We are grateful for the Reviewer #3 for all relevant comments and suggestions and we want to say that it wasn't our intention to the reviewer felt that his/her comments and remarks are not important to us and ignored by us. We also want to say that every comments and remarks of the reviewers were thoroughly and carefully examined, all were very useful and helpful to us make better our work. Any comments, we tried to adjust and respond to it and never wanted to any reviewer felt overlooked in his/her constructive criticism. Sorry, if as a result of misunderstanding each other you felt ignored in your comments. We try to do everything we can to improve our article to be useful and meet the requirements of the scientific article on highest level. Below the authors refer to the individual comments and specify changes that were made to the text as well as marked up version of manuscript where Topic Editor and Reviewer can see all corrections.

The authors have addressed some (but not all) of my major comments in the previous round, albeit not satisfactorily. E.g., in addressing my suggestion to include CMIP5 models, the authors provided rebuttal, that the analysis would be "burdensome and actually pointless".

I have suggested to explore how the uncertainty of CO2 fluxes attributed by the wind speed parameterization may evolve in the future. They responded that "it has been done and results are published" by showing Fig. 12.19 of the IPCC-AR5. I don't understand this argument. The figure is completely different than what I suggested. Further, based on this figure, they claimed that "This is not anyone would call robust prediction". In fact, the figure indicates nothing about the robustness of the model predictions. It indicates that the inter-models mean projects a small future changes, which in most cases are still within the range of internal variability. Earlier comments: I think the authors should also consider studying how this uncertainty would evolve in the future, regionally and temporally. Questions such as will uncertainty due to atmospheric CO2 concentration dominates the uncertainty due to wind speed formulation, etc. has not been studied previously and can be addressed here.

We do agree but the questions are much beyond the scope of this paper.

Nevertheless, I understand that for those not familiar with processing CMIP5 models, this task could be overwhelming and time consuming. I do think that my suggestion on analyzing the SOC or other key ocean regions is feasible, and their argument for focusing on the North Atlantic and Arctic regions is not very convincing.

We understand what reviewer wanted us to do, but we are focusing only on the North Atlantic in this paper because that is our region of study. However we decided to calculate also the Southern Ocean fluxes for comparing with the North Atlantic, what adds useful information, as reviewer said. They are now in Table 1, Figure 7 (now 6) and commented on in the manuscript text. We concentrate on the Atlantic Ocean also is the new and interesting find that the spread between different k parameterizations is smaller there than globally

L282 For example in the South Atlantic annual mean of wind speed is within 8.48 m s⁻¹ (Takahashi et al., 2009) and sink of CO_2 (south of 45°) decrease significantly after 1990 with increasing wind speeds what can influence higher concentration of pCO_2 in surface water due to enhance vertical mixing of deep waters and biological activity. (Le Quèrè et al., 2007). Takahashi et al. (2009) also indicate that the flux difference in the Southern Ocean are very strong dependence to the choice of the

The authors also did not respond to my comments on replacing information in Figs. 6 and 7 into a further errors Table. This has led me to identify in the 42% should be 47% Line 237: according to values provided earlier on Line 231, 332: 42% 47% Line should be Line 272: *"50%"* should be "more 50%" than Fig 7b, purple bar does not look correct (i.e. it shows here smaller than the light blue bar, but it is *larger in Fig. 6b)*

Sorry for that. The reviewer did have legitimate concerns here. Now all % are correct as well as color bar in Fig. 6 (old Fig.7). We have used information in Fig. 6 and 7 into a new Table 1 and

updated information inside the manuscript. However, we decided to retain Figure 7 (now 6) because this graphical expression of one of the main conclusions of the paper. We have also corrected all the above the indicated errors.

Figure 7 (now 6) had actually problem that it has been calculated in a different way from that was described (as an average of "Nightingale normalized" monthly fluxes). It has now been corrected. This caused some minor changes to the reported values of differences between the parameterizations (they are now identical to ones in Table 1).

L25 42 % -> 46%, 40 % -> 44 %, 67 % -> 65 %

L239 3% -> 4%, 42% -> 44%, 27% -> 28%, 40% -> 44%, 34% -> 33%, 67% -> 65%, 18% -> 38%, 32 % -> 47 %, 45% -> 44%

L350 42% -> 46%, 40% -> 36%, 67% -> 65%

L247 The spread of the Arctic values was lower than the Atlantic ones (see Table 1). On the other hand, the values for the South Ocean were slightly higher than for the North Atlantic but lower than the global ones, with the exception of the OceanFlux GHG.

L309 The spread of flux values for the Southern Ocean seems to support this conclusion, being larger than the North Atlantic one. Southern Ocean has on average stronger winds than North Atlantic (including also the Arctic Seas) which seem to have the smallest spread of flux values for different parameterizations.

The suggestion to have an English native speaker reviewing the manuscript prior to resubmission appears to be ignored as well. Overall, I have the feeling that the authors did not spend sufficient time to grasp the reviewer's suggestions, which in most cases are constructive.

Sorry that you felt in that way. We really spend a lot of time to check our manuscript and, as you suggest, we have one more proofreading right now and we hope that now everything is good. Because the new native speaker was actually the leader of the OceanFlux GHG project (Jamie Shutler), we also gained some (minor) corrections to some details of how FluxEngine works. For example:

L31 The seasonal flux in the Arctic computed from two climatology data sets are opposite to one another, possibly due to insufficient spatial and temporal data coverage, especially in winter.

L84 We calculated net air-sea CO₂ fluxes using a set of software processing tools called the 'FluxEngine' (Shutler et al., 2016), which were created within European Space Agency funded OceanFlux Greenhouse Gases project (http://www.oceanflux-ghg.org).

L87 deleted: The software became publicly available in March 2016 under an open source license, but at the time we started this study we did not have more information about it than is included in the paper describes a set of tools (Shutler et al., 2016). This was a conscious decision because, even as we had access to the toolset developers, we wanted to test it as the end users (this is probably the first study using the toolset by authors who had no part in creating it).

L87 The tools were developed to provide the community with a verified and consistent toolbox and to encourage the use of satellite Earth Observation (EO) data for studying air-sea fluxes. The toolbox source code can be downloaded or alternatively there is a version that can be run through a web interface. Within the online web interface, a suite of reanalysis data products, *in situ* and model data are available as input to the toolbox. These data are freely available for the scientific community to use. The FluxEngine allows you to select several different air-sea flux parameterizations, as well as input data, allowing the generation of the monthly global gridded net air-sea flux products with 1° x 1° spatial resolution. The output consists of twelve NetCDF files (one file per month). Each monthly composite file includes the mean (first order moment), median, standard deviation and the second, third and fourth order moments. There is also information (meta data) about origin of data inputs.

L117 now: We used Earth Observation (EO) wind speed and sea roughness (σ_0 in Ku band from GlobWave L2P products) data obtained from the European Space Agency (ESA). The GlobWave satellite products give a "uniform" set of along track satellite wave data from all available Altimeters (spanning multiple space agencies) and from ESA Synthetic Aperture Radar (SAR) data.

GlobWave Project is an initiative funded by ESA and subsidised by CNES. The aim of the project is to improve the uptake of satellite-derived wind-wave and swell data by the scientific, operational and commercial user communities. This has been achieved by providing a uniform, harmonized, quality controlled, multi-sensor set of satellite wave data. Wave data is collected from both altimeters (ERS-1, ERS-2, ENVISAT, Topex/POSEIDON, Jason-1, Jason-2, CryoSAT, GEOSAT and GEOSAT Follow On) and from ESA Synthetic Aperture Radar (SAR) missions, namely ERS-1, ERS-2 and ENVISAT. All data come in netCDF-3 format.

L211 The mean wind speed...

L336 It is impossible to declare within this study which dataset is more accurate as only new data can settle this. However, such data have been recently published (Yasunaka et al., 2016). The observed in-water pCO₂ data (Fig. 3 in Yasunaka et al. 2016), especially since 2005, show clearly an annual cycle compatible with the SOCAT seasonal flux variability.

This is also evident from the authors' response to Reviewer#2 comments (In my initial review, I made a comment about Figure 7 as well as Figure 8. The authors misunderstood. Point about fig 7 - This figure does not add anything over Figure 6. Why do you need both figures? Point about fig 8 - This is an interesting result and should be described in more detail. It is fine that the authors think this is outside of the scope of the current manuscript. I just want to make it clear that there were 2 seaparate comments. Here I would like the authors to address only the comment about Figure 7.,) e.g., his/her request to add more discussion to Fig. 8, and NOT to remove it.

L330 Although, using both Takahashi climatology and SOCAT *p*CO₂ climatology (Fig. 7) result in similar annual net air-sea fluxes in the North Atlantic, it should be noted that they show different seasonal variations. This may have been caused by slightly different time periods of the datasets (i.e. the SOCAT based climatology contains more recent data). The difference is much larger in the European Arctic due to the underlying sparse data coverage and possible interpolation artifacts (Goddijn-Murphy et al., 2015). This discrepancy makes us treat the net air-sea CO₂ flux results from the Arctic with much less confidence than the values for the whole North Atlantic. It is impossible to declare within this study which dataset is more accurate as only new data can settle this. However, such data have been recently published (Yasunaka et al., 2016). The observed inwater pCO₂ data (Fig. 3 in Yasunaka et al. 2016), especially since 2005, show clearly an annual cycle compatible with the SOCAT seasonal flux variability.

L588 Yasunaka, S, A. Murata, E. Watanabe, M. Chierici, A. Fransson, S. van Heuven, M. Hoppema, M. Ishii, T. Johannessen, N. Kosugi, S. K. Lauvset, J. T. Mathis, S. Nishino, A. M. Omar, A. Olsen, D. Sasano, T. Takahashi, R. Wanninkhof.,: Mapping of the air—sea CO2 flux in the Arctic Ocean and its adjacent seas: Basin-wide distribution and seasonal to interannual variability, doi:10.1016/j.polar.2016.03.006, 2016

More minor errors identified:

L115-118: I suggest to clarify the sentence, e.g., by splitting into two sentences.

Line 129: 130, lower case north

Line 137: OF should be F?

L140: unit of alpha should be: g 'C'..unit: gm⁻³ µatm⁻¹ is correct

L147: So equation (1) now becomes ...

L152: Equation(2) can also be represented as:

L156: referred to as the ..

L275-276: I believe the Southern Ocean, a region with stronger surface wind, could contribute to the global differences and should be discussed here.

All done. We addressed all the points above. The SO data are now in Table 1 and additional SO panels have been added to Figures 7 (now 6).

L131 add: For comparison, we also calculated fluxes in the Southern Ocean (south of 40° S).

L247 On the other hand, the values for the South Ocean were slightly higher than for the North

Atlantic but lower than the global ones, with the exception of the OceanFlux GHG parameterizations.

L309 The spread of flux values for the Southern Ocean seems to support this conclusion, being larger than the North Atlantic one. Southern Ocean has on average stronger winds than North Atlantic (including also the Arctic Seas) which seem to have the smallest spread of flux values for different parameterizations.

L388: OceanFlux L300: gas flux

L552 and 557: I am not sure what is Ocean Science policy on citing reference materials that are in submission stage.

Units in Figs. 1 and 4 color panels: superscript "-2" and "-1"

Units in Fig. 3 color panel: superscript "-1"

Fig. 3 caption: unit is wrong. (also in L572)

Fig. 5 caption and y-axes label: units should be Pg C/month, (also L583)

All mistakes were corrected. Thank you for your help and corrections.

Effect of gas-transfer_-velocity parameterization choice on CO₂ air-sea fluxes in the North Atlantic and the European Arctic

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Abstract

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The oceanic sink of carbon dioxide (CO₂) is an important part of the global carbon budget. Understanding uncertainties in the calculation of this net flux into the ocean is crucial for-climate research comprehension the global carbon cycle. One of the sources of the uncertainty within this calculation is the parameterization chosen for the CO₂ gas transfer velocity. We used a recently developed software toolbox, called the FluxEngine, in order to estimate the monthly net carbon airsea flux for the extratropical North Atlantic, the European Arctic, and well as globally values (in order to compare) using several published available quadratic and cubic wind speed parameterizations of the gas transfer velocity for different 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. This region, which is considered a large oceanic sink of CO₂-, and it is also a region often characterised by strong winds but and the region-with a good in situ covering measurement coverage, characterised by strong winds. -We show that this uncertainty is smaller in the North Atlantic and the Arctic than globally. It is as little as within 5% in the North Atlantic and 4% in the European Arctic, in comparisonng to 9% for the global World oOcean when restricted to functions with quadratic wind dependence. Whereas this uncertainty becomes and respectively 462%, 440% and 657% respectively if you consider for all of the studied parameterizations studied. We propose thatan explanation of this smaller uncertainty is caused by a combination of higher than global average wind speeds in the North Atlantic and lack of any seasonal changes in the direction of the flux direction within most of the region. We also compare the impact of using two different availablein situ pCO₂ datasets (Takahashi and SOCAT) within the flux calculation. Differences in these - pCO₂ data in turn cause differences discrepancies in the annual net fluxes values of 8% in the North Atlantic and 19% in the European Arctic.-The seasonal flux in the Arctic computed from two climatology data sets are opposite to one another, possibly due to insufficient spatial and temporal data coverage, especially in winter. Seasonality of the flux changes in the Arctic are opposite to one other in both datasets, most likely caused by insufficient data coverage, especially in winter.

1. Introduction

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

The trends and variations in the North Atlantic CO₂ sink has been intensively studied since

observations have shown it <u>appears to beis</u> decreasing (<u>eLefevre Lefevre</u> et al., 2004). Thise sink decrease <u>onin</u> inter-annual time scales has been confirmed by further studies (Schuster and Watson, 2007) and <u>this trend has</u> continued in recent years North of 40° N (Landschützer et al., 2013). It is not certain how many of these changes are the results of long-term changes, <u>how many of decadal</u> changes in atmospheric forcing, namely the North Atlantic Oscillation (Gonzalez-Davila et al., 2007; Thomas et al., 2008; Gruber 2009; Watson et al., 2009) <u>orand</u> 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 <u>the global ocean net carbon uptake</u> (Wanninkhof et al., 2013) showed that the <u>cause is is still unproblem has not been yet knownresolved</u>.

To sStudying the rate of the ocean CO₂ sink and especially its long-terms trends, one needs to first constrain the total uncertainty in the flux—calculationuncertainty. SIts—sources of uncertainty includeare sampling coverage, the method 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 (Landschützer et al., 2014; Woolf et al., 2015a, 2015b). It has also been identified that the choice of the wind data product provides an additional source of uncertainty (Gregg et al., 2015). In this work we have chosen to analyze various empirical wind driven gas transfer parameterizations using wind speed. Although the North Atlantic is one of the regions of the world ocean best covered by CO₂ fugacity measurements (Watson et al., 2011), the Arctic seas coverage is much poorer, especially in winter (Schuster et al., 2013). It has to be noticed that also the choice of the wind product adds to the uncertainty (Gregg et al., 2015).

One of the factors influencing the value of the calculated air-sea gas flux is the choice of the formula for the gas transfer velocity. Within the lLiterature there are many different of the subject has several parameterizations to choose from, but most different depending on a cubic or quadraticthe wind speed relationship (cubic or quadratic). The choice of parameterisationis problem is not trivial as indicated even by the name of an international one of the meeting that focusseds on the topic implies (the COST-735 Action organized "k conundrum" workshop, (COST-735 Action organized meeting in Norwich, February 2008). The conclusions from this meeting Its results have been incorporated into a recent review book chapter (Garbe et al., 2014). This paper will concentrates on quantifying the very uncertainty caused by the choice of the gas transferflux velocity parameterization in the case of the North Atlantic and the European Arctic. These regions wereas chosen as they are the due to being the areas for which many of the parameterisations were originally derived. They are of many studies some of the parameterizations were based on and also as a regions with wind distributions skewed tilted towards higher winds (in comparison to than the global average) enabling to test the effect of stronger winds on the net flux calculations to be investigated through on the difference of net fluxes calculated using the published gas transfer velocity formulas.

2. Methods

2.1 Datasets

We calculated air-sea <u>CO₂ carbon dioxide</u> fluxes using a set of <u>software</u> processing tools <u>callednamed the</u> 'FluxEngine' (Shutler et al., 2016)) (which is available on the http://www.ifremer.fr/cersat1/exp/oceanflux/), which were created within European Space Agency funded OceanFlux Greenhouse Gases project (http://www.oceanflux-ghg.orgGoddijn Murphy et al., 2015). All gas flux calculations were performed using the FluxEngine software, we were only endusers of. The software became publicly available in March 2016 under an open source license, but

at the time we started this study we did not have more information about it than is included in the paper describes a set of tools (Shutler et al., 2016). This was a conscious decision because, even as we had access to the toolset developers, we wanted to test it as the end users (this is probably the first study using the toolset by authors who had no part in creating it). The tools were developedidea, beyond the tool development, was to provide the community with a verified and consistent toolbox and to encourage the use of satellite Earth Observation (EO) data for studying air-sea fluxes. The toolbox source code can be downloaded or alternatively there is a version that can be run through a web interface. Within the online FluxEngineweb interface, a suite of reanalyszies data products, in situ and model data are available as input to the toolbox. These data are freely available for the scientific community to that can be use by the scientific community and to aid the interpretation of the resultant flux data. The FluxEngine allows you to selectre is a choice of several different <u>air-sea</u> flux parameterizations, as well as input data, allowing the generation of the monthly global gridded air-sea flux products with 1°x1° spatial resolution. The output consists offiles contained twelve NetCDF filessets (one fileset per month) in a NetCDF files. Some Monthly Each compositeoutput file data set includes the mean (first order moment), median, standard deviation and the second, third and fourth order moments. calculated for each calendar month. There is also information (meta data) about origin of data inputs as well as results of our calculations. U Input data users can choose from all of the data available on the web portal Flux Engine program (example perhaps from monthly EO input data include: rain intensity and event, wind speed and direction, % of sea ice cover from monthly model data, ECMWF air pressure, whitecapping (Goddijn-Murphy et al., 2011), two options for from-monthly climatology ofas pCO₂, SST, salinity) and configure them in a various way (Shutler et al., 2016). The user then needs to choose the different components and structure of the air-sea gas fluxin a-calculation and choose the process as a way of computed transfer velocity, parametrization to the wind speed calculation, corrections etc. The FluxEngine has been developed not only to support the study of the air sea flux of CO₂, but also to aid the study of other gases as DMS and N₂O (Land et al., 2013; Shutler et al., 2016).

For the calculations, we used pCO_2 and salinity values from Takahashi et al. (2009) climatology which is based on more than 3 million measurements of surface water pCO_2 in open-ocean environments during non El Nino conditions, (recalculated to fugacity in the FluxEngine toolset). 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 et al., 2014) pCO_2 and associated 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. The SOCAT databases have been re-analysed and then converted converted to climatologies using the methodology described in Goddijn-Murphy et al. (2015). All the climatologies were calculated for year 2010 within the FluxEngine toolset. The SSTfnd values were taken from Operational Sea Surface Temperature and Sea Ice Analysis (OSTIA) (Donlon et al., 2011), and in the case of SOCAT database, while SST skin data that we use come from ARC/(A)ATSR Global Monthly Sea Surface dataset (Merchant et al., 2012). Both data sets have been preprocessed in the same way for use with the using the toolsets of FluxEngine (Shutler et al., 2016).

We used Earth Observation (EO) wind speed and sea roughness (σ_0 in Ku band from GlobWave L2P products) data obtained from the European Space Agency (ESA). The GlobWave satellite products give a "uniform" set of along track satellite wave data from all available Altimeters (spanning multiple space agencies) and from ESA Synthetic Aperture Radar (SAR) data. GlobWave Project is an initiative funded by ESA and subsidised by CNES. The aim of the project is to improve the uptake of satellite-derived wind-wave and swell data by the scientific, operational and commercial user communities. This has been achieved by providing a uniform, harmonized, quality controlled, multi-sensor set of satellite wave data. Wave data is collected from both altimeters (ERS-1, ERS-2,

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ENVISAT, Topex/POSEIDON, Jason-1, Jason-2, CryoSAT, GEOSAT and GEOSAT Follow On) and from ESA Synthetic Aperture Radar (SAR) missions, namely ERS-1, ERS-2 and ENVISAT. All data come in netCDF-3 format.

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 <u>analyses the data</u> were <u>performed usinged</u> global <u>dataly</u> within the FluxEngine software. From the gridded product (1 x 1 deg) we extracted the extratropical North Atlantic (north of 30° N), and its subset, the European Arctic (north of 64° N). <u>For comparison, we also calculated fluxes in the Southern Ocean (south of 40 °S). Hereafter we follow <u>We use everywhere</u> the convention of <u>that</u> sources <u>of CO₂</u> (upward ocean-to-atmosphere <u>gas fluxes</u>) <u>are being</u> positive and sinks (downward atmosphere-to-ocean <u>gas fluxes</u>) <u>arebeing</u> negative. We give all results of <u>net CO₂ earbon-fluxes</u> in the SI unit of Pg (<u>which is numerically identical to Gt</u>).</u>

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 (F, mg C m⁻² day⁻¹) as the product of gas transfer velocity (k, ms⁻¹) and also the difference in CO_2 concentration (gm⁻³) withinat the sea water and itsthe interface with the—air (Land et al., 2013). The concentration of CO_2 in sea water is thea product of its solubility (α , gm⁻³ μ atm⁻¹) and its fugacity (fCO_2 , μ atm). Solubility is in turn, a function of salinity and temperature. Hence FEq. (1) is defined represented as:

$$F = k \left(\alpha_W f CO_{2W} - \alpha_S f CO_{2A} \right) \tag{1}$$

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 equation (1) now becomes:

$$F = k \left(\alpha_W p C O_{2W} - \alpha_S p C O_{2A} \right) \tag{2}$$

We can also ignore the differences between the two solubilites and just use the water side solubility α_W . Equation (2) will be represented as:

$$F = k \alpha_{W} (pCO_{2W} - pCO_{2A})$$
 (3)

This formulation is often referred to as the 'bulk parametrization'.

In this work we chose to analyze the <u>air-sea gas</u> fluxes using five different <u>gas transfer</u> parameterizations (, within the FluxEngine software, using in terms of wind speed to parametrized k). All of them are <u>wind speed</u> parameterisationszed, but <u>with wind speed and differ</u> in the formula usedfor gas transfer velocity, k:

$$k = \sqrt{(660.0/ \text{ Sc}_{\text{skin}})} * (0.212 \text{ U}_{10}^2 + 0.318 \text{ U}_{10})$$
(Nightingale et al., 2000),

$$k = \sqrt{(660.0/ \text{ Sc}_{\text{skin}}) * 0.254 \text{ U}_{10}^2}$$
 (5) (Ho et al., 2006),

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$$k = \sqrt{(660.0/\,\text{Sc}_\text{skin})} * 0.0283 \,\,\text{U}_{10}{}^3 \tag{6}$$
 (Wanninkhof and McGillis, 1999), 208

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$$k = \sqrt{(660.0/ \text{ Sc}_{\text{skin}}) * 0.251 \text{ U}_{10}^2}$$
 (Wanninkhof, 2014),

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$$k = \sqrt{(660.0/ \text{Sc}_{\text{skin}}) * (3.3 + 0.026 \text{ U}_{10}^3)}$$
 (8)
213 (McGillis et al., 2001),

where the subscripts are Schmidt numbers at the skin surface (Sc_{skin}), 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 10 m above the sea surface.

In addition to the purely wind driven parameterizations, we have used the combined Goddijn-Murphy et al. (2012) and Fangohr and Woolf (2007) parametrization, which was developed as a test algorithm ereated within as part of OceanFlux GHG Evolution project and it is provided as an option in the FluxEngine toolbox. This parameterization separates contributions from direct and bubble-mediated gas transfer as suggested by Woolf (2005). Its purpose is to enable a separate evaluation of the effect of the two processes on air-sea gas fluxes and it is an algorithm that has yet to be calibrated should not be treated as the final product (one of the aims of the ongoing OceanFlux Evolution project is to develop a calibration for this algorithm improving this parameterization). We used these OceanFluxGHG Evolution parameterizations in two versions of this parameterisation: wind driven direct transfer (using the U10 wind fields) and radar backscatter driven direct transfer (using mean wave square slope) as described in Goddijn-Murphy et al. (2012).

3. Results

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Using the FluxEngine software, we have produced CO₂ global monthly gridded air-sea fluxes and from these we have extracted calculated from them the the values for the two study regions, the extratropical North Atlantic and separately for its subset, the European Arctic seas. Figure 1 shows maps of the monthly mean average CO₂ air-sea fluxes for the North Atlantic, calculated with Nightingale et al. (2000) (hereafter callednamed further as N2000) k parameterisationformula and the Takahashi et al. (2009) climatology for the whole year and for each season. The area, as a whole, is a carbon sink of CO2 but even the seasonal maps show 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 the above mentioned sinks areas close to the North Atlantic Drift And East Greenland current are which become sinks of CO₂ in the summer (likely due to effect the growth of of phytoplankton-blooms) while the southernmost areas of the regionstudy become CO₂ sources in summer and autumn (which is likely to be due to the effect of sea-water temperature changes). Much of this variability is caused by changes of the surface water pCO_2 average values, shown in Figure 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 $\Delta p CO_2$ and k. In most parameterizations k is a function of wind speed (eqs. 4-8). The mean average wind speed U_{10} for the whole year and each season are shown in Figure 3. The wind speeds in the North Atlantic are higher than the mean value in the world ocean, with mean average values higher than 10 m s⁻¹ in many regions of the study area in all seasons except for the summer (with highest values in winter). This is important because the <u>air-sea fluxes</u> depends 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 the <u>air-sea</u> fluxes calculated <u>using twoon</u> 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 <u>air-sea</u> flux values <u>when</u> —compared to <u>thea</u> "quadratic" functionone, in the regions of high winds, and lower <u>absolute air-sea</u> flux values in <u>with-weaker</u> winds.

Figure 5 shows the monthly values of CO₂ air-sea fluxes for the five parameterizations (eq. 4-8) for the North Atlantic and the European Arctic. The regions areas are a sinks of CO₂ in every month, although August is close to neutral for the North Atlantic. The results using two cubic parameterisations formula (eqs. 6 and 8) are higher in absolute values, respectively by up to 30% for Wanninkhof and McGillis (1999) and 55% for McGillis (2001), in comparisone to the "quadratic" of N2000 (eq. 4). The other two "quadratic" parameterizations (eqs. 5 and 7) resulted in fluxes within 5% of N2000. Annual fluxes for the North Atlantic and the European Arctic and global (<u>included</u> for comparison) are shown in <u>Table 1</u>Figure 6. In addition to the five parameterizations, the figure presents results for both of the OceanFluxGHG Evolution formulas (using wind and radar backscatter data). The mean and standard deviations of the parametrization ensemble are shown as greavy vertical lines. The standard deviation uncertainty in global fluxes is similar to previous estimates (Sweeney et al., 2007, Landschützer et al., 2014) but they cannot be directly compared due to different parameterization choices and methodologies. The results show that the annual North Atlantic net air-sea CO₂ sinkfluxes, depending on the formula used, varies from -0.38 Pg C for N2000 to -0.56 Pg C for McGillis et al. (2001). In the case of global net air-sea CO₂ sinkearbon flux the values are, respectively, -1.30 Pg C and -2.15 Pg C. Table 1 as well as Figure 76 shows the same data "normalized" to the N2000 data (divided by value), thise allows us to parameterization results in a lower absolute flux values to visualize the relative differences. In the case of the North Atlantic using the "quadratic" Wanninkhof (2014) and Ho et al. (2006) parameterisations results in a net air-sea flux that is respectively 43% and 5% higher in absolute value than the equivalent N2000 result, while the "cubic" Wanninkhof and McGillis (1999) and McGillis et al. (2001) results in values that are up to -28% and 442% higher values. The respective values for the Arctic are 43%, 4% for quadratic as well as 287% and 440% for cubic functions. In the case of global net air-sea CO₂ flux the equivalent respective values are 8% and 9% higher than the N2000 result flux for the quadratic functions as well as 334% and 657% for cubic ones. The OceanFluxGHG parameterization results in net air-sea CO₂ fluxes that are 31/8% and 47/32% higher for North Atlantic than the N2000 result (for the respectively backscatter and wind driven versions respectively). The spread of the Arctic values was lower than the Atlantic ones (see Table 1). On the other hand, the values for the Southern Ocean were slightly higher than for North Atlantic but lower than the global ones, with the exception of the OceanFlux GHG parameterizations. In the case of global values the values is surplus wereas, respectively, 445% and 52% respectively.

All the above results used the Takahashi (2009) *p*CO₂ climatology. For comparison we have also calculated the air-sea fluxes using the re-analysed SOCAT version 1.5 and 2.0 data (Goddijn-Murphy et al., 2015). , interpolated to create a climatology using the FluxEngine toolset (Shutler et al., 2016). Figure 78 shows the results using the N2000 *k* parameterization for all three of the three climatologies. In the case of the North Atlantic study area, although the monthly values show large differences (using both SOCAT datasets results in a larger sink in summer and smaller in winter compare to Takahashi), the annual values are similar: -0.38 PgC for both Takahashi and SOCAT v. 1.5 and -0.41 PgC for SOCAT v. 2.0. In the case of the European Arctic the situation is very different, with Takahashi and SOCAT dataset derived climatologies resulting in inverse seasonal variability but with even as annual net air-sea CO₂ flux results that are similar: -0.102 PgC for Takahashi, -0.085 PgC for SOCAT v. 1.5 and -0.088 PgC for SOCAT v. 2.0.

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4. Discussion

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Our results show that using the three "quadratic" parameterizations (Nightingale et al., 2000; Ho et al., 2006 and Wanninkhof 2014) results in air-sea fluxes values that are within 5% of each other in the case of the North Atlantic. This discrepancy is smaller than the 9% difference identified for the global net global carbon air-sea fluxcase (Fig. 76). This result would confirms that at present, these different parameterizations are interchangeable for the North Atlantic as this variation is being all within the experimental uncertainty (Nightingale, 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 ¹⁴C inventories in the case of Wanninkhof (2014). The This makes it possible to be highly confident that at least average air-sea CO₂ fluxes calculated with these three formulas are close to the unknown true values. However, the differences between these and the quadratic parameterisations 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 air-sea CO₂ fluxes that are considerably larger in absolute values, by -up to 33% in the North Atlantic and 50% globally.

The above results imply smaller relative differences between the parameterizations in the North Atlantic than globally. This is interesting because the North Atlantic is the region of strong winds and over most of its area there are no seasonal changes in the air-sea flux direction (Fig. 1). For example in the South Atlantic annual mean of wind speed is within 8.48 m s⁻¹ (Takahashi et al., 2009) and sink of CO₂ (south of 45°) decrease significantly after 1990 with increasing wind speeds what can influence higher concentration of pCO_2 in surface water due to enhance vertical mixing of deep waters and biological activity. (Le Quèrè et al., 2007). Takahashi et al. (2009) also indicate that the flux difference in the Southern Ocean are very strong dependence to the choice of the gas parameterizations and wind speed. This is even more surprising, for North Atlantic, -given if one realizes that at least some of the older parameterizations were developed using, based on a smaller range of winds range than can exist the ones present in the North Atlantic. After analysis of this unexpected fact, using the formula multiplied by the different wind distribution, we have found two reasons for thisat. First when comparing quadratic and cubic parameterizations (Fig. 98), it is clear that these cubic parameterisations ones imply higher air-sea 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:

$$F_1(U) = a U^2, (9)$$

$$F_2(U) = b \ U^3. (10)$$

The difference between the two functions ΔF is equal to:

$$\Delta F = F_2 - F_1 = b U^3 - a U^2 = b U^2 (U - a b^{-1}) = b U^2 (U - U_x)$$
 (11)

where $U_x = a \ b^{-1}$. The difference is positive for wind speeds greater than U_x and negative for smaller winds less U_x ones. U_x is the value of wind speed for which the two functions intersect. In the case of equations (6) and (7), where a = 0.251 and b = 0.0283, they imply thating $U_x = 8.87 \text{ m s}^{-1}$. In fact all of the functions presented in Fig. 98 producehave very similar values for U_x , all of

which are wind speeds close to 9 m s⁻¹. Thise value is very close to average wind speeds in the North Atlantic (Fig. 3). This is one of the reasons of the small relative difference in net air-sea flux differences. The spread of flux values for the Southern Ocean seems to support this conclusion, being larger than the North Atlantic one. Southern Ocean has on average stronger wind than the North Atlantic (including also the Arctic Seas) which seem to have the smallest spread of flux values for different parameterizations. The other reason is the lack of seasonal variation in the sign of the air-sea fluxity. In the case of seasonal changes in the air-sea flux direction (caused by seasonal changes in water temperature or primary productivity), with winds stronger than U_x in some seasons and weaker in others (usually strong winds in winter and weak in summer), the air-sea fluxes partly cancel each other while the difference between cubic and quadratic parameterizations add to each other due to simultaneous changes in the sign of both fluxes itself and the $U - U_x$ term. This effect of seasonal variationity has been suggested to us baseding on available observations data—(A. Watson – personal communication) but we are unaware of any paper investigating itexplaining it in the terms of arithmetic formulas, or even describing it explicitly.

In addition to the five parameterizations described above, we calculated the air-sea fluxes using for the OceanFluxGHG Evolution combined formula, which parameterises the separating contributions from direct and bubble-mediated gas transfer into separate components. The resulting air-sea fluxes are higher in absolute terms, than from all of the quadratic functions considered in this study, and are closer in value being closer to cubic parameterisations ones. This may mean that the bubble mediated term of Fangohr and Woolf (2007) is may be an overestimating ion for the bubble component, implying the need for a dedicated calibration effort CO₂-fluxes. This question will be the subject of further studies in the OceanFlux Evolution project.

Although, using both Takahashi climatology and SOCAT pCO_2 <u>climatologydataset</u> (Fig. <u>87</u>) results in similar annual <u>net air-sea</u> fluxes in the North Atlantic, it should be noted that they show different seasonal <u>variationschanges</u>._-

This may have been caused by slightly different time periods of the datasets (i.e. the SOCAT based climatology contains more is more recent data). The difference is much larger in the European Arctic due to the underlying sparse much worse data spatial coverage and possible interpolation artifacts (Goddijn-Murphy et al., 2015). This discrepancy makes us treat the net air-sea CO₂ flux results from the Arctic with much less confidence than the valuesones for the whole North Atlantic. This situation is likely to may improve as thefter SOCAT dataset grows. It is impossible to declare within this study which dataset is more accurate as only new data can settle this. However, such data have been recently published (Yasunaka et al., 2016). The observed water pCO₂ data (Fig. 3 in Yasunaka et al., 2016), especially since 2005, show clearly an annual cycle compatible with the SOCAT seasonal flux variability. v.3 which is planned to be released in 2016.

5. Conclusions

In this paper we have studied the effect of the choice of gas transfer velocity parameterization on the net CO₂ air-sea gas fluxes 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 it is globally. The difference in the annual net air-sea CO₂ flux difference caused by the choice of the 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. It and is respectively up to 426% different for North Atlantic, 3640% for Arctic and 657% when comparing between the cubic and quadratic functions. In both cases the uncertainty in the North Atlantic and Arctic regions are smaller than the global case. We explain that thee smaller North Atlantic variability isby the combination of firstly higher than

global average wind speeds in the North Atlantic, closer to 9 m s⁻¹, which is the wind speed at which when most k parameterization have similar values, –and secondly the all-season CO₂ sink conditions in most North Atlantic areas. We repeat the analysis using compare—Takahashi and a SOCAT pCO₂ derived climatology and datasets—finding that although the seasonal variability in the North Atlantic is different, the annual net air-sea CO₂ fluxes are within 8% in the North Atlantic and 19% in the European Arctic. The seasonal flux calculated from the two pCO₂ datasets changes—in the Arctic have inverse seasonal variations, change in both datasets—indicating possible under sampling (aliasing) of the pCO₂ in this polar region and therefore highlighting the need to collect for more polar pCO₂—observations in all months and seasons data before than available at present.

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Figure 1. Seasonal and annual mean air-sea fluxes of CO₂ (mg 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). The gaps (white areas) are due to missing data, land and ice masks and interpolation algorithms of the FluxEngine software.

Figure 2. Seasonal and annual pCO_2 values (µatm) in surface waters of the North Atlantic, estimated using the Takahashi (2009) climatology in a) annual, b) DJF (Winter), c) MAM (Spring), d) JJA (Summer), e) SON (Autumn). The gaps (white areas) are due to missing data, land and ice masks and interpolation algorithms of the FluxEngine software.

Figure 3. Wind speed distribution U_{10} (ms⁻¹) 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). The gaps (white areas) are due to missing data, land and ice masks-and interpolation algorithms of the FluxEngine software.

Figure 4. Differences maps for the air-sea CO₂ fluxes (mg 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). The gaps (white areas) are due to missing data, land and ice masks and interpolationalgorithms of the FluxEngine software.

Figure 5. Monthly values air-sea fluxes of CO₂ (Pg_/month⁻¹) for the five parameterizations (eq. 4-8) in a) North Atlantic, b) European Arctic.

Figure 6. Annual air-sea fluxes of CO₂ (Pg_/year⁻¹) for the five (eq. 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.

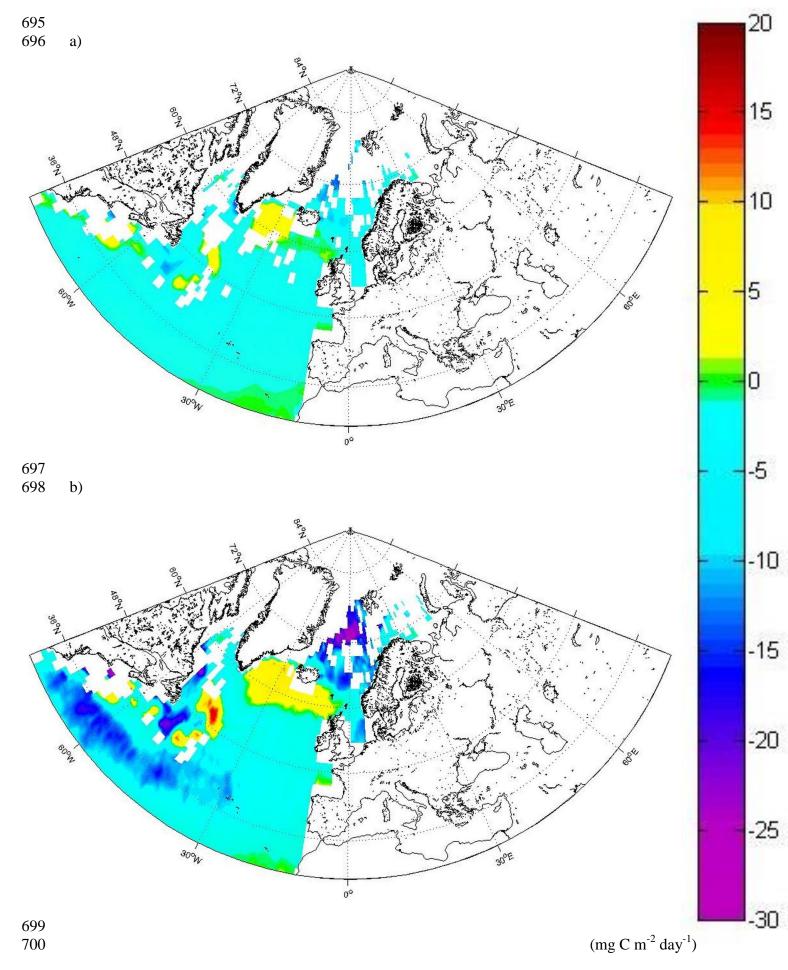
Figure <u>67</u>. Annual air-sea fluxes of CO₂ for the five (eq. 4-8) parameterizations as well as for backscatter (default) and wind driven OceanFluxGHG parameterization normalized to flux values of Nightingale et al. (2000) *k* parameterization (see text) in a)global, b) North Atlantic, c) European Arctic, d)-Southern Ocean. Average values for all parameterization and standard deviations are marked as vertical gray lines.

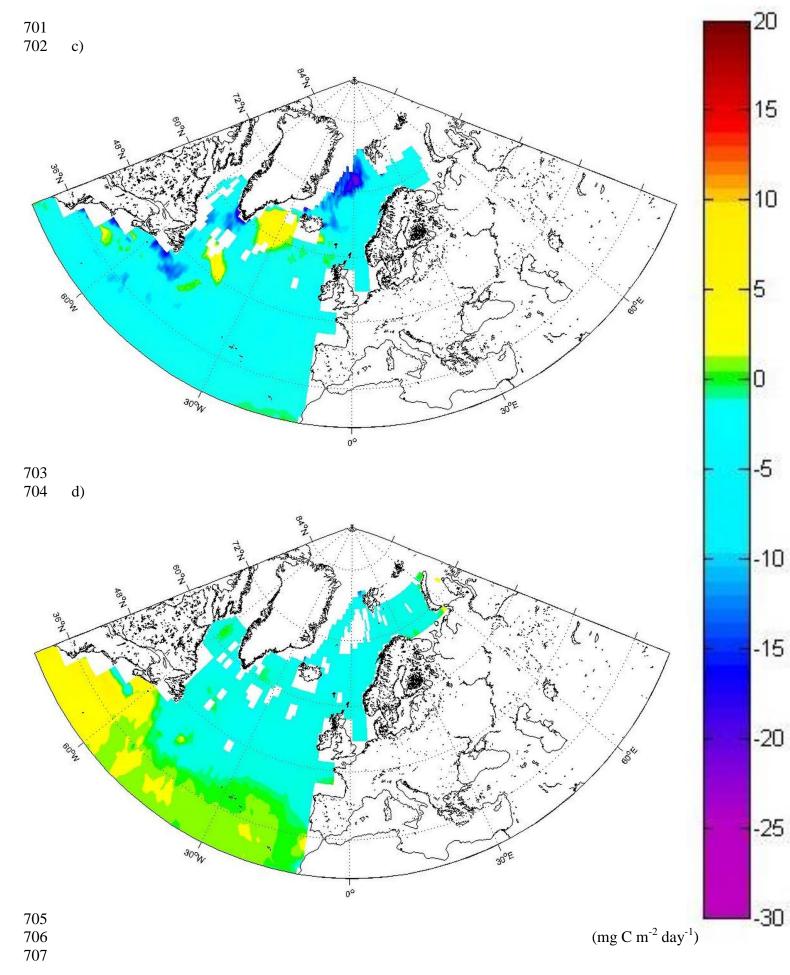
Figure 78. Comparison of monthly values fluxes of air-sea CO_2 fluxes calculated with different pCO_2 datasets (Takahashi et al., 2009, SOCAT v. 1.5 and 2.0) using the same k parameterization (Nightingale et al., 2000) in a) North Atlantic, b) European Arctic.

691 | Figure <u>89</u>. Different k660 parameterizations as a function of wind speed.

<u>Table 1. Annual air-sea CO2 fluxes (in Pg) using different k parameterizations. The values in parentheses are fluxed normalized to Nightingale et al., 2000 (as in Fig. 6)</u>

	Global	<u>Arctic</u>	North Atlantic	Southern Ocean
Nightingale et al., 2000	<u>-1.30</u>	<u>-0.102</u>	<u>-0.382</u>	<u>-0.72</u>
	(1.00)	<u>(1.00)</u>	(1.00)	(1.00)
Ho et al., 2006	<u>-1.42</u>	<u>-0.106</u>	<u>-0.402</u>	<u>-0.76</u>
	(1.09)	(1.04)	(1.05)	(1.06)
Wanninkhof and McGillis, 1999	<u>-1.73</u>	<u>-1.130</u>	<u>-0.490</u>	<u>-0.93</u>
	(1.33)	(1.28)	(1.29)	(1.30)
Wanninkhof, 2014	<u>-1.40</u>	<u>-0.105</u>	<u>-0.398</u>	<u>-0.76</u>
	(1.08)	(1.03)	(1.04)	(1.05)
McGillis et al., 2001	- <u>2.15</u>	<u>-0.147</u>	<u>-0.557</u>	<u>-1.08</u>
	(1.65)	(1.44)	(1.46)	(1.49)
OceanFlux GHG wind driven	<u>-1.98</u>	<u>-0.138</u>	<u>-0.560</u>	<u>-1.14</u>
	(1.52)	(1.36)	(1.47)	(1.58)
OceanFluxGHG backscatter	<u>-1.88</u>	<u>-0.130</u>	<u>-0.526</u>	<u>-1.09</u>
	(1.44)	(1.27)	(1.38)	(1.51)





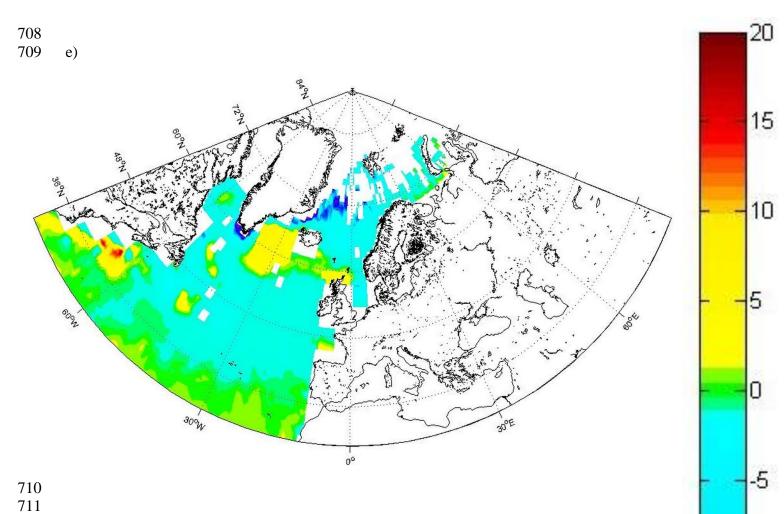


Figure 1. Seasonal and annual mean air-sea fluxes of CO₂ (mg C m⁻² day⁻¹) in the North Atlantic, combine using Nightingale et al. (2000) *k* parameterization and the Takahashi (2009) climatology in a) annual, b) DJF (Winter), c) MAM (Spring), d) JJA (Summer), e) SON (Autumn). The gaps (white areas) are due to missing data, land and ice masks-and interpolation algorithms of the FluxEngine software.

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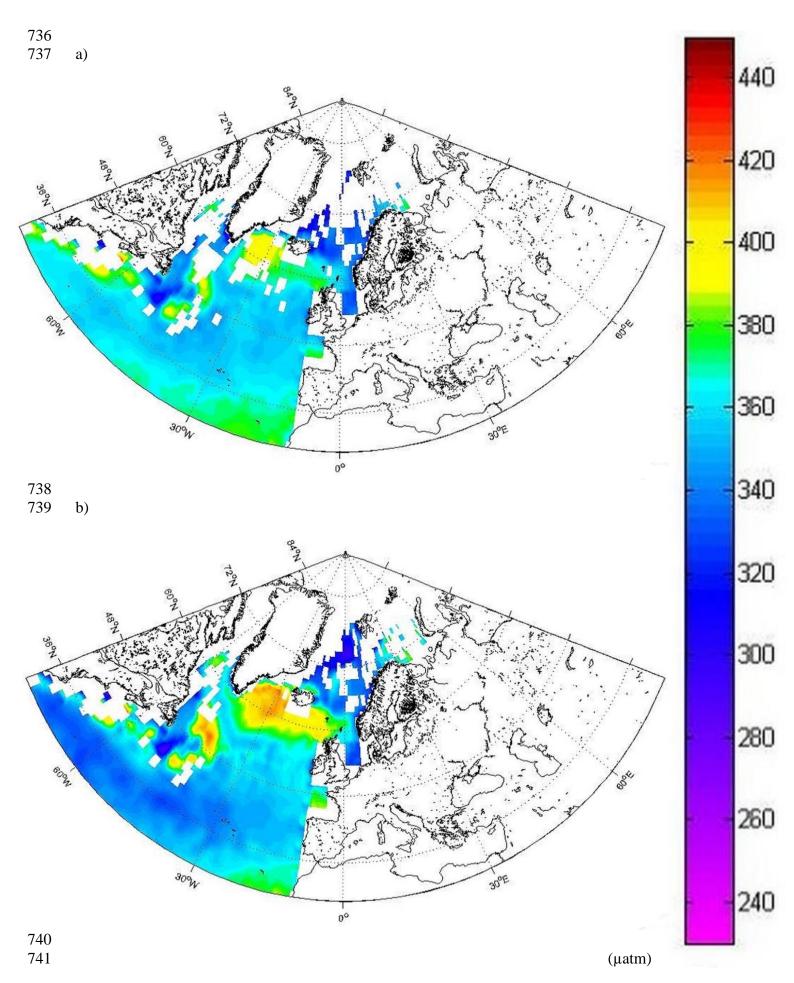
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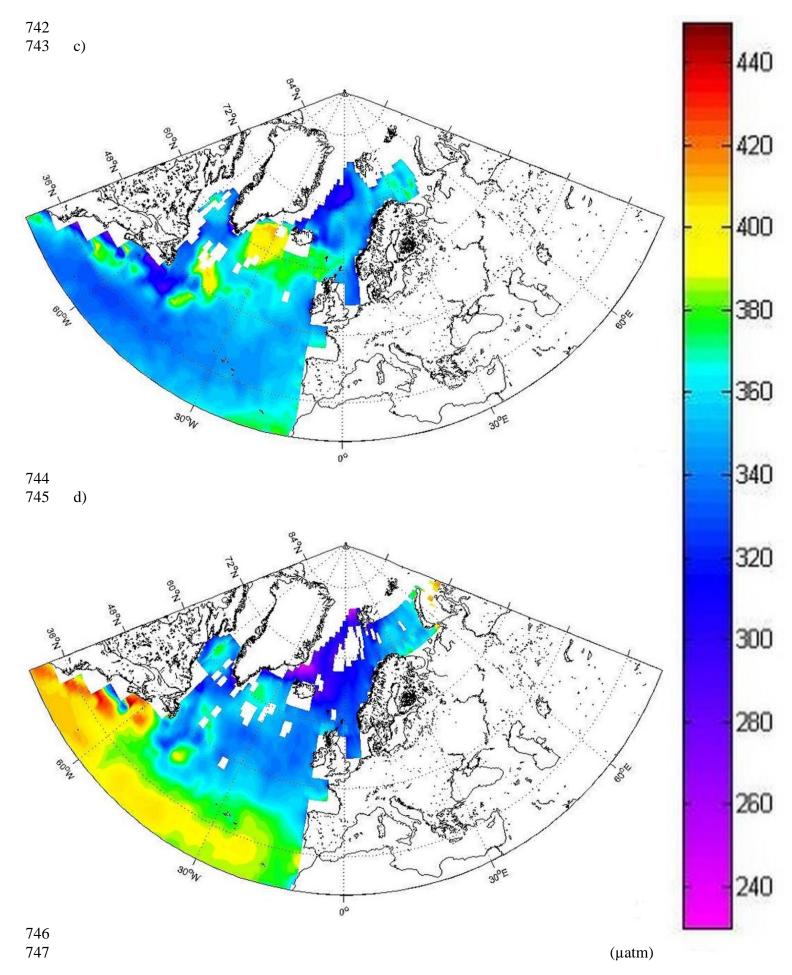
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(mg C m⁻² day⁻¹)





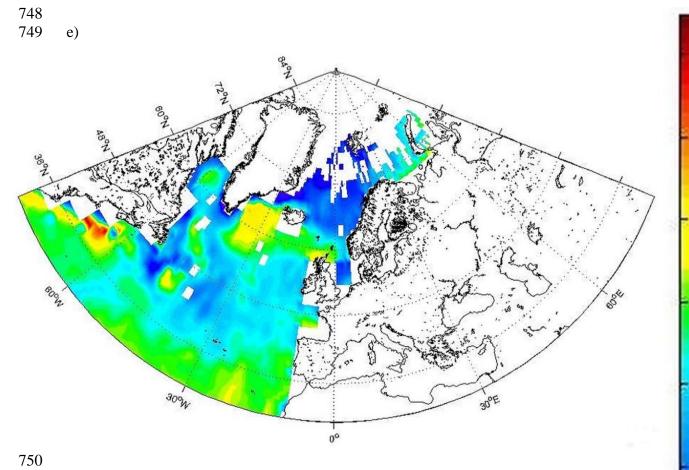
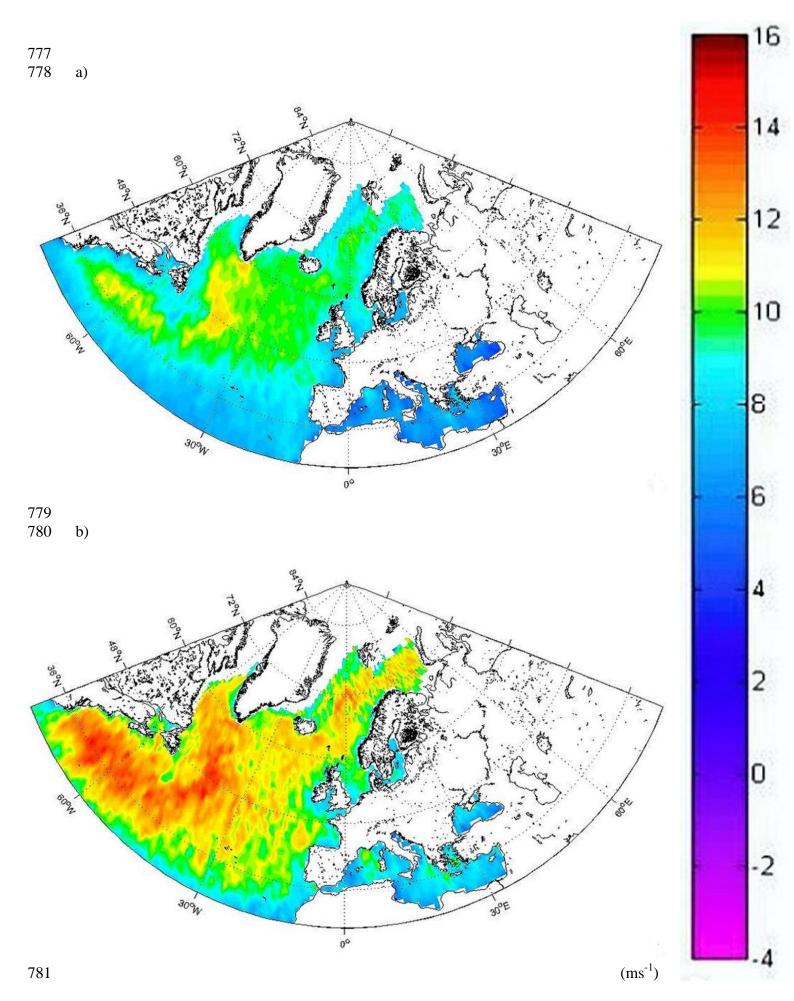
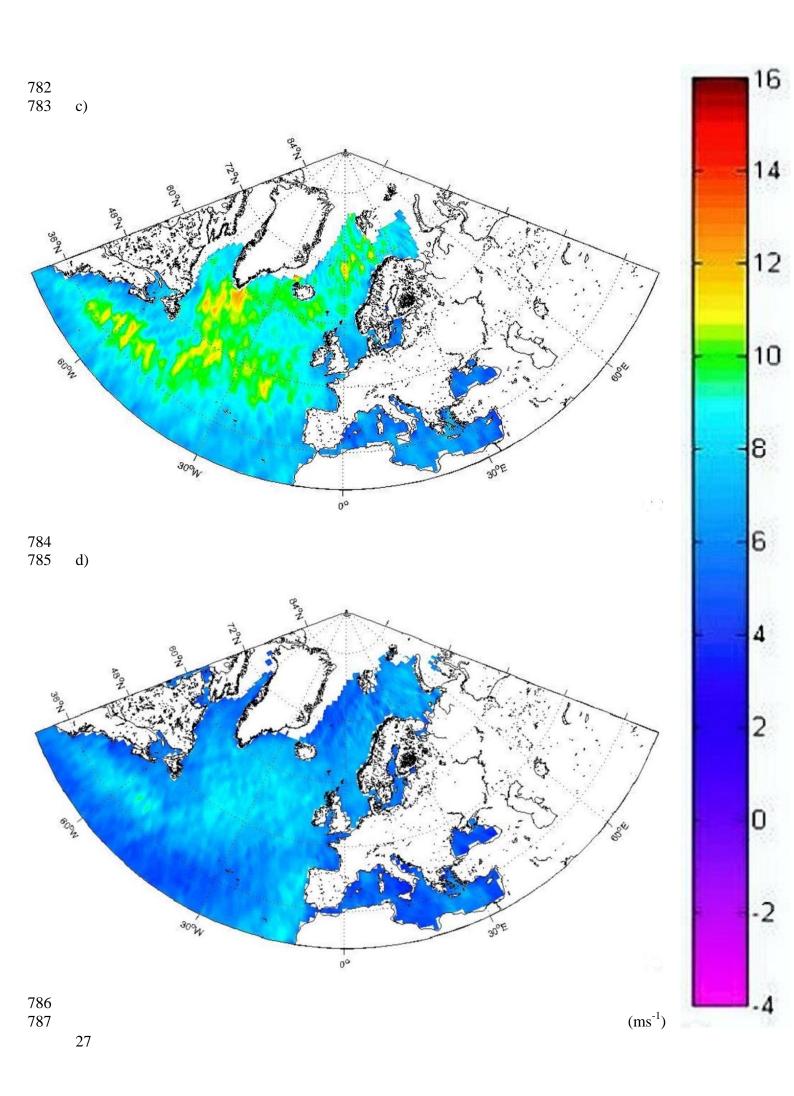
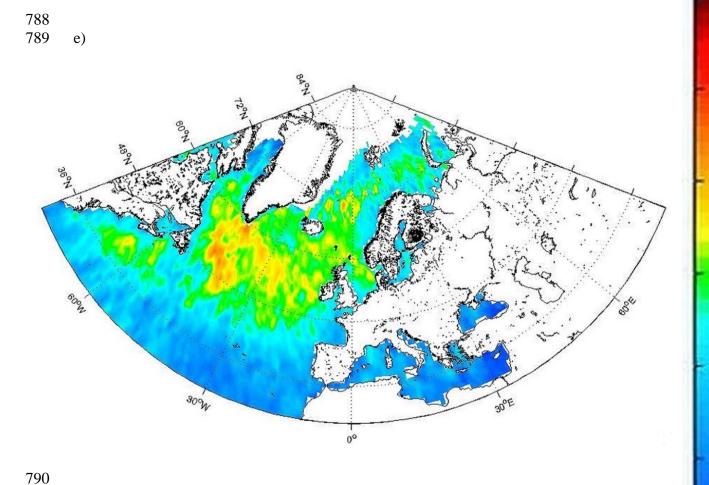


Figure 2. Seasonal and annual pCO_2 values (μ atm) in surface waters of the North Atlantic, estimated using Takahashi (2009) climatology in a) annual, b) DJF (Winter), c) MAM (Spring), d) JJA (Summer), e) SON (Autumn). The gaps (white areas) are due to missing data, land and ice masks-and interpolation algorithms of the FluxEngine software.

(µatm)



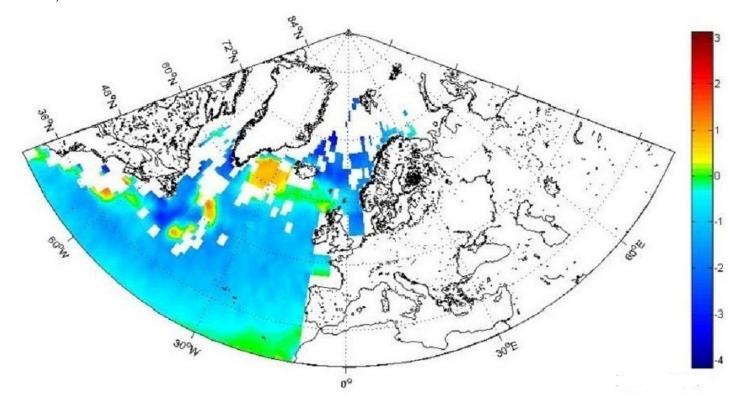




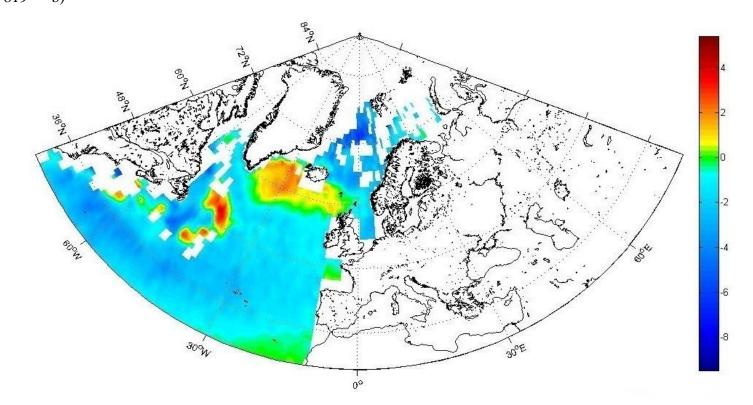
 (ms^{-1})

Figure 3. Wind speed distribution U_{10} (ms⁻¹) 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). The gaps (white areas) are due to missing data, land and ice masks and interpolation algorithms of the FluxEngine software.

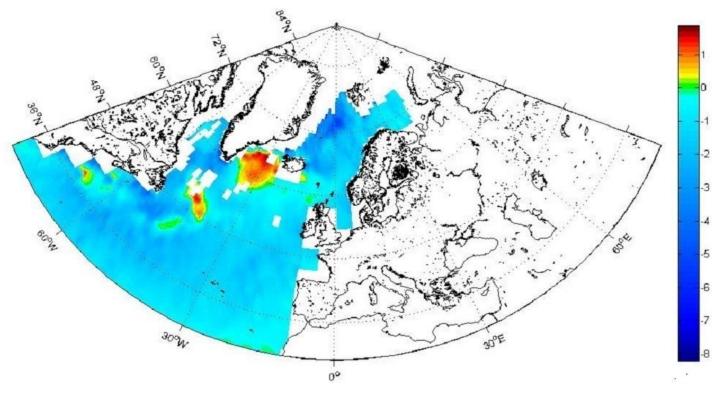
816 817 a)



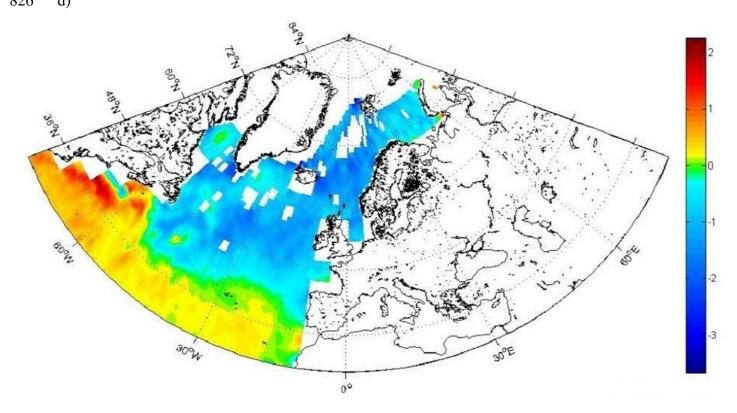
818 (mg C m⁻² day⁻¹) 819 b)



820 821 822 (mg C m⁻² day⁻¹) 823 824 c)



825 826 (mg C m⁻² day⁻¹) d)



(mg C m⁻² day⁻¹) 827 828 829



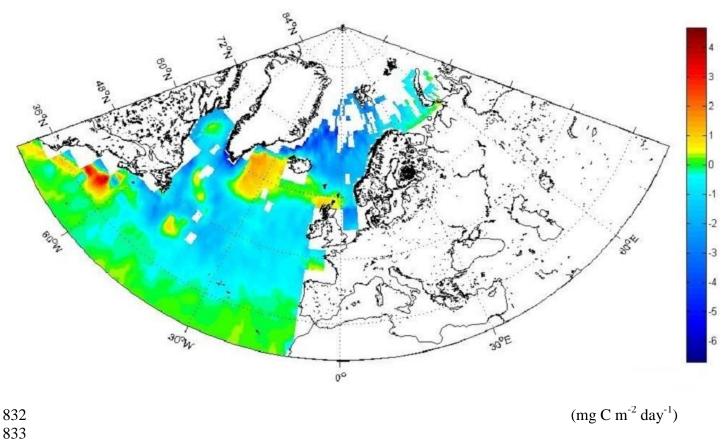


Figure 4. Differences maps for the air-sea CO₂ fluxes (mg C m⁻² day⁻¹) in the North Atlantic, between a wind cubed and squared parameterizations (Wanninkhof and McGillis 1999 and Wanninkhof 2014) in a) annual, b) DJF (Winter), c) MAM (Spring), d) JJA (Summer), e) SON (Autumn). The gaps (white areas) are due to missing data, land and ice masks—and interpolation algorithms of the FluxEngine software.

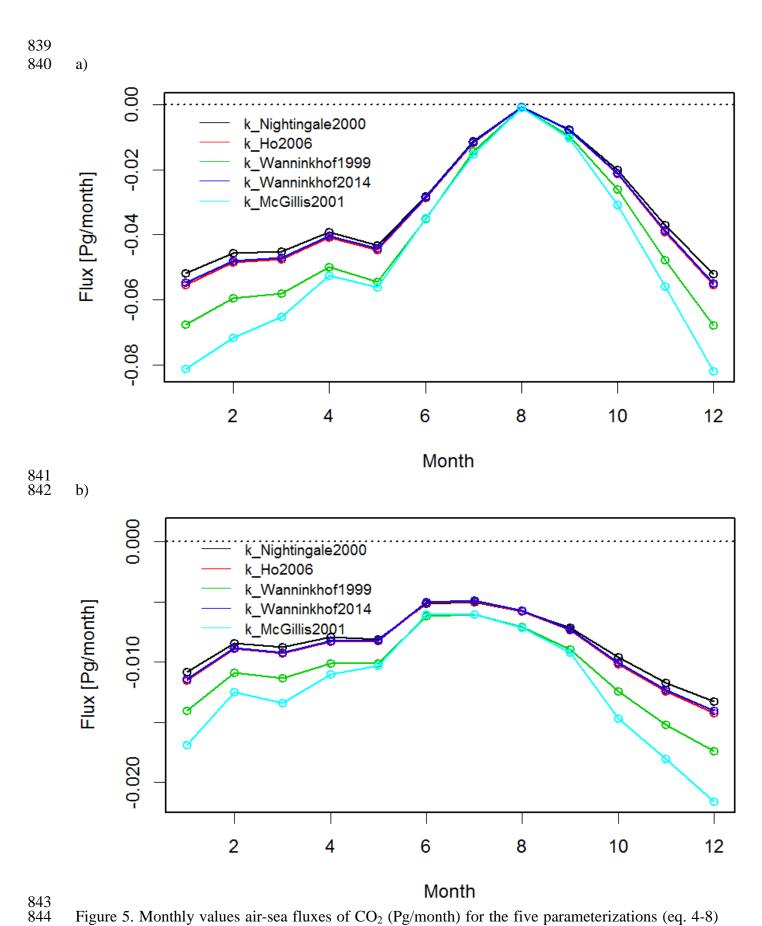
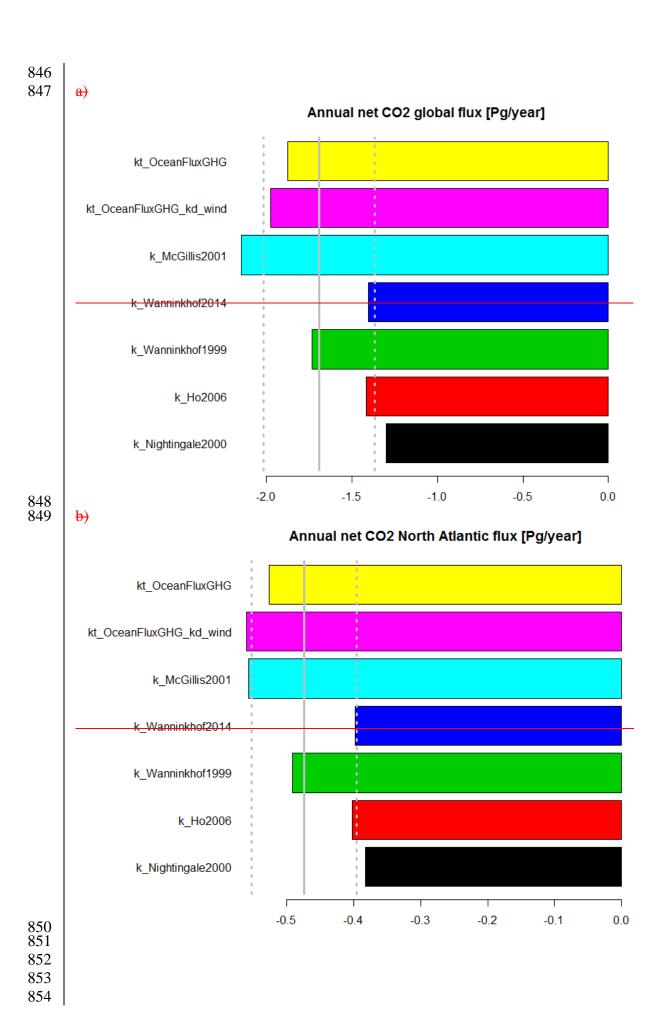


Figure 5. Monthly values air-sea fluxes of CO₂ (Pg/month) for the five parameterizations (eq. 4-8) in a) North Atlantic, b) European Arctic.





e)

Annual net CO2 Arctic flux [Pg/year]

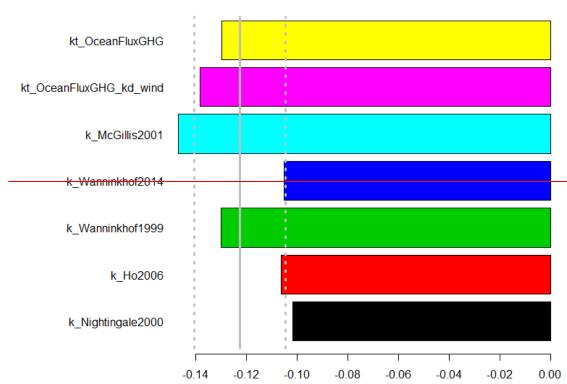
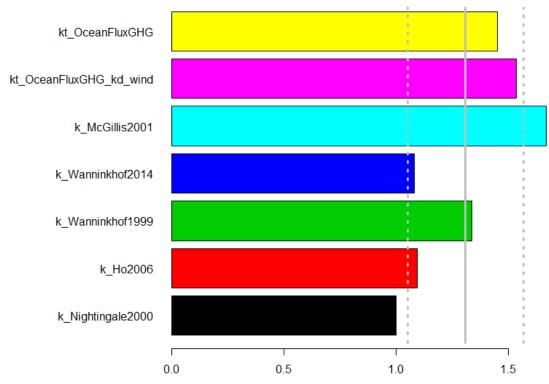
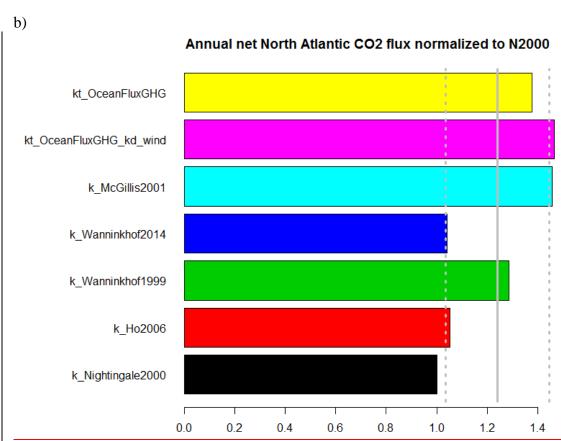


Figure 6. Annual air-sea fluxes of CO₂ (Pg/year) for the five (eq. 4-8) parameterizations as well as for backscatter (default) and wind driven OceanFluxGHG parameterization (see text) in a) global, b) North Atlantic, c) European Arctic. Average values for all parameterization and standard deviations are marked as vertical gray lines.

864 a)

Annual net global CO2 flux normalized to N2000







c)

Annual net Arctic CO2 flux normalized to N2000 kt_OceanFluxGHG kt_OceanFluxGHG_kd_wind k_McGillis2001 k_Wanninkhof2014 k_Wanninkhof1999 k Ho2006 k_Nightingale2000 0.0 0.2 0.4 0.6 8.0 1.0 1.2 1.4

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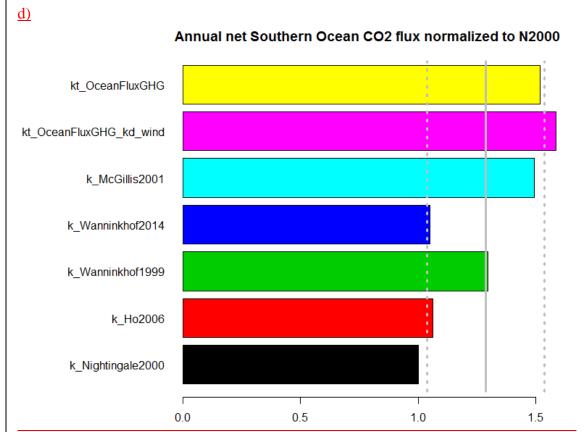


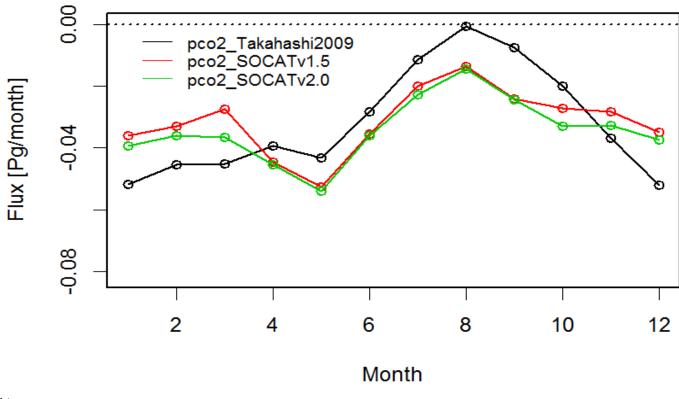
Figure <u>67</u>. Annual air-sea fluxes of CO₂ for the five (eq. 4-8) parameterizations as well as for backscatter (default) and wind driven OceanFluxGHG parameterization normalized to flux values of Nightingale et al. (2000) *k* parameterization (see text). Average values for all parameterization and standard deviations are marked as vertical gray lines.

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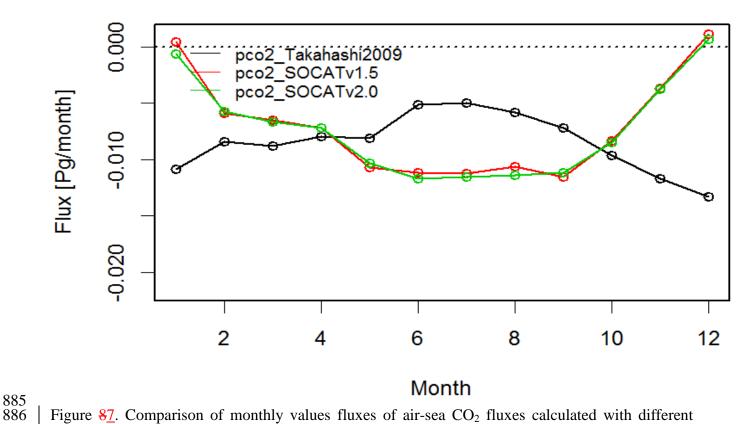
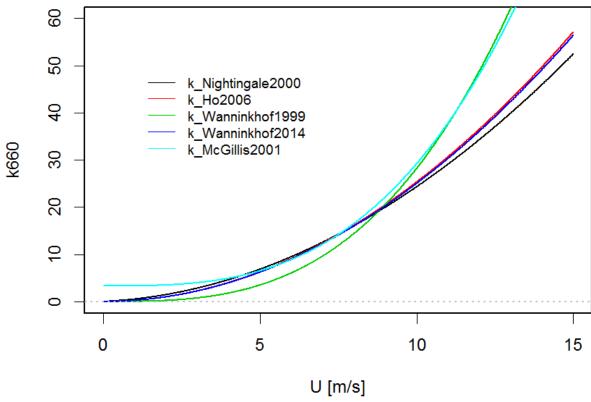


Figure \$7. Comparison of monthly values fluxes of air-sea CO_2 fluxes calculated with different pCO_2 datasets (Takahashi et al., 2009, SOCAT v. 1.5 and 2.0) using the same k parameterization (Nightingale et al., 2000) in a) North Atlantic, b) European Arctic.





892 | Figure <u>89</u>. Different k660 p

arameterizations as a function of wind speed.