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

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Aim

This work aims at refining the gas transfer velocity parameterization during low wind conditions with and without buoyancy, to be used in regional and global climate models. This parameterization and the enhanced understanding of the small-scale processes present in the vicinity of the air-water interfaces can of course be used for other purposes as well such as chemical and environmental engineering.

Main findings

The relative importance of buoyancy and shear forcing is characterized via a Richardson number $Ri = Bv/u_*^4$. Here B, ν, and u_* are the buoyancy flux, kinematic viscosity, and friction velocity, respectively. 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

$$\mathbf{k_g = A_{Shear} u_* (Ri/Ri_c + 1)^{1/4} Sc^{-n}} \qquad (1)^1$$

where $Ri_c = \left(A_{Shear}/A_{Buoy}\right)^4$ is a critical Richardson number and $A_{Shear} = 0.1$ is the transfer velocity coefficient for shear-stress forcing. Here $A_{Buoy} = 0.4$ and $A_{Shear} = 0.1$ are constants, $Sc = \nu/D$ is the Schmidt number, D is the gas diffusivity in water, and n is an exponent that depends on the water-surface characteristics.

Background

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.

Available gas transfer velocity parameterizations show mutual large variability for low wind conditions and are often given as functions of the mean wind velocity U_{10}, at a height 10 m above the water surface, only and do usually not consider the influence of buoyancy flux (as a result of vertical heat flux).

A positive (negative) buoyancy flux due to a heat flux out of the water increase (decrease) the gas transfer velocity and mixing due to destabilization (stabilization) of the water in the vicinity of the surface.

Results and conclusions

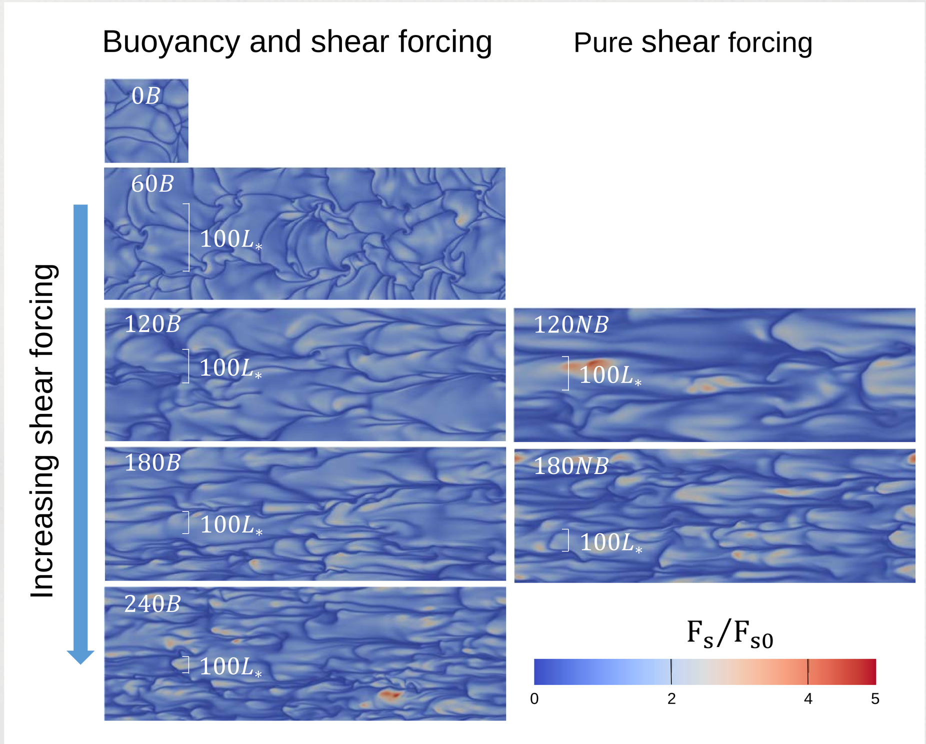

Figure 1. Normalized surface-normal scalar flux fields. The cases are named as the $Re_* = (u_* \nu)/H$ and B for Buoyancy and NB for No-Buoyancy. The same scaling is used for all subplots. The length scale $100L_*$, where $L_* = \nu/u_*$, is indicated in the subplots for cases with $u_* > 0$.

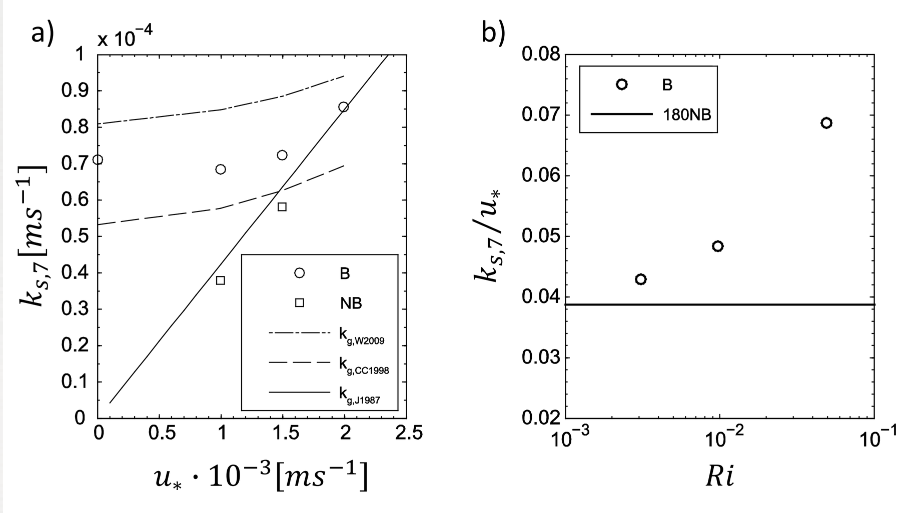

Figure 2. a) The scalar transfer velocity $k_{s,Sc} = k_{s,Sc}/\Delta s_{s,Sc}$. Circles denote cases with buoyancy and squares denote no-buoyancy cases. The dashed and dash-dotted lines denote the wind parameterizations with $n = 1/2$ according to equations (3) and (4) respectively. The solid line denotes a linear increase of the transfer velocity as a function of the friction velocity (2). b) Transfer velocity constant $k_{s,7}/u_*$ as a function of Ri.

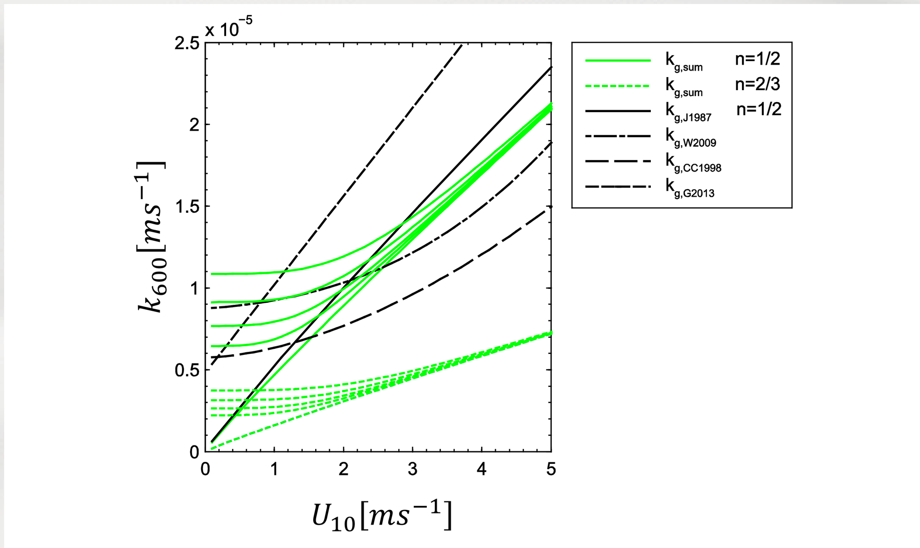

Figure 3. Transfer velocity constant $k_{g,600}$ according to equation (23) in green for $Q_0 = 0, 50, 100, 200$ and $400\ Wm^{-2}$ and $n = 1/2$ (clean) and dotted line $n = 2/3$ (saturated surfactant). The parametrizations in equations (2-5) are given for reference. The transfer velocity estimated with equation (4) is transformed into $Sc = 600$ using $k_{g,Sc_1} = k_{g,Sc_2}(Sc_1/Sc_2)^{-n}$ and $n = 1/2$.

1 The flow pattern for buoyancy driven flows is characterized by thin descending plumes of cold dense water, warm wider ascending plumes, and occasionally surface-normal vortices. The surface normal scalar flux follows this pattern. It is seen that once the shear stress is applied to the surface (in the x-direction towards right in the figures), the pattern and vortices start to be bended and stretched and a fish-scale pattern becomes visible.

Wall-bounded flows have been shown to typically create streaky structures in the vicinity of a wall with a spanwise spacing of about $100L_*$, where $L_* = \nu/u_*$. It can here be seen that these coherent structures typically are finer with than without buoyancy comparing $120B$ with $120NB$ and $180B$ with $180NB$. The scalar flux variation is increasing with increasing shear forcing and the variation is higher for pure shear forcing than for combined forcing. These difference between cases with pure shear and combined forcing decrease with increasing shear-stress (increasing Re_*), indicating that the buoyancy forcing becomes less important.

2 The scalar transfer velocities $k_{s,7}$ $(Sc = 7)$ increase linearly with u_* for cases with pure shear-stress forcing. These results are close to the measurements of gas transfer velocities in a wind tank (1). given in the same figure. Combined forcing gives on the other hand a more or less constant $k_{s,7}$ for low u_*, and then $k_{s,7}$ seems to connect to the linear trend as u_* increases. Another way of expressing this can be seen in b) where $k_{s,7}/u_*$ as a function of Ri is presented. Here $k_{s,7}/u_*$ is declining down to a limiting magnitude for decreasing Ri. This limiting magnitude is set by the no-buoyancy cases. A Richardson number $Ri \approx 0.004$ is found to express the conditions when the scalar transfer starts to change from being dominated by buoyancy forcing to shear-stress forcing which is relevant for determining the buoyancy influence.

3 The parameterization in equation (1) is here plotted for surface heat fluxes in the range of $0 < Q_0 < 400\ Wm^{-2}$. Here the buoyancy flux influences the gas-transfer velocity up to approximately 2- 4 ms^{-1}.

It is seen that the parametrization (3-5), however not explicitly, most likely implicitly account for some background heat flux. For low wind conditions it is though advisable to take the buoyancy (heat) flux into account as is done in equation (1).

The large influence of surfactants on the gas flux is also seen.

$$k_{J1987} = A_{J1987} u_* Sc^{-n}, A_{J1987} = 8.9^{-1} \qquad (2)^2$$

$$k_{g,W2009,660} = 0.1U_{10} + 0.064U_{10}^2 + 0.011U_{10}^3 + 3 \qquad (3)^3$$

$$k_{g,CC1998,600} = 0.215U_{10}^{1.7} + 2.07 \qquad (4)^4$$

$$k_{g,G2013,600} = (5.4 \cdot 10^{-6})U_{10} + (4.8 \cdot 10^{-6}) \qquad (5)^5$$

Material and methods

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 $F_g = k_g(C_{wb} - \vartheta C_{as})$ 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 ϑ is the dimensionless Ostwald solubility coefficient. 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 work focuses on the low wind condition where the forcings due to shear stress, natural convection, and surfactants are important.

Direct numerical simulations, DNS, are used to study how the turbulence and the gas-transports depend on different flow conditions. The gas is modeled as a passive scalar, s, which can be seen as an inert gas. The flow conditions are varied via (i) different surface boundary conditions for the velocity (including shear and surfactants) and the temperature (surface heat flux), (ii) different depths, and (iii) different molecular diffusivities for the scalar.

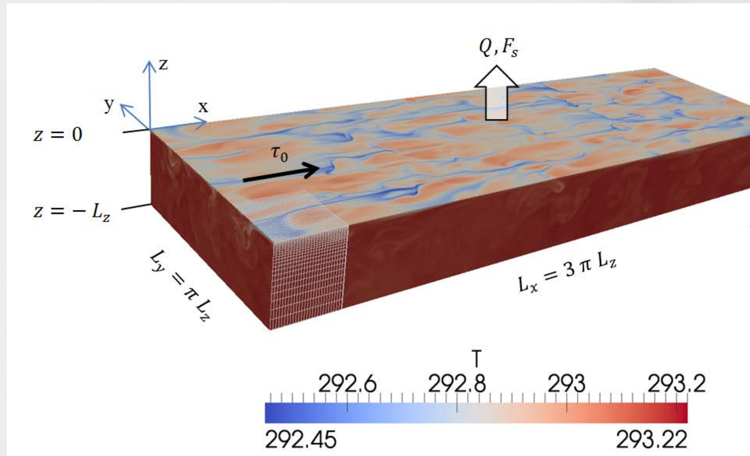

Figure 4. Computational domain for the cases with combined buoyancy and shear stress forcing. The domain size is given by $L_z = 0.1204\ m$, $L_x = 3\pi L_z$ and $L_y = \pi L_z$ in the depth, streamwise, and spanwise direction, respectively. The surface is subject to a constant outward-going heat flux, Q_0, and a constant scalar concentration, S_0, while the bottom is subject to zero flux boundary conditions. The velocity boundary conditions are either slip, no-slip or constant shear stress, τ_0, at the surface boundary and slip at the bottom boundary. Periodic (cyclic) boundary conditions are used for all variables in the horizontal (x- and y-) directions.

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