotron are planned using a 1567nm 100W erbium fiber laser.

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Diurnal BOOGIE: An investigation into spatiotemporal and climate change effects on organic matter in the sea surface microlayer and its movement between marine and atmospheric environments

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Abstract

The Sea Surface Microlayer (SML) is a complex boundary layer between the ocean and atmosphere that modulates the air-sea gas exchange and has biological, chemical and physical properties that are significantly different from Subsurface Waters (SSW) (Engel et al., 2017). Currently, there is a large uncertainty on how the SML composition of the ocean impacts fluxes of climate active gases and how climate change may influence this process in the future (Woolf et al., 2019).

Previous work has revealed organic matter in the SML, including surfactants (surface-active agents) can suppress the air-sea gas exchange, known as the Surfactant Suppression Effect (SSE; Mustafa et al., 2019; Pereira et al., 2016; Schmidt and Schneider, 2011). The SSE has been related to Sea Surface Temperature (SST) but the rate of suppression across the oceans is variable and likely due to variances in surfactants and their composition in the SML (Pereira et al., 2018). The BOOGIE program (Breathing Oceans: understanding the organic skin that modulates the exchange of greenhouse gases between the atmosphere and the ocean) will explore the spatial and temporal effects of organic matter in the Atlantic Ocean, and its effects on the air-sea gas exchange over time.

Diurnal BOOGIE (PhD project within BOOGIE) aims to understand spatiotemporal challenges, through the production of a SML time series to investigate key processes that drive organic matter changes over time. Here we will present first results from the first field campaign in 2022 that explores seasonal influences (SST and atmospheric deposition) and selective transfer mechanisms between the ocean and atmosphere (van Pinxteren et al., 2020). The effect of these changes on the SSE and air-sea gas exchange will be explored in the Cape Verde region of the Atlantic Ocean using a variety of techniques (e.g. Garrett screen, Air-sea gas exchange tank, Polarograph, LC-OCD-OND-UVD) to examine the SSE and surfactant composition of the SML, SSW, and aerosols in collaboration with the Cape Verde Atmospheric Observatory (CVAO) and Cape Verde Ocean Observatory (CVOO).

Oral presentation

Global estimates of air-sea CO₂ fluxes: Contributions of Wallace Broecker and Taro Takahashi

Author list (presenting author in bold):

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Abstract

Here we describe the fundamental contributions that Wallace Broecker and Taro Takahashi of LDEO/Columbia University made towards determining air-sea CO₂ fluxes through observations and insights on use of radio-isotopes, air-water CO₂ concentration differences, and inorganic carbon chemistry of seawater. The transfer of CO₂ between atmosphere and ocean is a key aspect of the global carbon system, for instance in its use to determine the amount of anthropogenic CO₂ sequestered by the ocean. The first estimates to measure this transfer took advantage of natural radio isotopes, Radon-222 (222Rn) and Carbon-14 (14C). The global exchange using natural 14C was based on the global disequilibrium between atmosphere and ocean. Subsequently, local estimates of gas transfer were obtained based on the deficit of 222Rn in surface water compared to its parent, radium-226, 226Ra. A general agreement between local and global estimates of transfer were obtained. However, the 222Rn estimates did not show any strong correlation with environmental forcing. Subsequent global exchange estimates relied heavily on utilizing the bomb-14C inventories in the ocean and atmosphere. The initial estimates of bomb 14C inventories have been revised downward by ≈30 % through better means to separate the bomb and natural 14C, and improved interpolation methods. The bomb 14C estimate of global CO₂ exchange is a fundamental constraint for parameterizations of air-sea gas transfer with wind. Estimates of regional and global CO₂ fluxes have been obtained using wind speed parameterizations, air-water partial pressure difference of CO₂, and novel interpolation methods. While the global air-sea flux estimates have improved with greatly increased number measurements and new techniques, the contributions of Broecker and Takahashi remain a cornerstone of global ocean carbon cycle research.

Oral presentation

New, substantially larger, estimates of global air-sea CO₂ flux from surface data

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Lonneke Goddijn-Murphy (Environmental Research Institute, University of the Highlands and Islands, UK)

Abstract

In recent years, an international effort has assembled well-documented and quality-controlled data sets for $f\text{CO}_2$, the surface ocean carbon dioxide fugacity. The free availability of these data sets has enabled time-resolved calculations of ocean-atmosphere fluxes of CO₂ from surface observations regionally and globally. However, previous studies have not corrected the data for temperature gradients between the surface and sampling depth at a few metres or for the effect on fluxes of the cool ocean surface skin. We calculate a time history of ocean-atmosphere fluxes of CO₂ from 1992 to 2018 corrected for these effects. These increase the calculated net flux into the oceans by 0.8-0.9 PgCyr⁻¹ over this period, at times doubling the uncorrected values. We estimate the uncertainty in our flux calculations by using both simple and sophisticated interpolation methods, but all configurations give convergent results when estimating fluxes globally after about 2000, or over the northern hemisphere throughout the period. Our corrections reconcile surface fluxes with independent estimates of the increase in ocean CO₂ inventory. Comparison with the inventory suggests that the pre-industrial flux of CO₂ from the open ocean to the atmosphere was ~0.5 PgC yr⁻¹ and that it exhaled mostly from the southern hemisphere.

The effects of surfactants on air-water gas transfer.

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Abstract

Near-surface turbulence is known to be very effective in promoting gas transfer across the air-water interface. The presence of even small quantities of surfactants, however, can drastically dampen this turbulence, thereby reducing the transfer velocity K_L . To systematically investigate the aforementioned damping effects, a series of three-dimensional direct numerical simulations (DNS) was performed, in which the level of surfactant-pollution was varied. In order to produce the near-surface turbulent flow field, isotropic turbulence of fixed intensity was introduced at the bottom of the computational domain.

The presence of surfactants reduces the water-surface tension. Hence, a non-uniform distribution of these surfactants, associated with a nonzero surface divergence, results in a non-uniform surface tension. The latter induces so-called Marangoni forces that act to progressively reduce surface divergence, and consequently the near surface turbulent flow field. In the simulations, the level of pollution is expressed by the ratio of the Marangoni number Ma and the turbulent capillary number Ca_t . In the DNS, convection diffusion equations for the gas concentration at five different Schmidt numbers were solved simultaneously. This enabled us to investigate the scaling of K_L as a function of the Schmidt number, $K_L \sim Sc^q$. This scaling was found to vary from $q=-1/2$ for $Ma/Ca_t = 0$ (clean surface) to $q=-2/3$ for large Ma/Ca_t (very dirty surfaces). The results also show that with increasing Ma/Ca_t the portion of the surface that is virtually surfactant-free (the clean surface fraction) becomes smaller and smaller. Using the above scaling results, a Schmidt number independent equation was derived that relates the mean gas transfer velocity to the aforementioned clean surface fraction for isotropic turbulence. The effects of Reynolds number and non-isotropic turbulence still needs to be verified.

Underway seawater and atmospheric measurements of volatile organic compounds in the Southern Ocean

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Abstract

Volatile organic compounds are present ubiquitously throughout the atmosphere. Particularly over the remote marine atmosphere, they play a role in ozone and particle formation. For gases such as methanol, acetone and acetaldehyde it is unclear whether the ocean acts as a source or a sink. For isoprene and dimethyl sulfide, the oceanic source strength is poorly constrained. This is in part due to a paucity of in situ measurements, especially in remote oceanic regions such as the Southern Ocean.

In this work we present shipborne underway ambient air and seawater measurements of methanol, acetone, acetaldehyde, isoprene and dimethyl sulfide. The ship transected the Atlantic sector of the Southern Ocean at approximately 60° S during March. The measurements were taken using a recently developed Segmented Flow Coil Equilibrator coupled to Proton Transfer Reaction-Mass Spectrometer. Using air and water measurements, hourly fluxes and saturations are computed for a broad range of compounds simultaneously. Binning of the data in 24 hourly bins revealed a diel cycle in acetaldehyde air and water concentrations. A smaller diel cycle was observed for isoprene and acetone surface water concentrations.

Ambient air concentrations of acetone and acetaldehyde were very low, possibly due to the remoteness of the sampling location. The underway measurements suggest that the ocean acts as a net sink for methanol, despite an episode of outgassing observed in an area of high biological activity. Depending on location, the Southern Ocean was either a source or a sink of acetone and acetaldehyde. Using our high temporal and spatial resolution fluxes, we compute that the Southern Ocean is a net weak sink of acetone and acetaldehyde at this time of the year. Underway measurements reveal episodic high concentrations of dimethyl sulfide. Isoprene is supersaturated in the surface ocean and was found to correlate with chlorophyll a. In this dataset, surface isoprene and methanol concentrations correlate, likely due to a common biological origin. Surface acetone, methanol and isoprene concentrations were found to correlate negatively with surface underway fCO_2 , suggesting a role of biology in the production of these compounds.

Oral presentation

Sea ice concentration impacts dissolved organic gases in the Canadian Arctic

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Abstract

The marginal sea ice zone has been identified as a source of different climate active gases to the atmosphere due to its unique biogeochemistry. However, it remains highly undersampled and the impact of summertime changes in sea ice concentration on the distributions of these gases is poorly understood. To address this, we present measurements of dissolved acetone and dimethyl sulfide* in the sea ice zone of the Canadian Arctic from the surface down to 60 m. The measurements were made using a Segmented Flow Coil Equilibrator coupled to a Proton Transfer Reaction Mass Spectrometer. These gases varied in concentrations with depth, with the highest concentrations generally observed near the surface. Underway (3-4 m) measurements showed generally higher concentrations in partial sea ice cover compared to ice-free waters. The combination of a large number of depth profiles and underway measurements at different sea ice concentrations enables proposition of the likely dominant production processes of these compounds in this area. Despite obvious in situ production, we estimate that the sea ice zone is absorbing acetone from the atmosphere. In contrast, DMS is consistently emitted from the ocean, with marked episodes of high emissions during ice-free conditions, suggesting that DMS is produced in ice covered areas and emitted once the ice has melted. These novel measurements and insights will allow us to better constrain the cycling of acetone and dimethyl sulfide in the polar regions and their effect on the oxidative capacity and aerosol budget in the Arctic summertime atmosphere.

*Methanol, acetaldehyde and isoprene were also measured during this deployment. Please get in touch with the author if these gases are of particular interest to you.

The peculiar characteristics of air-water gas transfer across a broken surface

Author list (presenting author in bold):

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Abstract

Air-water gas transfer is usually framed as a diffusive process across a defined interface. A “broken surface” implies that instead of a single surface, there is a main interface between the air and water and additional boundaries at the surface of bubbles and particles. Gas transfer across a broken surface is qualitatively different and challenges paradigms derived from the standard framing.

Bubbles and particles can be created in many industrial and natural processes, but the focus here will be on bubbles and drops generated by deep-water breaking on the wind-driven open ocean. When we consider the total effect of the formation of bubbles and drops at the sea surface, we should include the interaction with surface stirring and direct transfer across the sea surface, but the most interesting part is the “mediated” transfer that uses a bubble or particle as an intermediate reservoir in transit between ocean and atmosphere. With some caveats (notably for droplets), we can treat the mediated transfer as a parallel process that adds to the direct transfer. The process is a sequence with important subtleties that differ between bubbles and aerosol. The formation of bubbles and drops depends, at a minimum, on wind speed, sea state and water temperature. The suspension of bubbles and aerosol will also depend on the wind and interactions of the wind with the wave field. The internal dynamics and the transfer at the surface of a bubble depend on hydrostatic pressure, surface tension, the saturation of nitrogen and oxygen and specific bubble and gas properties. For aerosol, similar principles apply, but humidity profiles displace nitrogen and oxygen saturation. Parameterization of mediated gas transfer requires all of the above to be encapsulated. A description is emerging, which provides a complementary perspective to the simpler models of air-water gas transfer.

The peculiarities of transfer across a broken surface are interesting and important. The practical importance is made apparent by considering the calculation of global air-sea fluxes. These calculations are sensitive to the dependence of gas transfer velocities on wind speed, sea state and water temperature; and can be biased by asymmetries in exchange. It is relatively simple, conceptually and computationally, to propagate the uncertainty in the dependence on wind speed to the final flux, but other uncertainties are more difficult to evaluate and to propagate. While we have, for example, credible estimates of the global air-sea flux of carbon dioxide, not all uncertainties are fully explored and some surprises may await. Air-water gas transfer should be studied further, with an emphasis on mechanistic processes such as mediation of transfer by bubbles and aerosol, since the mechanistic insight is essential to confidence in the final result.

Autonomous eddy covariance system for long-term shipboard measurements of CO₂ flux - setup, data processing, and uncertainty analysis

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Abstract

Here we describe an autonomous eddy covariance (EC) system that has been used to continuously measure CO₂ flux on a research ship from 2018 to present. The large dataset requires streamlining of the data processing steps and enables a better understanding of the uncertainties in EC CO₂ flux measurements.

We find that with an ideal setup (i.e. well exposed and aligned EC system, latest closed-path CO₂ analyzer with a dryer, high flow rate and accurate delay estimate), the vast majority of the uncertainty in air-sea CO₂ flux is random. Ambient variability in atmospheric CO₂ and motion-sensitivity of the CO₂ sensor are the largest contributors to the random flux uncertainty, followed by motion-correction of the measured wind velocities. With the latest infrared absorption and cavity-based CO₂ sensors, the white noise contribution to the random flux uncertainty is relatively minor.

Temporal averaging greatly reduces the random uncertainty, but only to an extent. Natural variability starts to dominate once the temporal averaging period exceeds ~6 hours. The bias in our mean CO₂ flux measurements is estimated to be less than 10%. These uncertainty considerations are important when comparing EC CO₂ fluxes against CO₂ fluxes computed with the bulk formula.

Natural variability in air–sea gas transfer efficiency of CO₂

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Abstract

The flux of CO₂ between the atmosphere and the ocean is often estimated as the air–sea gas concentration difference multiplied by the gas transfer velocity (K₆₆₀). The first order driver for

K₆₆₀ over the ocean is wind through its influence on near surface hydrodynamics. However, field observations have shown substantial variability in the wind speed dependencies of K₆₆₀. In this study we measured K₆₆₀ with the eddy covariance technique during a ~ 11,000 km long Southern Ocean transect. In parallel, we made a novel measurement of the gas transfer efficiency (GTE) based on partial equilibration of CO₂ using a Segmented Flow Coil Equilibrator system. GTE varied by 20% during the transect, was distinct in different water masses, and related to K₆₆₀. At a moderate wind speed of 7 m s⁻¹, K₆₆₀ associated with high GTE exceeded K₆₆₀ with low GTE by 30% in the mean. The sensitivity of K₆₆₀ towards GTE was stronger at lower wind speeds and weaker at higher wind speeds. Naturally-occurring organics in seawater, some of which are surface active, may be the cause of the variability in GTE and in K₆₆₀. Neglecting these variations could result in biases in the computed air–sea CO₂ fluxes.

Global synthesis of air-sea CO₂ transfer velocity estimates from ship-based eddy covariance measurements

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Abstract

The air-sea gas transfer velocity (K_{660}) is typically assessed as a function of the 10-m neutral wind speed (U_{10n}), but there remains substantial uncertainty in this relationship. Here K_{660} of CO₂ derived with the eddy covariance (EC) technique from eight datasets (11 research cruises) are reevaluated with consistent consideration of solubility and Schmidt number and inclusion of the ocean cool skin effect. K_{660} shows an approximately linear dependence with the friction velocity (u^*) in moderate winds, with an overall relative standard deviation (relative standard error) of about 20% (7%). The largest relative uncertainty in K_{660} occurs at low wind speeds, while the largest absolute uncertainty in K_{660} occurs at high wind speeds. There is an apparent regional variation in the steepness of the K_{660} - u^* relationships: North Atlantic \geq Southern Ocean $>$ other regions (Arctic, Tropics). Accounting for sea state helps to collapse some of this regional variability in K_{660} using the wave Reynolds number in very large seas and the mean squared slope of the waves in small to moderate seas. The grand average of EC-derived K_{660} ($-1.47 + 76.67u^* + 20.48u^{*2}$ or $0.36 + 1.203U_{10n} + 0.167U_{10n}^2$) is similar at moderate to high winds to widely used dual tracer-based K_{660} parameterizations, but consistently exceeds the dual tracer estimate in low winds, possibly in part due to the chemical enhancement in air-sea CO₂ exchange. Combining the grand average of EC-derived K_{660} with the global distribution of wind speed yields a global average transfer velocity that is comparable with the global radiocarbon (¹⁴C) disequilibrium, but is \sim 20% higher than what is implied by dual tracer parametrizations. This analysis suggests that CO₂ fluxes computed using a U_{10n}^2 dependence with zero intercept (e.g., dual tracer) are likely underestimates at relatively low wind speeds.

Poster presentation

Whitecaps Identification and Extraction in Infrared Imagery Using Machine Learning

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Abstract

Whitecaps are the direct manifestation of wave breaking and are often classified as either active (stage A) or residual (stage B). Infrared (IR) imagery provides a reliable and objective discrimination between whitecaps stages based on their unique signals at IR wavelengths. Each whitecap stage is related to different physical and chemical processes which alter the marine boundary layer and have direct or indirect influence on gas transfer. Measurement of whitecap stages is therefore essential to quantify these processes. To this end, we will present results from a whitecap study cruise in the Gulf of Mexico. Over 60 hours of video were recorded by infrared imagery system mounted at the port-side. Whitecap fraction (W), the percentage area of whitecaps over a region of interest, is used to quantify coverage, with both whitecap stages contributing ($W = W_A + W_B$). Here we apply machine learning to track individual whitecaps and extract W , W_A , and W_B . Preliminary analysis shows that machine learning provides a more robust method to identify whitecap stages when compared to simple thresholding which is typically applied. The method will be presented and results will be explored in relation to environmental conditions and compared to previous results.

Poster presentation

The Impact of Wind Gusts on the Ocean Skin Layer

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Abstract

The thermodynamic and emissive properties of the ocean thermal skin layer are crucial contributors to air-sea heat flux. In order to properly observe ocean surface temperature without disturbing any delicate fluid mechanical processes, thermal infrared imaging is often used. However, wind impacting the ocean surface complicates the extraction of meaningful information from thermal imagery; this is especially true for transient forcing phenomena such as wind gusts. Here, we describe wind gust-water surface interaction through its impact on skin layer thermal and emissive properties. Two key physical processes are identified: (1) the growth of centimeter-scale wind waves, which increases interfacial emissivity and (2) microscale wave breaking and shear, which mix the cool skin layer with warmer millimeter-depth water and increase the skin temperature. As more observations are made of air-sea interaction under transient forcing, the full consideration of these processes becomes increasingly important.

Using Ship-Deployed High-Endurance Unmanned Aerial Vehicles for the Study of Ocean Surface and Atmospheric Boundary Layer Processes

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Abstract

Unmanned aerial vehicles (UAVs) are proving to be an important modern sensing platform that supplement the sensing capabilities from platforms such as satellites, aircraft, research vessels, moorings, and gliders. UAVs, like satellites and aircraft can provide a synoptic view of a relatively large area. However, the coarse resolution provided by satellites and the operational limitations of manned aircraft and ships has motivated the development of unmanned systems. UAVs offer unparalleled flexibility of tasking; for example, low altitude flight and slow airspeed allow for the characterization of a wide variety of geophysical phenomena at the ocean surface and in the marine atmospheric boundary layer. Here, we present the development of cutting-edge payload instrumentation for UAVs that provides a new capability for ship-deployed operations to capture a unique, high-resolution spatial and temporal variability of the changing air-sea interaction processes than was previously possible. The instrument payloads are built with a modular design for ease of interchangeability. Additionally, we implement a novel capability for vertical take-off and landing (VTOL) from research vessels. We succeeded in the first fully-autonomous deployment of a hybrid-VTOL fixed winged UAV from a moving ship on the open ocean, with an endurance of over 12 hours and the ability for multiple aircraft tandem orchestrated simultaneous flight. Real-time high-bandwidth data telemetry (100+ Megabits at up to 50 nm) allowed for the ability to adapt to observations in real-time for more efficient and targeted measurements towards our science goals. The payloads developed include thermal infrared, visible broadband and hyperspectral, and near-infrared hyperspectral high-resolution imaging. Additional capabilities include quantification of the longwave and shortwave hemispheric radiation budget (up- and down-welling) as well as direct air-sea turbulent fluxes. These technological advancements provide the next generation of instrumentation capability for UAVs. We will demonstrate these capabilities by showing the results two field campaigns, one in rural Alaska in Spring 2018 and 2019, and the other aboard the R/V Falkor near Fiji in Nov-Dec 2019. For example, we highlight the use of UAVs for reconnaissance to find features of interest that included large-scale temperature fronts, the discovery of floating pumice on the ocean surface likely the remnants of an undersea volcanic eruption near Tonga, and the discovery of a number of gigantic *Trichodesmium* blooms. When deployed from research vessels, UAVs will provide a transformational science prism unequalled using 1-D data snapshots from ships or moorings alone, and improve asset mobilization for targeted efficient data collection.