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NOTES AND CORRESPONDENCE

Comments on "Air–Sea Gas Transfer: Mechanisms and Parameterization"

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1. Introduction

Fluxes of gases like CO_2 across the sea surface constitute an important part of the global climate. However, direct field measurements of these fluxes are problematic and much effort has been put into parameterizations of the mean air–sea gas exchange flux F in terms of air–sea partial pressure differences Δp, gas solubility α, and wind-speed-dependent transfer velocity k (e.g., Liss and Merlivat 1986):

$$F = -k\alpha\Delta p. \tag{1}$$

The transfer velocities are mostly derived from laboratory studies. In a recent paper Wu (1996) attempts to summarize available field data to derive two formulas for k versus wind-speed-dependence discriminating situations found in the open ocean from those in lakes. We differ from Wu (1996) on the following assumptions:

1) The sudden increase of the air–sea gas transfer at low wind speeds due to the abrupt onset of capillary waves that is found in laboratory studies cannot be transferred to the open sea when considering mean fluxes as described by Eq. (1).
2) The bubble-mediated gas transfer is different for different gases because of their different solubility. The flux measurements performed with the dual-tracer method cannot simply be extrapolated to higher wind speeds. Conflicting opinions expressed in the literature are not fully discussed.
3) The description of the air–sea gas exchange, which is a complicated turbulent boundary-layer problem, can no longer be simply based on wind speed alone as done in the past 20 years. The knowledge of

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processes involved allows for more adequate parameterizations today.

2. Sudden increase of the air–sea gas transfer at low wind speeds

Wu (1996) refers to laboratory measurements reporting a rapid increase in the gas transfer rate coincided with the onset of capillary waves on the water surface (Kanwisher 1963; Broecker et al. 1978). After a critical discussion of the laboratory findings Wu (1996) adopts the idea of a sudden change of the gas transfer rate due to the influence of the steep capillary waves on the aqueous molecular sublayer.

The sudden change merely is a reaction to the enhanced roughness modifying the wind field than an effect of the ripples on the molecular aqueous layer. The change of roughness results in an increase of the frictional velocity and hence in a sudden increase of the air–water gas exchange. This is of course not a direct influence of the capillary waves on the air–water gas exchange. Csanady (1990) showed that for the molecular sublayers, the surface under capillary waves still appear to be smooth from the water side. Unless there are substantial divergences occurring in parts of the wavelets as described for the rollers on top of short gravity waves (Csanady 1990), this is hardly a direct effect. Csanady estimated that the capillary waves do not contribute substantially to the convergence in the aqueous molecular sublayer. A more realistic explanation is that the change of surface roughness influences the flow on the air side of the interface.

As seen on the surface, the capillary waves indeed appear suddenly when the wind speed suddenly increases by a gust. However, the wind speed has not only a mean but also a variance that makes the sea surface patchy with respect to the coverage with capillary waves. Some areas are covered by capillary waves, others are not. As the wind speed increases the

area covered by ripples gradually increases so that the surface averaged over a larger area should show a smooth transition from no capillary waves to full coverage without an obvious "jump." This is relevant to the mean gas transfer as described by the usual parameterization (1). The sudden increase should only be observed on a small scale that might be relevant to single surface renewals but not to the mean exchange.

Also, the sudden increase in gas transfer was observed mainly (if not exclusively) in laboratory studies where the natural variance of the wind speed and the implied variance of the surface patches covered with ripples does not occur. This is a difference between the tank airflow and the field wind pattern. It is basically because of the timescale difference in wind velocity fluctuations in the laboratory and in the field conditions.

In this respect it is also worthwhile to mention studies of a related phenomenon—the cool skin on the ocean surface. The thermal (cool skin) and diffusion (gas) sublayers are known to be governed by the similar laws (Soloviev and Schluessel 1994). It is interesting that the laboratory tank measurements of the cool skin (Fedorov and Ginzburg 1992) also show a strong change (decrease) of the temperature difference at onset of the steep capillary waves in the tank. However, there are no reports in the literature about such sudden change of the cool skin parameters during field studies.

3. Bubble-mediated gas transfer and dual-tracer techniques

Another aspect of Wu's (1996) analysis is that he ignores the fact that the bubble-mediated gas transport is different for different gases (because of their different solubility). Hence, the interpretation of the dual-tracer gas flux measurements at medium and high wind speeds cannot be so straightforward as in his analysis. This is especially important in the application of the results to well soluble gases like CO_2. The influence of the bubble mediated gas transfer on the air–sea exchange of CO_2 may start only at very high wind speed conditions (Woolf and Thorpe 1991). So any extrapolation of the dual tracer techniques for the CO_2 should be done very carefully.

4. Parameterization based on known physical processes

The description of air–sea gas exchange is a complicated turbulent boundary-layer problem. A realistic parameterization of the air–sea gas exchange must therefore include a comprehensive analysis of the main physical processes involved. It should also be based on dimensionless dependencies. The pure empiricism presented in Wu (1996) will not help much to further improve the parameterization of the gas transfer, especially in view of the small number of data available.

While during the past 20 years this empiricism was of help to get an early insight into simple bivariate dependencies, more recent studies show that the dynamic processes involved require the inclusion of other variables than the wind speed to successfully parameterize the gas flux at the sea surface. In particular, important processes such as stabilization of the upper ocean by insolation or transition from free convection at calm seas to mechanical turbulence at higher wind speeds are not only a function of wind speed (e.g., Woods 1980; Soloviev and Schluessel 1996). Apart from the limited fetch over lakes, the main difference between ocean and lakes certainly consists in the salinity. This implies processes involving enhanced evaporation at the surface during daytime that leads to an enrichment of the salinity in the upper ocean, which in turn causes increased convection. On the other hand, the negligible salinity in lakes allows for an excessive stabilization during strong insolation that might suppress the gas transfer to a certain extent. Again, these known processes do not depend only on the wind speed. Additional effects that have an impact on the air–sea gas transfer include the cool-skin gas exchange effect due to the temperature dependency of the solubility (Robertson and Watson 1992) and possible irreversible thermodynamics (Phillips 1994; Doney 1995).

5. Conclusions

Quantifying the air–sea gas exchange is a complicated turbulent boundary-layer problem that cannot be solved by simply relating the transfer velocity to only one of many impact variables. While simple k versus wind speed relationships were a first approach at their time, new parameterizations should acknowledge the physical processes involved. It has been shown that the inclusion of processes driving the air–sea gas transfer is necessary and possible and that the simple relationship between transfer velocities and wind speed will fail in many cases.

A serious problem is the absence of sufficient field datasets. Moreover, most of the known datasets are not accompanied by standard meteorological observations, which are important for calculation of the air–sea gas flux under low wind speed conditions (Soloviev and Schluessel 1994). Unfortunately, existing gas-flux measurement techniques are either nonapplicable for the sea conditions or too complicated for collecting statistically representative datasets. The development of techniques for the measurement of gas fluxes at the ocean–air interface is still a challenge. Unless such measurements are performed on a more operational basis significant progress can only be made by including knowledge of the dynamical processes at the air–sea interface in predictive models of the air–sea gas flux.

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