Modification of Turbulence at the Air-Sea Interface Due to the Presence of Surfactants and Implications for Gas Exchange. Part I: Laboratory Experiment

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 Modification of turbulence at the air–sea interface due to the presence of surfactants and implications for gas exchange. Part I: Laboratory experiment

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Abstract. The air-sea gas transfer of gases like CO2 is substantially determined by the properties of the aqueous diffusion sublayer and free-surface turbulent boundary layer. Little is known about the effect of surfactants on turbulence in the near-surface layer of the ocean. In order to investigate the effect of surfactants on turbulent exchanges below the air-sea interface, we have conducted a series of laboratory experiments at the UM RSMAS Air-Sea Interaction Saltwater Tank (ASIST) facility. Results from these experiments demonstrate that the surfactant monolayer suppresses turbulence and reduces drag below the water surface and increases the surface drift velocity. This effect is important for parameterization of the interfacial component of gas exchange under low wind speed conditions. From the theoretical standpoint, the mechanism of the turbulence reduction can be explained by the modification of the “streaks” in the buffer zone near the interface by visco-elastic properties of the water surface when surfactants are present. These findings are consistent with results from high-resolution non-hydrostatic numerical simulations presented in a companion paper.
Key Words: laboratory experiments, surfactants, near-surface turbulence

1. Introduction

The effect of surfactants on the air-sea exchange processes is of primary importance at low wind speeds (Frew 1997; Atmane et al. 2004). The resistance to air-sea gas transfer is associated with the aqueous diffusion sublayer, which is of the order of 50 µm thick (Bolin 1960). This diffusion sublayer is a result of the interaction of turbulence with the sea surface. The effect of surfactants on the
gravity-capillary waves has been intensively studied in relation to water wave damping (Alpers and Hühnerfuss 1989) and to synthetic aperture radar (SAR) imaging of the ocean surface (Hühnerfuss et al. 1987; Gade et al. 1998). However, less is known about the effect of surfactants on near-surface turbulence.

The aim of this paper is to investigate the effect of surfactants on near-surface turbulence in application to the problem of air-sea gas exchange. For this purpose, we have conducted a series of laboratory experiments and numerical simulations (in an accompanying article by Matt et al. this volume).

Section 2 of this paper describes instrumentation and techniques of laboratory experiments and provides an overview of the measurements conducted at the ASIST facility. Section 3 considers the effect of surfactants on surface roughness from the air side of the air-sea interface. The effect of surfactants on turbulence in the near-surface layer is analyzed in Section 4. Section 5 is the discussion of the results; Section 6 concludes the article.

2. Measurements

2.1. Experimental techniques

Experimental data presented in this work were acquired in the Air-Sea Interaction Saltwater Tank (ASIST) at the Rosenstiel School of Marine and Atmospheric Science, University of Miami (UM RSMAS).

ASIST is a state-of-the-art wind-wave tank (Figure 1), its primary purpose is to study various physical processes at the air-sea interface (Donelan et al. 2010;
Haus et al. 2010; Babanin and Haus 2009).

The working section of ASIST is 15 m long, 1 m wide and 1 m high; in a typical setup, the water level is 0.42 m. For simulations of realistic ocean conditions, it is equipped with a digital mechanical wave generator (wave frequencies 0.25 Hz–3 Hz, amplitudes 0–0.1 m), wind generator (wind speed in the tank’s centerline 0–30 m s$^{-1}$), current generator (current speed 0–0.5 m s$^{-1}$), and water temperature control (5–40ºC). The air circulation has a closed and open loop option. In the open loop option fresh air is coming from the atmosphere and being exhausted back after passing the working section (Figure 2). Open loop mode is used for gas, humidity, and heat transfer to avoid quick saturation. In our experiments, we have used only closed loop mode. We did not use mechanical wave and current generation.

Fully transparent acrylic glass side walls, bottom, and top of the tank allow using optical non-intrusive methods for flow measurements and visualization. The methods used in our experiments are described below.

### 2.1.1 Digital Laser Elevation Gauge

The Digital Laser Elevation Gauge (DLEG) (Savelyev 2009) is a setup using a line-scan camera and a laser beam crossing the water surface. Two Argon-Ion (beam power 150 mW, wavelength 488 nm) air-cooled lasers are equipped with beam splitters and mirrors to provide up to six vertical beams at any points in the tank. The intersection of these beams with the surface is detected by line-scan cameras looking at the beam through the tank’s side wall. Fluorescein added to the water makes the laser beam highly visible; this creates a brightness contrast as the beam crosses the air-water interface. The brightness threshold location on a line image signifies the water elevation. Each camera has a one-dimensional sensor consisting of 2048 pixels and sampling rate up to 1000 Hz (we only used 1024 pixel resolution and 250 Hz sampling rate in our experiments) and the range of heights is determined by the choice of lenses. This technique provides the most

![Figure 2](image-url)  
**Figure 2** Air-Sea Interaction Saltwater Tank schematics.
accurate surface elevation measurement (maximum resolution < 0.2 mm).

For sequences of surface elevation images, an edge detection algorithm was developed, which provided a clean and continuous surface elevation signal (Figure 3).

2.1.2 Digital Particle Image Velocimetry

The Digital Particle Image Velocimetry (DPIV) system manufactured by Dantec Dynamics was used for flow measurements and visualization. For this purpose water was seeded with polyamide spheres (50 μm diameter) and illuminated by two consecutive laser sheet pulses 4–7 ms apart. The laser sheet was positioned vertically along the centerline of the wave tank, or horizontally

![Figure 3](image)

**Figure 3** Example of a surface elevation signal acquired by a line scan camera. Top image represents multiple vertical scans stacked together. Bottom plot is the resulting water elevation.

![Figure 4](image)

**Figure 4** Velocity vector map, deduced from DPIV images using the adaptive correlation algorithm provided by Dantec Dynamics in its Flowmanager software.
below the water surface, at about 6.6 m wind fetch. A digital Hisense camera synchronized with laser pulses recorded pairs of consecutive images. The adaptive correlation algorithm developed by Dantec Dynamics is applied to these images to yield velocity maps of about 10x7 cm² in size, with grid resolution of approximately 1 mm (see Savelyev (2009) for more detail). These maps were acquired at a 15 Hz sampling rate, 150 realizations for each flow regime, and provided sufficient data for ensemble averaging of turbulent properties. An example of a flow map resulting from the processing of one pair of images is shown in Figure 4.

2.2 Experimental setup and conditions

In this article, we discuss results from the DPIV and elevation measurements that are summarized in Table 1. For the DPIV, we describe two experimental setups: in the first setup, we use the DPIV with a vertical laser sheet to measure a vertical section of near-surface velocities (Experiment 1 and Experiment 2); the second setup uses the DPIV with a horizontal laser sheet 2 cm below the water surface to take measurements of a horizontal field of sub-surface velocities (Experiment 3). The dates of the experiments are 13 February, 23 March, and 16 April 2009, respectively. The air and water temperatures for Experiment 2 are $T_{\text{air}} = 24.5°C$ and $T_{\text{water}} = 23.2°C$, and for Experiment 3, $T_{\text{air}} = 24.6°C$ and $T_{\text{water}} = 24.3°C$. The air and water temperatures for Experiment 1 were not measured (we believe they were similar to the temperatures in Experiment 2). As a part of Experiment 2, we also collected elevation data to study the effect of surfactants on short surface waves.

Table 1  Summary of laboratory experiments at ASIST. In the text, data collected on 13 February, 23 March, and 16 April 2009 are referred to as Experiment 1, 2, and 3, respectively. *DPIV pulse interval of 7 ms, all others 4 ms.

<table>
<thead>
<tr>
<th>Date, 2009</th>
<th>Run #</th>
<th>Measurement</th>
<th>$U_{10}$, m s⁻¹</th>
<th>Surfactant</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>13 February</td>
<td>1.7</td>
<td>Vertical DPIV*</td>
<td>2.9</td>
<td>None</td>
<td>Figure 8a</td>
</tr>
<tr>
<td>13 February</td>
<td>1.8</td>
<td>Vertical DPIV*</td>
<td>2.9</td>
<td>Oleic acid</td>
<td>Figure 8a</td>
</tr>
<tr>
<td>23 March</td>
<td>2.3</td>
<td>Vertical DPIV</td>
<td>2.9</td>
<td>None</td>
<td>Figure 8b</td>
</tr>
<tr>
<td>23 March</td>
<td>2.8</td>
<td>Vertical DPIV</td>
<td>2.9</td>
<td>Oleyl alcohol</td>
<td>Figure 8b</td>
</tr>
<tr>
<td>23 March</td>
<td>2.11</td>
<td>Elevation Gauge</td>
<td>11.0</td>
<td>None</td>
<td>Figure 7</td>
</tr>
<tr>
<td>23 March</td>
<td>2.12</td>
<td>Elevation Gauge</td>
<td>7.0</td>
<td>None</td>
<td>Figure 6</td>
</tr>
<tr>
<td>23 March</td>
<td>2.13</td>
<td>Elevation Gauge</td>
<td>2.9</td>
<td>None</td>
<td>Figure 5</td>
</tr>
<tr>
<td>23 March</td>
<td>2.14</td>
<td>Elevation Gauge</td>
<td>2.9</td>
<td>Oleyl alcohol</td>
<td>Figure 5</td>
</tr>
<tr>
<td>23 March</td>
<td>2.15</td>
<td>Elevation Gauge</td>
<td>7.0</td>
<td>Oleyl alcohol</td>
<td>Figure 6</td>
</tr>
<tr>
<td>23 March</td>
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<td>Elevation Gauge</td>
<td>11.0</td>
<td>Oleyl alcohol</td>
<td>Figure 7</td>
</tr>
<tr>
<td>16 April</td>
<td>3.2</td>
<td>Horizontal DPIV</td>
<td>2.9</td>
<td>None</td>
<td>Figure 9a, 10</td>
</tr>
<tr>
<td>16 April</td>
<td>3.7</td>
<td>Horizontal DPIV</td>
<td>2.9</td>
<td>Oleyl alcohol</td>
<td>Figure 9b, 10</td>
</tr>
<tr>
<td>16 April</td>
<td>3.8</td>
<td>Horizontal DPIV</td>
<td>2.9</td>
<td>Oleyl alcohol</td>
<td>Figure 10</td>
</tr>
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We diluted the surfactant, oleic acid or oleyl alcohol - both are insoluble in water - in 95% ethanol to a concentration of approximately 8 mmol/liter and 3 mmol/liter, respectively. We added 60 ml of the surfactant solution to the wave tank for each successive experimental run. The surfactant was added with a syringe through an opening in the side of the tank. Care was taken to minimize the disturbance created on the water surface by the addition of surfactant. The surfactant was released at the upwind side of the tank. The surfactant plume then transited through the measurement zone with the surface drift current generated by the wind stress. The DPIV and laser elevation measurements were timed to coincide with the passage of the surfactant plume.

In our experiments, we released surfactant at concentrations exceeding the saturation level. These surfactants form a monomolecular layer on the water surface, where excess surfactant is concentrated in microscopic droplets, so-called “lenses”. The surfactant removed from the surface by renewal processes is resupplied from these surfactant lenses and the saturated films thus act on surface waves like a monolayer completely covering the surface. The dynamic elasticity of such a film is constant (Ermakov 2007), which corresponds to a constant decrement of damping of small-scale surface waves on film-covered water.

The assumption of constant dynamic elasticity is also used in the implementation of an elastic boundary condition for the numerical simulations of the effect of surfactants on near-surface turbulence described in a companion paper (Matt et al. 2010).

3. Measurements of the effect of surfactants on surface roughness

The damping effect of surfactants on small-scale surface gravity-capillary waves is well known and is due to the dilational elasticity of the monomolecular surface film (Hühnerfuss et al. 1987). We reproduced this damping effect in the laboratory experiments for three different wind conditions (Figures 5, 6, 7).

At the lowest wind speed $U_{10} = 2.9 \text{ m s}^{-1}$, the resolution from the elevation gauge limited the observation of damping for the very low elevation waves (Figure 5). The damping was still clearly visible by eye at $U_{10} = 2.9 \text{ m s}^{-1}$ but is more readily observed for higher wind speeds, $U_{10} = 7 \text{ m s}^{-1}$ and $U_{10} = 11 \text{ m s}^{-1}$ (Figures 6, 7).

Suppression of surface waves due to surfactant decreases the sea surface roughness, which affects the transfer of momentum from the air to the ocean; this is equivalent to the reduction of the drag coefficient from the air side. Since the main resistance to the air-sea gas transfer is on the water side of the air-sea interface, it is important to understand how the presence of surfactants affects near-surface turbulence. For analysis of the turbulence regime below the water
surface in the presence of surfactant, we selected the low wind speed case (Figure 5).

Figure 5  Wind wave spectra of clean water surface and in the presence of oleyl alcohol surface film – measurements in ASIST facility of UM RSMAS. The 95% confidence interval is shown by the dash-dot lines. The wind speed referenced to a 10 m height was approximately $U_{10} = 3 \, \text{m} / \text{s}$. The dotted line represents the ratio of wave damping decrements for surfactant-covered and clean surface for oleyl alcohol (eqs. 1-4 in Hühnerfuss 1987).

Figure 6  As Figure 5 but for wind speed $U_{10} = 7 \, \text{m/s}$. 
4. Effect of surfactants on near-surface turbulence

Continuity of momentum flux at the air-water interface implies that

\[ M = C_a U_a^2 = C_w U_w^2, \]  
(1)

where \( U_a \) is the wind speed and \( C_a \) is the drag coefficient from the air side; \( U_w \) is the speed of the wind induced current and \( C_w \) is the drag coefficient from the water side.

The addition of surfactant reduces sea surface roughness from the air side of the interface due to suppression of short gravity-capillary waves. This results in reduction of the drag coefficient from the air side and thus a reduction of the momentum flux at the air-sea interface. The momentum flux at the air-water interface in the presence of surfactants is

\[ M' = C'_a U_a'^2 = C'_w U_w'^2, \]  
(2)

where the prime denotes the same variable as in (1) but after the addition of surfactants and \( C'_a < C_a \).

In our measurements with DPIV using a vertical laser sheet, we observed that the presence of a surfactant film increases the surface drift velocity by approximately 25% (Figure 8). This observational result can be expressed as follows:

\[ U_w' \approx 1.25 U_w. \]  
(3)

Combining (1), (2) and (3), we obtain the following relationship:
\[ C'_{w} = C_w (U_w/U'_w)^2 M'/M. \]  

(4)

If we assume for a moment that \( M' = M \), then an estimate of the drag coefficient from the water side in the presence of surfactants according to (3), (4)
will be as follows:

\[ C'_w \approx C_w \left( \frac{1}{1.25} \right)^2 = 0.64 C_w. \]  

(5)

This is equivalent to the reduction of the drag coefficient from the water side by 36% due to effect of surfactants.

The momentum transmitted from the air to the water surface is reduced in the presence of surfactant due to reduced surface roughness from the air side of the interface, which means that at the same wind speed \( M' < M \). Consequently, according to (4), the effect of surfactants on the drag coefficient from the water side in reality is even stronger.

In order to understand the nature of the effect of surfactants on the drag reduction and increase in surface drift velocity, we investigated the horizontal structure of the velocity field just below the water surface by using the DPIV with a horizontal laser sheet at 2 cm depth. Without addition of surfactant, we observed “streak-like” features oriented in the along-tank direction. In our experiment, the streak-like features appear noticeably damped after the addition of surfactant (Figure 9). This damping is also reflected in the corresponding values of variance of velocity components shown in Table 2. Spectra calculated from the fields in Figure 9 - and averaged over space and time - confirm the damping for wavelengths in the centimeter range (Figure 10). Note that these spectra are calculated across the tank from the along tank \((u)\) and transverse \((v)\) velocity components. Experiment details are given in Table 1.

### 5. Discussion

The streak-like features observed in our experiment have previously been reported from experiments and numerical simulations near the rigid wall (Lesieur 2008) and below the free surface (Dhanak et al. 1999; Tsai 2001). Streaks of low and high longitudinal velocity relative to the local mean velocity profile are a generic feature of the turbulent boundary layer near a rigid wall. These streaks are a type of coherent structure developing in the buffer layer between the viscous sublayer and the area of developed turbulence. Such streaks are observed between
approximately $z^+ = 5$ and $z^+ = 40 - 50$, when scaled in wall units, $z^+ = z/(\nu/\text{u}_*)$, where $z$ is the distance to the surface, $\nu$ is the kinematic viscosity, and $\text{u}_*$ is the friction velocity. The streaks near the rigid wall are of spanwise size of about 100 wall units and of average length of 500 wall units (Lesieur 2008).

Figure 11 shows a schematic representation of the effect of the surfactant monolayer on the near-surface circulation. Concentration of the surfactant in convergence zones increases, which results in the reduction of surface tension. Concentration of the surfactant in divergence zones decreases, which results in the increase in surface tension. As a result, forces opposing fluid motion develop at the water surface, which results in suppression of velocity fluctuations in the vicinity of the surface. The thickness of the affected layer can be linked to Kolmogorov’s internal scale of turbulence, which near the surface can be scaled in wall units, $\nu/\text{u}_*$.

Near rigid walls, the near-surface streaks are subject to the Tollmien-Schlichting (T-S) type instability leading to the development of ‘hairpin vortices’ and ejection of fluid from the viscous sublayer (Kim et al. 1987). A similar instability, though possibly not exactly of the same type as that near the rigid wall, can also develop near a flexible wall (Benjamin 1960; 1963) or a free surface (Caulleiz et al. 2007). Soloviev and Lukas (2006) linked this type of instability (resulting in fluid ejection from the near surface layer) to ‘ramp-like structures’, which are almost always observed in turbulent boundary layers in atmosphere (Antonia et al. 1979) and ocean (Thorpe 1985; Soloviev 1990). This type of coherent structure is responsible for about 40% of the momentum transport across the boundary layer.

A consequence of streak suppression below the surface is the reduction of the drag coefficient from the water side of the air-water interface, which may explain
the effect of surface drift velocity increase observed in our laboratory experiment in the presence of surfactants.

Suppression of turbulence below the water surface due to the presence of surfactants should also lead to the reduction of the air-sea gas exchange velocity. In this laboratory experiment we did not investigate the effect of surfactants on gas transfer velocity. This effect is discussed in more detail in the companion paper (Matt et al. this issue).

6. Conclusions

Understanding the effect of surfactants on the air-sea gas exchange is important for estimation of the CO₂ uptake by the oceans in low wind speed zones. A series of laboratory experiments conducted at the Air-Sea Interaction Saltwater Tank (ASIST) facility has demonstrated that the surfactant monolayer suppresses turbulence and reduces the drag coefficient below the water surface and increases the surface drift velocity. We assume that “streaks” and their intermittent instability are essential phenomena in this process. Suppression of the near-surface turbulence due to the elastic properties of the surface films is expected to reduce the air-sea gas transfer velocity.

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