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Estimating air-water gas transfer velocity during low wind condition with and without buoyancy

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The increasing abundance of atmospheric carbon dioxide, CO_2, and methane, CH_4, affects the global carbon cycle as well as the climate both regionally and globally. Understanding of the air-water gas exchange and its temporal and spatial distribution is therefore of both regional and global importance. Gas-transfer occurs in both directions across the interface where the oceans take up atmospheric $CO_{2,}$ whereas the coastal areas and inland waters emit CO_2 and CH_4 estimated as yearly and spatial averages. The interfacial gas-flux of CO_2 and CH_4, controlled by the water side is typically estimated as $F_g = k_g(C_{wb} - \vartheta C_{as})$. Here k_g is the gas transfer velocity, C_{wb} and C_{as} are the gas concentration in the water bulk and in the air at the surface respectively, and ϑ is the dimensionless Ostwald solubility coefficient. This study focuses on low wind conditions where the gas transfer typically is increased by surface shear (from wind) and natural convection, and is attenuated by the presence of surfactants. Direct Numerical Simulations (DNS) are used to study the micro-scale turbulence close to the surface and to find the relative importance of buoyancy and shear forcing characterized via a Richardson number $= Bv/u_*^4$. Here B is the buoyancy flux, v is the kinematic viscosity, and u_* is the friction velocity.

It is shown that the transition between convection- to shear-dominated gas transfer velocity is at $Ri \approx 0.004$ meaning that the buoyancy flux during natural conditions isn't important for gas transfer velocity (or gas exchange) at wind velocities U_{10} above approximately 3 ms^{-1}. It is further shown that the gas transfer velocity can be represented by either of two different approaches: (i) <u>Additive</u> 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, or (ii) <u>either</u> buoyancy driven for $Ri > Ri_c$ as $k_g = A_{Buoy}(B\nu)^{1/4} Sc^{-n}$ or shear-stress driven for $Ri > Ri_c$ as $k_g = A_{Shear} u_* Sc^{-n}$. Here A_{Buoy} and A_{Shear} are constants, $Sc = \nu/D$ is the Schmidt number, D is the gas diffusivity in water, and n is an exponent depending on the water-surface characteristics.