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Kyoto University
Characteristics of gas-flux density distribution at the water surfaces

Wu-ting Tsai¹ and Li-ping Hung²

¹ Institute of Hydrological and Oceanic Sciences, National Central University, Jhong-li City, Tao-yuan 32001, Taiwan, E-mail: wtttsai@ncu.edu.tw
² Image Processing and Modeling, Interdisciplinary Center for Scientific Computing, University of Heidelberg, Speyerer Straße 6, D-69115 Heidelberg, Germany, Email: li-ping.hung@iwr.uni-heidelberg.de

Abstract. Numerical simulation of a wind-driven turbulent flow underneath a wavy surface is conducted to characterize the distribution of gas-flux density across the water surface, and to explore the potential correlation between the gas-flux density and other surface quantities, including the temperature, the wave slope and the velocity divergence. It is found that the underlying elongated streamwise eddies, which are either associated with the coherent vortical flows within the turbulent boundary layer or formed from the nonlinear interaction between the drift current and the surface waves, contribute to the major transfer rate at the water surface. These longitudinal eddies induce streaky thermal signature at the water surface, which is found to highly correlate with the gas-flux density.

Key Words: gas flux, surface waves, turbulence, numerical simulation

1. Introduction

The air-water exchange of weakly soluble gases is generally governed by the nonlinear interactions between the wavy interface and the underlying aqueous flow. These interaction processes also result in various characteristic signatures of temperature, roughness and velocity at the water surface. This provides the basis for the use of heat as a proxy tracer in conjunction with infrared thermography to measure the instantaneous air-water gas transfer velocity (e.g., Haußecker et al. 1995). Similar attempts have also been undertaken to relate air-water gas flux to surface slope (e.g., Bock et al. 1999) or surface velocity divergence (e.g., McKenna and McGillis 2004).

The objective of this study therefore is two-fold: to characterize the surface signatures induced by the underlying coherent flows; and to quantify the relationship between the gas-flux density and various surface quantities, such as temperature, wave slope and velocity field. Our approach is to conduct numerical
simulation of a model problem, in which we can extensively characterize the state of the flows and the interface. The focuses are to identify the predominant signatures of gas-flux density at the water surface, and then to reveal the underlying flow processes associated with these dominant interfacial transfer.

2. Numerical simulation

We consider the three-dimensional flow of a viscous fluid bounded by a free-moving water surface. A numerical model has been developed by the present authors for simulating such a fully nonlinear free-surface flow (Tsai and Hung 2007). The model solves the complete conservation equations of mass and momentum subject to the fully-nonlinear boundary conditions on the water surface, and can accurately resolve surface waves of various length scales, ranging from the gravity waves to capillary ripples.

Advection-diffusion equations governing the temperature field of the sensible heat $\theta$ and the gas concentration $c$ are also integrated with the flow simulation. For the transport of sensible heat, the air temperature and humidity are considered to remain constants to maintain constant evaporation. This results in a uniform sensible heat flux at the water surface, which gives rise to a Neumann condition for the surface temperature. In our simulations, the temperature field is treated as a passive tracer; the buoyancy effect due to temperature fluctuations and the imposed surface heat flux does not modify the vertical momentum equation. While in the case of transfer of sparingly soluble gas, the resistance is dominated by the subsurface aqueous flow, and the atmosphere can be regarded as an infinite reservoir with a constant tracer gas concentration. This results in a Dirichlet condition for the concentration field at the water surface.

Being restricted by the computing capacity, the diffusion coefficients of sensible heat and dissolved gas, $\nu_\theta$ and $\nu_c$, are assumed to be the same as the kinematic viscosity $\nu$, i.e., both the Prandtl number $Pr = \nu/\nu_\theta$ and the Schmidt number $Sc = \nu/\nu_c$ are equal to one. The thicknesses of viscous, thermal and mass sublayer immediately next to the water surface, therefore, are of the same order of magnitude; and the transport of the scalar across the diffusion sublayer is controlled by the boundary condition at the water surface (Tsai et al. 2005).

The simulation is posed to mimic the subsequent development of a mechanically-generated gravity-capillary wave driven by a wind field. The wavelength of the carrier wave $\lambda=7.5$ cm, the phase velocity $c \approx 34$ cm/s, and the steepness $ak \approx 0.25$. The action of the wind field results in distributions of shear stress $\tau = 0.5\tau_0 [1 + \cos(kx - ct + \Theta_t)]$ and normal stress $\tau_n = p_0 \cos(kx - ct + \Theta_n)$, where the amplitudes of the shear stress $\tau_0 = 2$ dyne/cm² and the normal stress $p_0 = 2$ dyne/cm², the phase lags $\Theta_t = 5/18\pi$ and $\Theta_n = 8/9\pi$. These stress distributions
are idealized approximations of the laboratory measurements by Banner (1990) and Banner and Peirson (1998).

3. Results and discussion

3.1 Surface signatures

Three-dimensional prospective surface profiles $\eta$ and the distributions of surface temperature $\Theta_s$, showing four characteristic surface signatures in the evolution of the simulation results, are depicted in Figure 1. The corresponding distributions of gas-flux density $j_s$ and cross-wind velocity on the water surface $v_s$, and the distribution of streamwise vorticity $\omega_x$ on a representative cross-stream vertical section are also plotted in the figure.

The first two characteristic surface signatures, which appear rapidly at the early stage of the simulation, are the parasitic capillary ripples riding on the carrier gravity wave and the elongated thermal streaks (Figure 1a and b). These elongated streaks with narrower spacing are postulated to be induced by the coherent streamwise eddies within the turbulent boundary layer (Tsai et al. 2005). For an upward heat flux, the temperature within the elongated streaks is cooler, indicative of downwelling flows underneath the regions. This results in accompanying upwelling flows between the cooler streaks with higher gas-flux density at the water surface. The capillary ripples are also observed to enhance gas flux (see the streamwise fluctuation in the distribution of flux density in Figure 1b) as revealed in the previous studies (e.g., Hasse and Liss 1980; Jähne et al. 1987). The contribution by the capillary ripples, however, is not as significant as that by the longitudinal eddies. Despite the different surface conditions, gas-flux density at the water surface $j_s$ exhibits very similar distribution pattern to that of the surface temperature $\Theta_s$.

At the latter time, the spacing between the elongated streaks increases and the cores of counter-rotating vortex pairs expand as depicted in Figure 1c and d. It is postulated that these elongated streaks with wider spacing are caused by the Langmuir circulations associated with wave-current interaction. Approximately, three pairs of Langmuir cells are observed in Figure 1c and d. The upwelling flows between the pairs of Langmuir cells can be deduced from the elongated distributions of cross-wind velocity with alternating directions at the water surface. Within the diverging region, the surface water is warmer and the gas flux is enhanced significantly. A distinct feature of these elongated streaks is that the characteristic signature becomes discontinued or less intense near the crest of the carrier wave, indicating the effect of the orbital velocity field of the carrier wave on the transfer processes.
3.2 Correlations of gas flux with surface properties

To further examine the evolutions of these surface properties with the development of surface wave, and to explore the potential correlation among various mean surface quantities, temporal variations of the total gas flux $\langle j \rangle$, the
mean surface temperature $\theta_s$, the root-mean-square wave slope $\nabla_s \eta$, and the root-mean-square velocity divergence $\nabla_s \cdot \mathbf{u}_s$ are shown in Figure 2, where $\langle \cdot \rangle$ represents averaging over the water surface. Note that the results of the first two wave periods are still in the spinning-up stage of the simulation.

Both the mean gas flux and the surface temperature increase with time to maintain a constant surface concentration and a constant upward heat flux. Uplifts of gas flux are observed within the intervals $t \approx 2T_0$ to $5T_0$ and $12T_0$ to $16T_0$. In
contradiction to the previous studies, no obvious dependence on either the mean-
-square wave slope or the mean-square surface-velocity divergence of the gas flux
is observed in the present numerical simulation.

To quantify the correlation between the distribution of gas-flux density and
other surface parameters, temporal evolutions of the cross-correlation coefficients,
\[ C(j_s; \theta_s), C(j_s; \| \nabla \cdot \eta \|) \] and \( C(j_s; \| \nabla \cdot \mathbf{u} \|) \) are shown in Figure 3, where
\[ C(f; g) = \langle f \times g \rangle \left( \langle f^2 \rangle^{1/2} \langle g^2 \rangle^{1/2} \right)^{-1}. \] Through the entire period of wave development, the gas-flux density highly correlates with the surface temperature \( (C(j_s; \theta_s) \approx 90\%) \). Whereas, the correlation between the gas-flux density and the square wave slope remains to be about 70%; and only a low correlation between the gas-flux density and the surface-velocity divergence \( (\approx 50\% \text{ to } 60\%) \) is observed.

4. Concluding remarks

In this study, we have identified the characteristic distribution of gas-flux
density, which contributes to the major gas transfer at the water surface, and also
revealed the corresponding underlying coherent vortical flows that induce the
surface signature. The detailed mechanisms, which cause these dominant flows,
however, remain unclear. For example, pairs of counter-rotating longitudinal
vortices are observed to form the elongated streaky signatures of gas-flux density
and surface temperature in both the early and later stages of the flow evolution. It,
however, still needs to be verified that the surface streaks with narrower spacing
appears in the early stage of the simulation are associated with the coherent
vortices within a turbulent boundary layer; whereas the elongated streaks with
wider spacing observed later are caused by the well known Langmuir circulations
arising from the nonlinear interaction between surfaces wave and drift current.

Another unresolved issue is the impact of parasitic capillary wavelets on the
transfer process near the water surface. There are laboratory experimental
evidences (e.g., Jähne et al. 1987) suggesting that the transfer of low-solubility
gases across air-water interface is strongly enhanced by the presence of capillary
wavelets. Our simulation result, however, shows only small enhancement in gas
flux by the parasitic capillaries, in comparison with that caused by the coherent
longitudinal eddies. The presence of parasitic capillaries on the carrier gravity
wave has two counter effects on the transfer process near the water surface. The
increase in velocity straining and vorticity production underneath the capillary
wave train can potentially enhance near-surface transfer (Hung and Tsai 2009). On
the other hand, the increase in velocity straining also enhances energy dissipation
within the carrier wave, and attenuates the near-surface velocity field of the carrier
gravity wave (Tsai and Hung 2010). This may leads to reduction in transfer rate.

These are issues under current investigation.
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