Effect of gas-transfer-velocity parameterization choice on CO$_2$ air–sea fluxes in the North Atlantic and European Arctic

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Abstract

The ocean sink is an important part of the anthropogenic CO₂ budget. Because the terrestrial biosphere is usually treated as a residual, understanding the uncertainties of the net flux into the ocean sink is crucial for understanding the global carbon cycle. One of the sources of uncertainty is the parameterization of CO₂ gas transfer velocity. We used a recently developed software tool, FluxEngine, to calculate monthly net carbon air–sea flux for the extratropical North Atlantic, European Arctic as well as global values (or comparison) using several available parameterizations of gas transfer velocity of different dependence of wind speed, both quadratic and cubic. The aim of the study is to constrain the uncertainty caused by the choice of parameterization in the North Atlantic, a large sink of CO₂ and a region with good measurement coverage, characterized by strong winds. We show that this uncertainty is smaller in the North Atlantic and in the Arctic than globally, within 5 % in the North Atlantic and 4 % in the European Arctic, comparing to 9 % for the World Ocean when restricted to functions with quadratic wind dependence and respectively 42, 40 and 67 % for all studied parameterizations. We propose an explanation of this smaller uncertainty due to the combination of higher than global average wind speeds in the North Atlantic and lack of seasonal changes in the flux direction in most of the region. We also compare the available pCO₂ climatologies (Takahashi and SOCAT) pCO₂ discrepancy in annual flux values of 8 % in the North Atlantic and 19 % in the European Arctic. The seasonal flux changes in the Arctic have inverse seasonal change in both climatologies, caused most probably by insufficient data coverage, especially in winter.

1 Introduction

The region of extratropical North Atlantic, including the European Arctic, is a region where a large part of ocean deep waters are formed (Talley, 2013). This process, part of the global overturning circulation, makes the area a large sink of CO₂ (Takahashi
et al., 2002, 2009; Le Landschützer et al., 2014; Quéré et al., 2015), including its anthropogenic fraction (Orr et al., 2001). Therefore, there is a widespread interest in tracing the changes in North Atlantic carbon fluxes, especially as models predict a decrease in the sink volume later in this century (Halloran et al., 2015).

The trends in North Atlantic CO₂ sink has been intensively studied since observations showed it is decreasing (Lefevre et al., 2004). The sink decrease on interdecadal time scales has been confirmed by further studies (Schuster and Watson, 2007) and continued in recent years north of 40° N (Landschützer et al., 2013). It is not certain how much of this change is due to long-term changes, how much to decadal changes in atmospheric forcing, namely the North Atlantic Oscillation (Gonzalez-Davila et al., 2007; Thomas et al., 2008; Gruber, 2009; Watson et al., 2009) and changes in meridional overturning circulations (Perez et al., 2013). Recent assessments of the Atlantic and Arctic sea–air CO₂ fluxes (Schuster et al., 2013) and global ocean carbon uptake (Wanninkhof et al., 2013) showed that this problem has not been yet resolved.

Studying the volume of the ocean CO₂ sink and especially its trends, one needs to constrain the flux uncertainty. Its sources are sampling coverage, methods of data interpolation, in-water fugacity data quality, the method used for normalization of fugacity data to a reference year in a world of ever increasing atmospheric CO₂ partial pressure and the choice of gas transfer velocity \( k \) parameterization (Takahashi et al., 2009). Although North Atlantic is one of the regions of the world ocean best covered by CO₂ fugacity measurements (Watson et al., 2011), Arctic seas coverage is much poorer, especially in winter (Schuster et al., 2013). The uncertainties in the contemporary global air–sea flux of carbon dioxide have been discussed in two recent papers (Woolf et al., 2015a, b).

One of the factors influencing the value of calculated air–sea gas flux is the choice of formula for gas transfer velocity. Literature of the subject has several parameterizations to choose from with different dependence on wind speed (cubic or quadratic). This problem is not trivial as indicated even by the name of one of the meetings on the topic the COST-735 Action organized “\( k \) conundrum” workshop (in Norwich, Febru-
ary 2008). Its results have been incorporated into a recent review book chapter (Garbe et al., 2014). This paper will concentrate on the very uncertainty caused by the choice of gas flux velocity parameterization in the case of the North Atlantic and European Arctic.

2 Methods

2.1 Datasets

We calculated air–sea carbon dioxide fluxes using a set of processing tools named “Flux Engine” (Shutler et al., 2015), created within European Space Agency founded OceanFlux Greenhouse Gases project (Goddijn-Murphy et al., 2015). The idea beyond the tool development has been to encourage the use of satellite Earth Observation (EO) data for studying air–sea fluxes. Within the Flux Engine, a suit of reanalyses, in situ and model data are available as input to the toolbox and to aid the interpretation of the resultant flux data. There is a choice of several different flux parameterizations as well as input data allowing the generation of monthly global gridded flux products.

For the calculations, we used $pCO_2$ and salinity values from Takahashi et al. (2009) climatology based on more than 3 million measurements of surface water $pCO_2$ in open-ocean environments during non El Nino conditions. For some calculations we used, as an alternative, Surface Ocean CO$_2$ Atlas (SOCAT) ver. 1.5 and 2.0 (Sabine et al., 2013; Pfeil et al., 2013; Bakker and al. 2014) $pCO_2$ and SST data. SOCAT is a community driven dataset containing respectively 6.3 and 10.1 million surface water CO$_2$ fugacity values with a global coverage. Both climatologies were calculated for year 2010. The SST values were taken from Operational Sea Surface Temperature and Sea Ice Analysis (OSTIA) (Donlon et al., 2011), and in the SOCAT case from the database, while SST skin data we used came from ARC/(A)ATSR Global Monthly Sea Surface dataset (Merchant et al., 2012).
We used Earth Observation (EO) wind speed and sea roughness ($\sigma_0$ in Ku band from GlobWave L2P products) data obtained from the European Space Agency (ESA) Environmental monitoring satellite, Envisat. Envisat was launched in 2002 with 10 instruments onboard into sun-synchronous near-polar orbit (SSO) with 35 day repeat cycle. It carries, among others, two atmospheric sensors monitoring trace gases. EO data supports earth science research and allows monitoring of the evolution of environmental and climate change.

All the data were used globally within the FluxEngine software. From the gridded product ($1^\circ \times 1^\circ$) we extracted extratropical North Atlantic (North of 30° N), and its subset, the European Arctic (North of 64° N).

### 2.2 $k$ parameterizations

The flux of CO$_2$ at the interface of air and the sea is controlled by wind speed, sea state, sea surface temperature (SST) and other factors. We estimate the air–sea flux of CO$_2$ (OF, gC m$^{-2}$ day$^{-1}$) as the product of gas transfer velocity ($k$, m s$^{-1}$) and also the difference in CO$_2$ concentration (g m$^{-3}$) at the sea water and the interface–air (Land et al., 2013). The concentration of CO$_2$ in sea water is a product of its solubility ($\alpha$, g m$^{-3}$ µatm) and its fugacity ($f$CO$_2$, µatm). Solubility is, in turn, a function of salinity and temperature. Hence Eq. (1) is represented as:

$$F = k(\alpha_W f CO_{2W} - \alpha_S f CO_{2A})$$  \hspace{1cm} (1)$$

where the subscripts denote values in water (W) and the air–sea interface (S) and in the air (A). We can exchange fugacity to the partial pressure (their values differ by < 0.5 % over the temperature range considered) (McGillis et al., 2001). So Eq. (2) now becomes:

$$F = k(\alpha_W p CO_{2W} - \alpha_S p CO_{2A})$$  \hspace{1cm} (2)$$
We can also ignore the differences between the two solubilities and just use the water side solubility $\alpha_w$. Eq. (3) will be represented as:

$$F = k\alpha_w(p_{CO_{2w}} - p_{CO_{2a}})$$

(3)

This formulation is often taken as the “bulk parametrization”.

In this work we chose to analyze the fluxes using five different parameterizations, within the FluxEngine software, using in terms of wind speed to parametrized $k$. All of them are parameterized with wind speed and differ in the formula for gas transfer velocity, $k$:

$$k = \sqrt{\frac{660.0}{Sc_{skin}}} \cdot (0.212U_{10}^2 + 0.318U_{10})$$

(Nightingale et al., 2000),

(4)

$$k = \sqrt{\frac{660.0}{Sc_{skin}}} \cdot 0.254U_{10}^2$$

(Ho et al., 2006),

(5)

$$k = \sqrt{\frac{660.0}{Sc_{skin}}} \cdot 0.0283U_{10}^3$$

(Wanninkhof and McGillis, 1999),

(6)

$$k = \sqrt{\frac{660.0}{Sc_{skin}}} \cdot 0.251U_{10}^2$$

(Wanninkhof, 2014),

(7)

$$k = \sqrt{\frac{660.0}{Sc_{skin}}} \cdot (3.3 + 0.026U_{10}^3)$$

(McGillis et al., 2001),

(8)

where the subscripts are Schmidt numbers at the skin surface ($Sc_{skin}$), as a function of SST (=[(kinematic viscosity of water)/(diffusion coefficient of CO$_2$ in water)]), 660.0 is the Schmidt number for carbon dioxide at 20°C temperature in seawater, $U_{10}$ is the wind speed above 10 m sea surface.

In addition to the purely wind driven parameterizations, we have used a combined Goddijn-Murphy et al. (2012) and Fangohr and Woolf (2007) parametrization, created as part of OceanFlux GHG project and provided as an option in FluxEngine. This parameterization separates contributions from direct and bubble-mediated gas transfer as suggested by Woolf (2005). Its purpose is to enable separate evaluation of the effect of the two processes on gas fluxes and should be not treated as a final product (one of the aims of the ongoing OceanFlux Evolution project is improving this parameterization). We used these OceanFluxGHG parameterizations in two versions: wind driven and radar backscatter driven.
3 Results

Using the FluxEngine software, we have produced CO₂ global monthly gridded air–sea fluxes and calculated from them the values for the study region, extratropical North Atlantic and separately for its subset, the European Arctic seas. Figure 1 shows maps of average CO₂ fluxes for North Atlantic, calculated with Nightingale et al., 2000 (named further as N2000) $k$ formula and Takahashi (2009) climatology for the whole year and for each season. The area as a whole is a carbon sink but even the all-season map shows that some regions close to North Atlantic Drift and East Greenland Current are net sources. The seasonal maps show even more variability. For example the above mentioned sink areas become sinks in summer (effect of phytoplankton blooms) while the southernmost areas of the study become CO₂ sources in summer and autumn (effect of sea-water temperature changes). Much of this variability is caused by changes of the surface water $pCO₂$ average values, shown in Fig. 2 for the whole year and for each season (and variability in atmospheric CO₂ partial pressure, not shown). However, the flux is proportional to the product of $\Delta pCO₂$ and $k$. In most parameterizations $k$ is a function of wind speed (Eqs. 4–8). The average wind speed $U_{10}$ for the whole year and each season is shown in Fig. 3. The wind speeds in the North Atlantic are higher than the mean in the world ocean, with average values higher than 10 m s$^{-1}$ in many regions of the study area in all seasons except the summer (with highest values in winter). This is important because the fluxes depend not only on average wind speed but also on its distribution (see also the Discussion). This effect is especially visible between formulas with different powers of $U_{10}$. Figure 4 shows the difference in calculated fluxes on the example of two parameterizations: one proportional to $U_{10}^3$ (Eq. 6) and one to $U_{10}^2$ (Eq. 7), namely Wanninkhof and McGillis (1999) and Wanninkhof (2014). It can be seen that the “cubic” function results in higher absolute flux values, comparing to a “quadratic” one, in regions of high winds and lower with weaker winds.

Figure 5 shows the monthly values of CO₂ fluxes for the five parameterizations (Eqs. 4–8) for the North Atlantic and European Arctic. The areas are a sink in ev-
ery month, although August is close to neutral for the North Atlantic. The results using
two cubic formula (Eqs. 6 and 8) are higher in absolute values respectively by 30 % for
Wanninkhof and McGillis (1999) and 55 % for McGillis (2001), comparing to “quadratic”
N2000 (Eq. 4). The other two “quadratic” parameterizations (Eqs. 5 and 7) resulted
in fluxes within 5 % of N2000. Annual fluxes for North Atlantic, European Arctic and
global (for comparison) are shown in Fig. 6. In addition to the five parameterizations,
the figure presents results for both the OceanFluxGHG formulas (using wind and radar
backscatter data). The mean and standard deviations of the parametrization ensemble
are shown as gray vertical lines. Annual North Atlantic flux, depending on the formula
used, varies from \(-0.38\) TgC for N2000 to \(-0.56\) TgC for McGillis et al. (2001). In the
case of global carbon flux the values are, respectively, \(-1.30\) and \(-2.15\) TgC. Figure 7
shows the same data “normalized” to N2000, the parameterization resulting in lower
absolute flux values, to visualize the relative differences. In the case of North Atlantic,
using the “quadratic” Wanninkhof (2014) and Ho et al. (2006) results in a flux respec-
tively 3 and 5 % higher in absolute value than N2000, while the “cubic” Wanninkhof and
McGillis (1999) and McGillis et al. (2001) results in respectively 28 and 42 % higher val-
ues. The respective values for the Arctic are 3, 4 % for quadratic as well as 27 and 40 %
for cubic functions. In the case of global flux the respective values are 8 and 9 % higher
than N2000 flux for the quadratic functions as well as 34 and 67 % for cubic ones. The
OceanFluxGHG parameterization results in fluxes 18 and 32 % higher for North At-
lantic than N2000 (for respectively backscatter and wind driven versions). In the case
of global values this surplus was, respectively, 45 and 52 %.

All the above results used the Takahashi (2009) $p$CO$_2$ climatology. For comparison
we have also calculated fluxes using SOCAT version 1.5 and 2.0. Figure 8 shows the
results using N2000 $k$ parameterization for all the three climatologies. In the case of
the North Atlantic study area, although the monthly values show large differences (both
SOCAT climatologies result in larger sink in summer and smaller in winter comparing to
Takahashi), the annual values are similar: \(-0.38\) TgC for both Takahashi and SOCAT v.1.5 and \(-0.41\) TgC for SOCAT v. 2.0. In the case of European Arctic the situation is
4 Discussion

Our result show that using the three “quadratic” parameterizations (Nightingale et al., 2000; Ho et al., 2006 and Wanninkhof, 2014) results in fluxes values within 5% of each other in the case of North Atlantic. This discrepancy is smaller than the 9% difference for the net global carbon air–sea flux (Fig. 7). This would confirm that, to our knowledge, the parameterizations are interchangeable being all within the experimental uncertainty. This view was supported by the results of a discussion session convened by the leading authors of the three parameterizations (“Relationship between wind speed and gas exchange over the ocean: which parameterization should I use?” during SOLAS Open Science Conference in Kiel, Germany on 7–11 September 2015). The three parameterizations were derived using different methods and data from different regions, namely passive tracers and dual-trace experiments in the North Sea in the case of Nightingale et al. (2000), dual tracers in the Southern Ocean in the case of Ho et al. (2006) and global ocean $^{14}$C inventories in the case of Wanninkhof (2014). This makes it possible to be highly confident that at least average fluxes calculated with the three formulas are close to the unknown true values. However, the uncertainties are still large and although the quadratic functions are supported by several lines of evidence (see Garbe et al. 2014 for discussion), other powers are not completely refuted by the available observations. Therefore it is important to notice that a choice of one of the available cubic functions may lead to net fluxes larger in absolute values up to 33% in the North Atlantic and 50% globally.

The above results imply smaller relative differences between the parameterizations in North Atlantic than globally. This is interesting because North Atlantic is a region of strong winds and in most of its area no seasonal changes in the flux direction (Fig. 1).
We see two reasons for that. First, when comparing quadratic and cubic parameterizations (Fig. 9), it is clear that cubic ones imply higher fluxes for high winds while quadratic one for weak winds. This difference can be presented in arithmetic terms. Let us assume two functions of wind speed \( U \), \( F_1(U) \) quadratic and \( F_2(U) \) cubic:

\[
F_1(U) = aU^2, \quad \text{(9)}
\]

\[
F_2(U) = bU^3. \quad \text{(10)}
\]

The difference between the two functions \( \Delta F \) is equal to

\[
\Delta F = F_2 - F_1 = bU^3 - aU^2 = bU^2(U - ab^{-1}) = bU^2(U - U_x) \quad \text{(11)}
\]

where \( U_x = ab^{-1} \). The difference is positive for wind speeds greater than \( U_x \) and negative for smaller ones. \( U_x \) is the value of wind speed for which the two functions intersect. In the case of Eqs. (6) and (7), \( a = 0.251 \) and \( b = 0.0283 \), implying \( U_x = 8.87 \text{ ms}^{-1} \). In fact all the functions presented in Fig. 9 have very similar values for wind speeds close to 9 ms\(^{-1}\). The value is very close to average wind speeds in the North Atlantic (Fig. 3). This is one of the reasons of small relative net flux differences. The other is lack of seasonality. In case of seasonal changes in the flux direction (caused by seasonal changes in water temperature or primary productivity), with winds stronger than \( U_x \) in some seasons and weaker in other (usually strong winds in winter and weak in summer), the fluxes partly cancel each other while the difference between cubic and quadratic parameterization add to each other due to simultaneous change in the sign of both flux itself and the \( U - U_x \) term. This effect of seasonality has been recognized previously (A. Watson, personal communication, 2015) but we are unaware of any paper explaining it in the terms of arithmetic formulas.

In addition to the five parameterization described above, we calculated fluxes for the OceanFluxGHG combined formula, separating contributions from direct and bubble-mediated gas transfer. The resultant fluxes are higher, in absolute terms, from all the quadratic functions considered in this study, being closer to cubic ones. This may mean
that the bubble mediated term of Fangohr and Woolf (2007) may be an overestimation for CO$_2$ fluxes. This question will be the subject of further studies in the OceanFlux Evolution project.

Although using both the Takahashi and SOCAT $p$CO$_2$ climatologies (Fig. 8) results in similar annual fluxes in the North Atlantic, it should be noted that they show different seasonal changes. This may have been caused by slightly different time periods of the climatologies (SOCAT is more recent). The difference is much larger in the European Arctic due to much worse data spatial coverage and possible interpolation artifacts (Goddijn-Murphy et al., 2015). This discrepancy makes us treat the flux results from the Arctic with much less confidence than the ones for the whole North Atlantic. This situation may improve after SOCAT v.3 which is planned to be released in 2016.

5 Conclusions

In this paper we have studied the effect of choice of gas transfer velocity parameterization on the net CO$_2$ air–sea gas flux volume in the North Atlantic and European Arctic using the recently developed FluxEngine software. The results show that the uncertainty caused by the choice of the $k$ formula is smaller in the North Atlantic and in the Arctic than globally. The annual net flux difference caused by the choice of parameterization is within 5% in the North Atlantic and 4% in the European Arctic, comparing to 9% globally for the studied functions with quadratic wind dependence and respectively 42% for North Atlantic, 40% for Arctic and 67% between the cubic and quadratic functions. We explain the smaller North Atlantic variability by the combination of higher than global average wind speeds in the North Atlantic, closer to 9 m s$^{-1}$, the wind speed when most $k$ parameterization have similar values and the all-season CO$_2$ sink conditions in most North Atlantic areas. We compare the Takahashi and SOCAT $p$CO$_2$ climatologies finding that although the seasonal variability in the North Atlantic is different, annual net fluxes are within 8% in the North Atlantic and 19% in the European Arctic. The seasonal flux changes in the Arctic have inverse seasonal change in both
climatologies indicating possible under sampling and therefore the need for more polar $pCO_2$ data before than available at present.

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References


Constraining North Atlantic and Arctic CO$_2$ fluxes

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Figure 1. Seasonal and annual mean air–sea fluxes of CO$_2$ (g C m$^{-2}$ day$^{-1}$) in the North Atlantic, combine using Nightingale et al. (2000) $k$ parameterization and Takahashi (2009) climatology in (a) annual, (b) DJF (winter), (c) MAM (spring), (d) JJA (summer), (e) SON (autumn).
Figure 2. Seasonal and annual $p$CO$_2$ values (µatm) in surface waters of the North Atlantic, estimated using Takahashi (2009) climatology in (a) annual, (b) DJF (winter), (c) MAM (spring), (d) JJA (summer), (e) SON (autumn).
Figure 3. Wind speed distribution $U_{10}$ (ms$^{-2}$) in the North Atlantic used to determine the relationship between gas transfer velocity and air–sea CO$_2$ fluxes in (a) annual, (b) DJF (winter), (c) MAM (spring), (d) JJA (summer), (e) SON (autumn).
Figure 4. Differences maps for the air–sea CO₂ fluxes (g C m⁻² day⁻¹) in the North Atlantic, between a wind cubed and squared parameterizations (Wanninkhof and McGillis 1999 and Wanninkhof 2014) in (a) annual, (b) DJF (winter), (c) MAM (spring), (d) JJA (summer) (e) SON (autumn).
Figure 5. Monthly values air–sea fluxes of CO$_2$ (gC m$^{-2}$ day$^{-1}$) for the five parameterizations (Eqs. 4–8) in (a) North Atlantic, (b) European Arctic.
Figure 6. Annual air–sea fluxes of CO$_2$ (g C m$^{-2}$ day$^{-1}$) for the five (Eqs. 4–8) parameterizations as well as for backscatter (default) and wind driven OceanFluxGHG parameterization (see text) in (a) global, (b) North Atlantic (c) European Arctic. Average values for all parameterization and standard deviations are marked as vertical gray lines.
**Figure 7.** Annual air–sea fluxes of CO$_2$ (g C m$^{-2}$ day$^{-1}$) for the five (Eqs. 4–8) parameterizations as well as for backscatter (default) and wind driven OceanFluxGHG parameterization normalized to flux values of Nightingale et al. (2000) $k$ parameterization.
Figure 8. Comparison of monthly values fluxes of air–sea CO$_2$ fluxes calculated with different $p$CO$_2$ climatologies (Takahashi et al., 2009, SOCAT v. 1.5 and 2.0) using the same $k$ parameterization (Nightingale et al., 2000) in (a) North Atlantic, (b) European Arctic.
Figure 9. Different k660 parameterizations as a function of wind speed.