Estimating the Air-Water Gas Transfer Velocity during Low Wind Conditions

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ABSTRACT
The abundances of atmospheric carbon dioxide, CO$_2$, and methane, CH$_4$, are increasing. These increases affect e.g., the global carbon cycle and the climate both regionally and globally. To better understand the present and future atmospheric CO$_2$ and CH$_4$ concentrations and their climate impact, the gas exchange between water and the atmosphere is important. This exchange can occur in two directions. Oceans take up approximately one third of the anthropogenic CO$_2$ release (the ocean carbon sink). At the same time coastal waters and inland waters emit large amounts of CO$_2$ and CH$_4$, altogether corresponding to a similar amount as the ocean sink.

The interfacial gas-flux for CO$_2$ and CH$_4$ is controlled by the water-side. The gas-flux, $F_g$, is for such gases typically estimated as $F_g = k_g(C_{wb} - \theta C_{as})$ where $k_g$ is the gas transfer velocity, $C_{wb}$ and $C_{as}$ are the gas concentrations in the water bulk and in the air at the surface, and $\theta$ is the dimensionless Ostwald solubility coefficient. The subject of this thesis is to describe and estimate $k_g$ for gases that have a water-side controlled gas-flux (e.g., CO$_2$, and CH$_4$). Besides being important for the geophysical sciences, $k_g$ is also used to design and optimize many applications in e.g., chemical and environmental engineering.

The transfer velocity is influenced by interfacial shear stress from wind, natural convection due to surface heat flux, microscale breaking waves at moderate wind speeds, breaking waves at high wind speeds, bubbles, surfactants, and rain. This thesis focuses on the low wind condition where the forcings due to shear stress, natural convection, and surfactants are important. The relative importance of buoyancy and shear forcing is characterized via a Richardson number $Ri = Bv/u^4$. Here $B$, $v$, and $u^*$ are the buoyancy flux, kinematic viscosity, and friction velocity, respectively. The thesis summarizes three papers where $k_g$ has been studied numerically with direct numerical simulations (DNS) and one paper where field observations have been used.

The results from the field measurements show close relationships for the method using flux-chambers and the parameterization using the rate of turbulent kinetic energy dissipation, and the quantities surface rms velocity and the significant wave height. A parameterization of area-integrated values of $k_g$ from wave measurements was proposed.

The DNS comprise flow conditions ranging from convection-dominated to shear-dominated cases. The results are used to: (i) evaluate different parameterizations of the air-water gas-exchange, (ii) determine, for a given buoyancy flux, the wind speed at which gas transfer becomes primarily shear driven, (iii) find an expression for the gas-transfer velocity for flows driven by both convection and shear, and (iv) investigate the influence of surfactants on gas transfer velocity.

Parameterizations using either the rate of turbulent kinetic energy dissipation or the horizontal surface flow-divergence show a larger disadvantageous dependence on the type of forcing than the parameterization using the surface-normal heat-flux. Two parameterizations using the wind-speed above the surface give reasonable estimates for the transfer-velocity, depending however on the surface heat-flux. The transition from convection- to shear-dominated gas-transfer-velocity is shown to be at $Ri \approx 0.004$. This means that buoyancy fluxes in natural conditions are not important for gas exchange at wind velocities $U_{10}$ above approximately 3 ms$^{-1}$. Below this wind speed the buoyancy fluxes should be taken into account.

The transfer velocity is shown to be well represented by two different approaches: (i) Additive forcing as $k_{g,sum} = A_{shear} u_* (Ri/Ri_c + 1)^{1/4}Sc^{-n}$, where $Ri_c = (A_{shear}/A_{Buoy})^4$ is a critical Richardson number, and (ii) either buoyancy or shear-stress forcing that gives $k_g = A_{Buoy} (Bv)^{1/4}Sc^{-n}$ for $Ri > Ri_c$ and $k_g = A_{shear} u_* Sc^{-n}$ for $Ri < Ri_c$. Here $A_{Buoy} = 0.4$ and $A_{shear} = 0.1$ are constants, $Sc = v/D$ is the Schmidt number, $D$ is the gas diffusivity in water, and $n$ is an exponent that depends on the water-surface characteristics.

Key words: air-sea gas exchange, turbulence, heat flux, natural convection, shear, direct numerical simulations, gas transfer velocity, IR, flux-chambers