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Lengthscales of motions that control air–water gas transfer in grid-stirred turbulence

H. Tsumori *, Y. Sugihara

Department of Earth System Science and Technology, Kyushu University, 6-1 Kasuga-koen, Kasuga 816-8580, Japan

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Abstract

The relationship between the gas transfer velocity and turbulent lengthscales is investigated experimentally in a grid-stirred turbulent flow. The horizontal velocity field at the water surface is measured using particle image velocimetry (PIV). The gas transfer velocity for oxygen is obtained through reaeration experiments. In addition, the gas transfer process by surface-renewal eddies is visualized using laser-induced fluorescence (LIF) technique, in which carbon dioxide is used as the tracer gas. The definition of the Taylor microscale holds that the root-mean-square (RMS) of the surface divergence is expressed by the square root of the turbulent kinetic energy divided by the Taylor microscale. Experimentally obtained data support this scaling. They show the gas transfer velocity to be proportional to the square root of the RMS of the surface divergence. These experimental results imply that the Taylor microscale is an important parameter for gas transfer velocity at the air–water interface. These relations indicate that a nondimensional gas transfer velocity is proportional to the $-1/4$ power of a turbulent-macroscale Reynolds number, which is similar to a small-eddy model, assuming that turbulent eddies with the Kolmogorov scale control the gas transfer process. However, this Reynolds number dependence does not necessarily mean the superiority of turbulent eddies with the Kolmogorov scale in the gas transfer. The LIF visualizations in horizontal and vertical planes close to the air–water interface indicate that the horizontal CO_2-concentration field has a fine spatial pattern, which resembles that of the surface divergence field, and that surface-renewal motions observed in the vertical plane have a larger lengthscale than the Kolmogorov scale. We infer from both PIV and LIF results that the Taylor microscale is an important lengthscale for air–water gas transfer.

Keywords: Gas transfer velocity; Surface divergence; Free-surface turbulence; Taylor microscale; Surface renewal; Particle image velocimetry; Laser-induced fluorescence

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1. Introduction

Air–water gas transfer is an important process in various fields such as chemical engineering, geophysical and environmental sciences. For low-diffusivity gases in water, such as carbon dioxide (CO_2) and oxygen (O_2), gas transfer across the air–water interface is controlled by resistance in the water. The concentration boundary layer on the water side is formed very close to the air–water interface. This boundary layer is peeled and the interface is renewed by turbulent eddies near the interface. Based on such turbulent processes, various models for computation of gas transfer velocity have been proposed to date (e.g., Danckwerts, 1951; Fortescue and Pearson, 1967; Theofanous et al., 1976; McCready et al., 1986). However, a universal model has not been developed because of the difficulty in understanding the turbulent structure close to the interface.

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* Corresponding author.
E-mail address: tsumori@esst.kyushu-u.ac.jp (H. Tsumori).

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Free-surface divergence is closely associated with surface-renewal motions. It is believed to be a control parameter of gas transfer. Based on surface divergence, various experiments and models have been produced for gas transfer velocity (e.g., McCready et al., 1986; Banerjee et al., 2004; McKenna and McGillis, 2004). Surface divergence β is expressed using the following continuity equation at the air–water interface:

$$\beta(x,y,t) = \left(\frac{\partial u}{\partial x}+\frac{\partial v}{\partial y}\right) = -\frac{\partial w}{\partial z}, \tag{1}$$

where u and v are the horizontal velocities at the interface in the directions of the horizontal coordinate axes x and y, respectively, and w the vertical velocity in the direction of the vertical coordinate axis z taken downward from the interface. In a region of small z, the concentration boundary layer becomes much thinner than the scale of turbulent eddies near the interface. For that reason, we can determine the vertical velocity w from the surface divergence β, i.e.,

$$w = -\beta z. \tag{2}$$

At a stagnation flow point near the air–water interface, the horizontal velocities are thought to become small. Therefore, using Eq. (2), we can approximate the convection–diffusion equation for the gas concentration C_g dissolved in the water at the point as

$$\frac{\partial C_g}{\partial t} - \beta z\frac{\partial C_g}{\partial z} = D\frac{\partial^2 C_g}{\partial z^2}, \tag{3}$$

where D is the molecular diffusivity of gas in the water. McCready et al. (1986) derived a linear equation for the concentration fluctuation from Eq. (3). On the basis of that equation, they related the concentration fluctuation with the surface divergence. Calculating the turbulent gas flux, as expressed in terms of surface divergence, they obtained the following relation:

$$k_L = C\sqrt{D\ \beta_{rms}}, \tag{4}$$

where k_L stands for the gas transfer velocity on the water side, C a dimensionless coefficient and β_{rms} the root-mean-square (RMS) of the surface divergence. The value of β_{rms} indicates the degree of convergence or divergence (i.e., downward and upward vertical flows) at the air–water interface. It is a physical parameter that denotes the magnitude of surface-renewal motions. The validity of the surface divergence model of Eq. (4) has been confirmed experimentally in various studies (e.g., McCready et al., 1986; Tamburrino and Gulliver, 2002; McKenna and McGillis, 2004). Recently, it has been possible to obtain instantaneous surface velocity fields using particle image velocimetry (PIV). Using it, the surface divergence can be calculated directly. In a grid-stirred turbulent flow, which is a simple turbulent field without the mean flow, Orlins and Gulliver (2002) measured surface velocity fields with a PIV method and reported that the surface divergence fluctuates more rapidly and has a smaller spatial scale than the vorticity. McKenna and McGillis (2004) also measured surface velocity fields in a grid-stirred turbulence using PIV and the gas transfer velocity based on a reaeration experiment for O_2. They found a linear relation between the gas transfer velocity and the square root of the RMS of the surface divergence. They concluded the validity and importance of the surface divergence model.

However, the relation between β_{rms} and characteristics of free-surface turbulence has not been sufficiently clarified. Particularly, the characteristic lengthscale of β_{rms} is important for development of a gas-transfer velocity model. For example, the dominance of either a large-eddy model proposed by Fortescue and Pearson (1967) or a small-eddy model proposed by Lamont and Scott (1970) has been discussed. Even in an oscillating-grid turbulence, Chu and Jirka (1992) supported the large-eddy model, whereas Asher and Pankow (1986) did the small-eddy model. Although previous studies have mainly examined the relation between statistical quantities of turbulence and the gas transfer velocity, such an analysis seems to present some limitations when we inspect the lengthscale of turbulent eddies, which play an important role in the gas transfer process.

Direct observations of the gas absorption process at the air–water interface provide useful information for characterization of turbulent eddies. In particular, visualization using laser-induced fluorescence (LIF) technique elucidates details of the concentration field close to the air–water interface and surface-renewal motions because of turbulent eddies. An LIF technique based on oxygen quenching of fluorescence by pyrene butyric acid has been used to measure the concentration profiles of dissolved oxygen in water (e.g., Wolff and Hanratty, 1994; Woodrow and Duke, 2001). Herlina and Jirka (2004) investigated the concentration field of oxygen near the air–water interface in a grid-stirred tank using the oxygen-quenching method. Using LIF visualization, they analyzed scales of turbulent eddies, which drive surface-renewal motions. Münsterer and Jähne (1998) investigated vertical profiles of the HCl concentration on the water side close to the air–water interface using LIF technique in a wind-wave tank. They showed that profiles of the mean concentration are explainable using a surface-renewal model with the Schmidt number exponent of 2/3, though these deviate from the prediction based on the small-eddy model. On the basis of those experimental results, they concluded