# Dear dr. Hoppema,

We want to thank you for your help, the devoted your time and investing our article in the improvement. Thanks yours and the reviewers, comments our manuscript became an article on the high scientific level. Thank you also, that you gave us a chance to correct it and do our best. We are very grateful for this chance. Below is a list of our correctness.

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First two cases where I am not satisfied with your answer to my previous comments:

"We deleted the seasonal part (it was indeed confusing). However, it is not so simple to show that Current and Drift in the Figure (we added information which colored means source and which sink, maybe this will help for a readers). Sorry for that, we really try to do something with this, but this is not so simple, as one can think. We do not have such maps, even to add as a new Figure, so if we want to add this we have to download this from the internet, which is not, in our opinion, a good idea. On balance, we would prefer to retain the regional discussion even if the current positions are not shown."

#### **MY ANSWER**

You can do better than this. Either you make a map using ODV (which is a free tool used by almost the whole oceanographic community) or you just put some letters in the first figure where those currents are situated. This is not that difficult. You cannot discuss something in the paper without showing where it is. Not every reader knows where the North Atlantic Drift is. It is good habit to always show in figures where and what you are talking about.

After your advice we decided to add a map with surface ocean currents in the Arctic, which we downloaded from the website: http://www.grida.no/graphicslib/detail/ocean-currents-and-sea-ice-extent\_4aa6 . We do this followed their permission, which is:

#### Use constraints

This graphic may be reproduced in any form of educational or non-profit purposes without special permission from the GRID-Arendal, provided acknowledgement of the source is made. GRID-Arendal would appreciate receiving a copy of any publication that features this graphic. For any form of publication, please include the link to this page and give the cartographer/designer credit (in this case Philippe Rekacewicz, UNEP/GRID-Arendal)

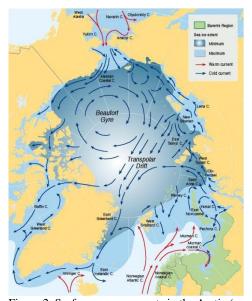


Figure 2. Surface ocean currents in the Arctic (sources: http://www.grida.no/graphicslib/detail/ocean-currents-and-sea-ice-extent\_4aa6, author: Philippe Rekacewicz, UNEP-GRID, Arendal, Norway)

"L323 Somewhat more info about the "OceanFlux GHG Evolution combined formula" would be good to be able to assess what this is all about. Now it is like a black box.

Luckily no longer. We also shared the sentiment that the method could be a black box for a reader. However, we did not feel it would be proper describing it in details, especially before it has been published. The good news is it has been recently published therefore we refer to it directly (Goddijn-Murphy et al., 2016). This is one of the problems when several papers from the same project are submitted in parallel (we were not even aware of this one until it has been published). L334(Goodijn-Murphy et al., 2016) [also the citation has been added to the Literature section]" MY ANSWER

It is great that the citation is available now. What I was referring to is a brief explanation. This does not have to be a long and detailed story. Just some info (one or two sentences) so that the reader knows what it is all about.

Following by Goddijn-Murphy et al. we add:

**L336-340**....OceanFlux GHG Evolution combined formula, which is based on knowledge that airsea exchange is enhanced by air-entraining wave breaking and bubble-mediated transfer, especially for the less soluble gases than CO<sub>2</sub>. Goddijn-Murphy et al. (2016) assume a linear wind relationship for dimethyl sulphide (DMS) and an additional bubble-mediated term for less soluble gases, parameterized with whitecap coverage.

I am using the marked up version of the manuscript for my comments and line numbers

#### L2 North Atlantic Ocean

L2 ... North Atlantic Ocean

L17 perhaps a reference to FluxEngine here?

L17 ...FluxEngine (Shutler et al., 2016)

L18 and the global ocean (not: and globally)

L18.. Ocean, including the European Arctic, and for the global ocean

**L20** ...parameterizations in the North Atlantic Ocean

L23 ...in the North Atlantic Ocean and the Artic than in the global ocean.

L27 parameterizations (+s)

**L26** ...parameterizations...

L30 ...) Takahashi et al (2009) and SOCAT versions 1.5 and 2.0)...

L34 from the two datasets (add: the)

**L32** ...from the two datasets

L55 Pérez (other accent)

L52 ...(Pérez et al., 2013).

L60-61 The global interannual variability in air-sea CO2 flux can be about 60% due to differences in pCO2 and 35% by uncertainty of k (k needs to be defined here as it has not been done earlier) **L57-59** ... and 35% by gas transfer velocity *k* parameterization (Couldrey et al., 2016).

L62-63 data quality of the fugacity of CO2 (fCO2) (why did you skip data quality?)

**L60** ..interpolation, data quality of the fugacity of  $CO_2$ ...

L72, and the choice of the wind speed parameterization may cause variability in k by as much as

about 50% (note comma at beginning)

**L68** ...even by 10% - 40%, and the choice of the wind speed parameterization may cause variability in k as much as about 50%...

L73-74 have analyzed solely the effects of the choice between ...

L70 In this work we have analyzed solely the effects of the choice between various empirical winds driven gas transfer parameterizations.

L79-80 "One of the factors influencing the value of the calculated net air-sea gas flux is the choice of the formula for the gas transfer velocity." This is a repetition of the previous paragraph and should be deleted.

L75 delete

L80 In (not: Within)

**L75** In the literature...

L98 as part of (not: within)

**L92** ...which was created as part of European Space Agency...

L99-100 "All gas flux calculations were performed using the FluxEngine software." Delete as this is exactly the same as in the previous sentence.

L93 deleted

L106 delete: ", as well as input data," (repetition of previous sentence)

L98 deleted

L111 delete: "Users can choose from all of the data available on the web portal;" (repetition of previous info)

L103 deleted

L114 define SST here.

L105...Sea Surface Temperature (SST)...

L140-143 delete: "GlobWave Project is an initiative funded by ESA and subsidised by Centre National d'Etudes Spatial (CNES). The aim of the project was to constrain a uniform, harmonized, quality controlled, multi-sensor set of satellite wind-wave data for using by different communities despite of in situ data." This is not the place to describe projects and it adds nothing to the paper. It only distracts the attention of the reader.

L128 deleted

L151 contained in the (not: within)

**L133** ... contained in the FluxEngine software.

L152 we extracted data from the extratropical North Atlantic Ocean

L134 ... we extracted data from the extratropical North Atlantic Ocean

L177 become

L159 become

L224-225 delete: "(from 224 blue to purple colored in the Fig. 1)" (Everyone can see that in Fig. 1; this is superfluous)

L225-226 but some regions close to North Atlantic Drift and East Greenland Current (Fig. 9), are net sources. (and delete: "(from yellow to red colored in the Fig. 1).", see comment above)

**L204-205** The area, as a whole, is a sink of CO<sub>2</sub> but some regions close to North Atlantic Drift and East Greenland Current (Figure 2) are net sources.

L227-228 "At the seasonal maps one can see more variability affects by physical process or biological activity." This sentence is not clear at all. Please rewrite.

**L205-208** At the seasonal maps one can see more variability controlled by physical process (with temperature changes causing maximum in-water  $pCO_2$  in summer) or biological activity (with phytoplankton blooms causing summer values to be lowest in the annual cycle).

L232 delete: average

L212 ... $pCO_2$  values,

L237 (which is 7 ...) (for clarity)

**L217** ...(which is 7 m s<sup>-1</sup>;...

L240 (see Discussion below)

**L220** ...distribution (see Discussion below).

L250 delete: respectively (unnecessary here)

**L231** ... values, by up to 30%...

L254 and the global ocean ...

**L239-245** Annual net fluxes for the North Atlantic, Southern and global ocean as well as for the European Arctic are shown in Table 1. The results show that the annual North Atlantic net air-sea CO<sub>2</sub> sink, depending on the formula used, varies from -0.38 Pg C for N2000 to -0.56 Pg C for McG2001. In the case of global net air-sea CO<sub>2</sub> sink the values are -1.30 Pg C and -2.15 Pg C, respectively. Table 1 as well as Figure 7 shows the same data "normalized" to the N2000 data (divided by value), which allows us to visualize the relative differences (in Table 1 values in parentheses).

L263 which (not: this)

L244 ...which allows...

L266 4% and 5% higher in absolute value, respectively, ("respectively" always behind) L267-268 are 28% and 44% higher, respectively, than N2000 results. ("up to" is not correctly used here; and higher must be added)

**L246-248**...4% and 5% higher in absolute values, respectively, than the equivalent N2000 result, while the "cubic" WMcG1999 and McG2000 results in values that are 28% and 44% higher, respectively, than N2000 results, for this regions.

L272-273 The OceanFlux GHG parameterization for the backscatter and wind driven versions, results in net ... (change order, increase clarity)

**L252-256** The OceanFlux GHG parameterization for the backscatter and wind-driven versions, results in net air-sea  $CO_2$  fluxes higher for the North Atlantic Ocean than N2000, that are 38% and 47%, respectively, and in the global case the values, for those two versions, were 44% and 52% higher, respectively, than N2000 results.

L276 than that of the Atlantic ones (see Table 1).

**L256** ...values was lower than that of the Atlantic ones (see Table 1).

L283 As it is written now, it seems that SOCAT is part of (Goddijn-Murphy et al., 2015). Please change wording.

**L260-263** All the above results were obtained with the Takahashi et al. (2009)  $pCO_2$  climatology and for comparison, we have also calculated the air-sea  $CO_2$  fluxes using the re-analysed SOCAT versions 1.5 and 2.0 data (which were converted to climatologies using methodology described in Goddijn-Murphy et al., 2015).

L285 both SOCAT versions

L264 ... and both SOCAT veriosns).

L296-298 Change to: Our results show that using the three "quadratic" parameterizations (Nightingale et al., 2000; Ho et 296 al., 2006 and Wanninkhof, 2014) air-sea fluxes are within 5% of each other in the case of the North Atlantic. (twice "results" doesn't look nice)

**L275-277** Our results show that using the three "quadratic" parameterizations (Nightingale et al., 2000; Ho et al., 2006 and Wanninkhof, 2014) air-sea fluxes are within 5% of each other in the case of the North Atlantic (Table 1, values in parentheses).

L299 This confirms that ...

**L278** This confirms that at present,...

**L288** ...in the North Atlantic Ocean and more than 50% in the global ocean.

**L292** ... Atlantic Ocean than in the global ocean.

L308 leads (not: may lead)

L287 ...may leads to net air-sea CO<sub>2</sub> fluxes...

L314-319 "For example in the South Atlantic, the annual mean wind speed is 8.5m s-1 (Takahashi et al., 2009), and of the CO2 sink (south of 45°) decreases significantly after 1990 with increasing wind speeds; this may cause higher CO2 concentration (and higher pCO2) in surface water due to enhanced vertical mixing of CO2-rich deep waters (Le Quèrè et al., 2007) and biological activity (seasonal changes in primary production)."

This long sentence is unclear. Moreover, I do not understand how this sentence can logically follow the previous one. If you want to describe a fact pertaining to the North Atlantic with the support of processes going on in the South Atlantic (is that what you intend?) then you have to make that clear. Finally, what the role of biological activity is in this explanation, remains elusive. Please explain and change the text.

We rewrite text above (see below) and also add explanation about biological activity (but not in inside the article). We have removed part which, in our opinion, could not be full understood for the readers and make any confusion for them

Biological activity may enhance mass transfer by modifying the concentration gradient in the mass boundary layers, on the water side of the interface.

**L291-300** The above results imply smaller relative differences between the parameterizations in the North Atlantic Ocean than in the global ocean. 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, the annual mean wind speed is 8.5 m s<sup>-1</sup> which is lower values of wind speed than in the North Atlantic (9 m s<sup>-1</sup>) and the <u>range of</u> seasonal changes in the air-sea <u>CO<sub>2</sub></u> fluxes are <u>from e higher than in the North Atlantic Ocean (±0.05 to -0.05 Pg C yr Takahashi et al., 2009) with differences between the parameterizations lower than in the North</u>

Atlantic (Le Quèrè et al., 2007; Takahashi et al., 2009)... This may cause higher CO<sub>2</sub> concentration (and higher pCO<sub>2</sub>) in surface water due to enhanced vertical mixing of CO<sub>2</sub>-rich deep waters (Le Quèrè et al., 2007). Takahashi et al. (2009) also indicate that the air-sea CO<sub>2</sub> fluxes difference in the Southern Ocean is strongly dependent on the choice of the gas transfer parameterizations and wind speed.

L322 Smaller differences in the North Atlantic Ocean than in the global ocean are surprising, given that at least ...

L324 range of winds than what occurs in the North Atlantic.

**L299** Smaller differences in the North Atlantic Ocean than in the global ocean are surprising, given that at least some of the older parameterizations (e.g. W2009 or WMcG1999) were developed using a smaller range of winds than what occurs in the North Atlantic.

L351-356 This is one sentence, and it is much too long. Please make two out of these.

**L326-331** In the case of seasonal changes in the air-sea flux direction (caused by 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 fluxes partly cancel each other. The difference between cubic and quadratic parameterizations adds to each other due to simultaneous changes in the sign of both fluxes itself and the U -  $U_x$  term.

L370 ... Atlantic; however, it should be ...

L347 ...in the North Atlantic; however it should be...

L372 ... of the datasets as the SOCAT-based dataset contains more recent data.

L348-349 ...time periods of the datasets as the SOCAT-based dataset contains more recent data.

L372-376 It should be noted that a significant part of the data from Takahashi et al. (2009) are included in SOCAT so the differences in the European Arctic may be due to the sparse data coverage and possible interpolation artifacts (Goddijn-Murphy et al., 2015) or to processing of the data through the FluxEngine.

**L349-352** It should be noted that a significant part of the data from Takahashi et al. (2009) are included in SOCAT so the differences in the European Arctic may be due to the sparse data coverage and possible interpolation artifacts (Goddijn-Murphy et al., 2015) or to processing of the data through the FluxEngine.

L376-378 "The results are improved in Couldrey et al. (2016) where modeled and observation data were compared and has been show the same relationships in high-latitude zone." This is not clear at all and should be rewritten. Be precise to describe what you want to say.

**L352-354** A recent paper (Couldrey et al., 2016) using even more high-latitude data than were available in the SOCAT versions 1.5 and 2.0, which we used, shows similar seasonal pattern as SOCAT. Still, this discrepancy...

L380 to decide in this study ...

**L356** It is impossible to decide in this study which datasets...

L382 delete: in-water (ackward)

L359 deleted

L392 delete: within

L369 deleted

L416 correcting
L392 ..correcting...

#### L421 IFREMER

L396

L656 delete: 1-12 (these are not the final page numbers)

L625 deleted

L631 and 714 Figure 2. Surface ocean currents in the Arctic (sources:

http://www.grida.no/graphicslib/detail/ocean-currents-and-sea-ice-extent\_4aa6, author: Philippe Rekacewicz, UNEP-GRID, Arendal, Norway). North Atlantic Drift forming the Norwegian Atlantic Current in the Arctic Ocean.

# Effect of gas-transfer velocity parameterization choice on air-sea CO<sub>2</sub> fluxes in the North Atlantic Ocean and the European Arctic

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Abstract

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The oceanic sink of carbon dioxide (CO<sub>2</sub>) 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. One of the sources of the uncertainty within this calculation is the parameterization chosen for the CO<sub>2</sub> gas transfer velocity. We used a recently developed software toolbox, called the FluxEngine (Shutler et al., 2016), to estimate the monthly air-sea CO<sub>2</sub> fluxes for the extratropical North Atlantic Ocean including,—the European Arctic, and for the global oceanglobally using several published quadratic and cubic wind speed parameterizations of the gas transfer velocity. The aim of the study is to constrain the uncertainty caused by the choice of parameterization in the North Atlantic Ocean. This region is a large oceanic sink of CO<sub>2</sub>, and it is also a region characterised by strong winds, especially in winter but with good in situ data coverage. We show that the uncertainty in the parameterization is smaller in the North Atlantic Ocean and the Arctic than in the global oceanity. It is as little as 5% in the North Atlantic and 4% in the European Arctic, in comparison to 9% for the global ocean when restricted to parameterizations with quadratic wind dependence. This uncertainty becomes 46%, 44% and 65% respectively, when all parameterizations are considered. We suggest that this smaller uncertainty (5% and 4%) is caused by a combination of higher than global average wind speeds in the North Atlantic (> 7 ms<sup>-1</sup>) 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 in situ pCO<sub>2</sub> datasets (Takahashi et al. (2009) and SOCAT versions 1.5 and 2.0) for the flux calculation. The annual fluxes using the two data sets differ by 8% in the North Atlantic and 19% in the European Arctic. The seasonal fluxes in the Arctic computed from the two datasets disagree with each other possibly due to insufficient spatial and temporal data coverage, especially in winter.

#### 1. Introduction

The region of extratropical North Atlantic Ocean, including the European Arctic, is a region responsible for the formation of deep ocean waters (see Talley (2013) for a recent review). This process, part of the global overturning circulation, makes the area a large sink of atmospheric CO<sub>2</sub> (Takahashi et al., 2002; Takahashi et al., 2009; Landschützer et al., 2014; Le Quéré et al., 2015). Therefore, there is a widespread interest in tracking the changes in the North Atlantic net carbon dioxide fluxes, especially as models appear to predict a decrease in the sink volume later this century (Halloran et al., 2015).

The trend and variations in the North Atlantic CO<sub>2</sub> sinks has been intensively studied since observations have shown it appeareds to be decreasing (Lefèvre et al., 2004). This decrease on inter-annual time scales has been confirmed by further studies (Schuster and Watson, 2007) and this trend has continued in recent years North of 40° N (Landschützer et al., 2013). It is not certain how many of these changes are the result of long-term changes, decadal changes in atmospheric forcingnamely the North Atlantic Oscillation (Gonzalez-Davila et al., 2007; Thomas et al., 2008; Gruber 2009; Watson et al., 2009) or changes in meridional overturning circulations (Pérez et al., 2013). Recent assessments of the Atlantic and the Arctic net sea-air CO<sub>2</sub> fluxes (Schuster et al., 2013) and the global ocean net carbon uptake (Wanninkhof et al., 2013) show that the cause is still unknown.

To study the rate of the ocean CO<sub>2</sub> sink and especially its long-term trend, one needs to first constrain the uncertainty in the flux calculation. The global interannual variability in air-sea CO<sub>2</sub> fluxes variability can be vary about 60% due to differentces in pCO<sub>2</sub> and 35% by gas transfer velocity k parameterization (Couldreney et al., 2016). Sources of uncertainty include sampling coverage, the method of data interpolation, data quality of the fugacity of CO<sub>2</sub> (fCO<sub>2</sub>), the method used for normalization of fugacity data to a reference year in a world of ever increasing atmospheric CO<sub>2</sub> the measurement uncertainty in all the parameters used to calculate the fluxes (including partial pressure in water and air, bulk and skin water temperatures, air temperatures, wind speed etc.) and some which are not usually included in the calculations but most probably influence the flux values (sea state parameters, air bubble void fraction, surfactant effects etc.) as well as the choice of gas transfer velocity k parameterization formula (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 in gas transfer velocity, even by 10% - 40%, and the choice of the wind speed parameterization may cause variabilitya difference in the results of k as much as about, even about 50% (Gregg et al., 2014; Couldrey et al., 2016). In this work we have analyzed solely the effects of the choice between various published empirical windwinds driven gas transfer parameterizations. The North Atlantic is one of the regions of the world ocean best covered by CO<sub>2</sub> fugacity measurements (Watson et al., 2011), the Arctic seas coverage is much poorer, especially in winter (Schuster et al., 2013).

One of the factors influencing the value of the calculated net air-sea gas flux is the choice of the formula for the gas transfer velocity. In Within the literature there are many different parameterizations to choose from and most depend on a cubic or quadratic wind speed relationship. The choice of the appropriate parameterization is not trivial as indicated by the name of an international meeting which focused on this topic ("k conundrum" workshop, COST-735 Action organized meeting in Norwich, February 2008). The conclusions from this meeting have been incorporated into a recent review book chapter (Garbe et al., 2014). This paper concentrates on quantifying the uncertainty caused by the choice of the gas transfer velocity parameterization in the North Atlantic and the European Arctic. These regions were chosen as they are the areas for which many of the parameterizations were originally derived. They are also regions with wind fields

skewed towards higher winds (in comparison to the global average) enabling the effect of stronger winds on the net flux calculations to be investigated by using published gas transfer velocity formulas.

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#### 2. Methods

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#### 2.1 Datasets

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We calculated net air-sea CO<sub>2</sub> fluxes using a set of software processing tools called the 'FluxEngine' (Shutler et al., 2016), which was created as part of within European Space Agency funded OceanFlux Greenhouse Gases project (http://www.oceanflux-ghg.org). All gas flux calculations were performed using the FluxEngine software. 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. The FluxEngine allows the usersyou to select several different air-sea flux parameterizations, as well as input data, producing monthly global gridded net air-sea fluxes products with 1° x 1° spatial resolution. The output consists of twelve NetCDF files (one file per month). One 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. Users can choose from all of the data available on the web portal; an For example, of the monthly EO input data includes: rain intensity, wind speed and direction, % of sea ice cover from monthly model data, ECMWF air pressure, whitecapping (Goddijn-Murphy et al., 2011), two options for monthly datasets of pCO<sub>2</sub>, SSTSea Surface Temperature (SST), salinity. The user then needs to choose the different components and structure of the net air-sea gas flux calculation and choose the transfer velocity parameterization.

For the calculations, we used *p*CO<sub>2</sub> and salinity values from Takahashi et al. (2009) climatology which wasis based on more than 3 million measurements of surface water *p*CO<sub>2</sub> in open-ocean environments during non El Nino conditions. For some calculations we used, as an alternative, Surface Ocean CO<sub>2</sub> Atlas (SOCAT) version 1.5 and 2.0 (Sabine et al., 2013; Pfeil et al., 2013; Bakker et al., 2014) *p*CO<sub>2</sub> and associated SST data. SOCAT is a community driven dataset containing 6.3 and 10.1 million surface water CO<sub>2</sub> fugacity values for version 1.5 and 2.0, respectively, with a global coverage. The SOCAT databases have been re-analysed and then converted to climatologies using the methodology described in Goddijn-Murphy et al. (2015). All the climatologies were calculated for year 2010 with the FluxEngine toolset. The SSTskin (defined within Group for High Resolution SST (GHRSST) as temperature of the surface measured by an infrared radiometer operating at the depth of ~10-20 μm) values were taken from the Advance Along Track Scanning Radiometer (ESA/ARC/(A)ATSR) Global Monthly Sea Surface dataset (Merchant et al., 2012) in the case of both datasets, and have been preprocessed in the same way for use with the FluxEngine (Shutler et al., 2016).

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We used Earth Observation (EO) wind speed and sea roughness ( $\sigma_0$  – altimeter backscatter signal 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) and are publicity available at the Ifremere/CERSAT (http://globwave.ifremer.fr/products/data-access). GlobWave Project is funded by ESA and subsidised by Centre National d'Etudes Spatial (CNES). The aim of the project was to constrain a uniform, harmonized, quality controlled, multi-sensor set of satellite wind-wave data for using by different communities despite of in situ data. Wave data are collected from six altimeter missions 137 (Topex/POSEIDON, Jason-1/22, CryoSAT, GEOSAT and GEOSAT Follow On) and from ESA 138 Synthetic Aperture Radar (SAR) missions, namely ERS-1/2 and ENVISAT. All data come in 139 netCDF-3 format.

All analyses were performed using global data <u>contained in within</u> the FluxEngine software. From the gridded product (1° x 1°) we extracted <u>data from</u> the extratropical North Atlantic <u>Ocean</u> (north of 30° N), and its subset, the European Arctic (north of 64° N). For comparison, we also calculated fluxes in the Southern Ocean (south of 40° S). Hereafter we follow the convention of that sources of CO<sub>2</sub> (upward ocean-to-atmosphere gas fluxes) are positive and sinks (downward atmosphere-to-ocean gas fluxes) are negative. We give all results of net CO<sub>2</sub> fluxes in the SI unit of Pg (Pg is 10<sup>15</sup> g which is numerically identical to Gt).

# 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 net air-sea flux of  $CO_2$  (F, mg C m<sup>-2</sup> day<sup>-1</sup>) as the product of gas transfer velocity (k, ms<sup>-1</sup>) and the difference in  $CO_2$  concentration (gm<sup>-3</sup>) in the sea water and its interface with the air (Land et al., 2013). The concentration of  $CO_2$  in sea water is the product of its solubility ( $\alpha$ , gm<sup>-3</sup>  $\mu$ atm<sup>-1</sup>) and its fugacity ( $fCO_2$ ,  $\mu$ atm). Solubility is in turn, a function of salinity and temperature. Hence F is defined 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 with the partial pressure (their values differ by <0.5 % over the temperature range considered) (McGillis et al., 2001). So equation (1) now becomes:

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

One can also ignore the differences between the two solubilites, and just use the waterside solubility  $\alpha_W$ . Equation (2) will then become:

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

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

In this study we chose to analyze the air-sea gas fluxes using five different gas transfer parameterizations (k). All of them are wind speed parameterizations, but differ in the formula used:

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

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

$$k = \sqrt{(660.0/ \text{ Sc}_{\text{skin}}) * 0.0283 \text{ U}_{10}^{3}}$$
 (6)

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

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$$k = \sqrt{(660.0 / Sc_{skin})} * (3.3 + 0.026 U_{10}^{3})$$
 (8)  
189 (McGillis et al., 2001),

where  $Sc_{skin}$  stands for the Schmidt numbers at the skin surface, a function of SST ([= (kinematic viscosity of water)/(diffusion coefficient of  $CO_2$  in water)]), 660.0 is the Schmidt number corresponding to values of carbon dioxide at 20 °C 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) parameterization, which was developed as a test algorithm within of OceanFlux GHG Evolution project. 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. We used two versions of this parameterization: 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 global gridded monthly net CO<sub>2</sub> air-sea fluxes and from these we have extracted the values for the two study regions, the extratropical North Atlantic Ocean and separately for its subset - the European Arctic seas. Figure 1 shows maps of the monthly mean air-sea CO<sub>2</sub> fluxes for the North Atlantic, calculated with Nightingale et al. (2000) (hereafter called N2000) k parameterization and the Takahashi et al. (2009) climatology for the whole year and for each season. The area, as a whole, is a sink of CO<sub>2</sub> (from blue to purple colored in the Fig. 1) but some regions, in some parts, close to North Atlantic Drift and East Greenland Current (Figure 2-), areis net sources (from yellow to red colored in the Fig. 1). At the seasonal maps one can see more variability affects by physical process (with temperature changes causing maximum in-water pCO<sub>2</sub> in summer) or biological activity (with phytoplankton blooms causing summer values to be lowest in the annual cycle). For example, the areas close to the North Atlantic Drift And East Greenland current are sinks of CO<sub>2</sub> in the summer (likely due to the growth of phytoplankton) while the southern most areas of the region become CO<sub>2</sub> 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 3 for the whole year and for each season (and variability in atmospheric CO<sub>2</sub> 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 wind speed  $U_{10}$  for the whole year and each season are shown in Figure 4. The wind speeds in the North Atlantic are higher than the mean value in the world ocean (which is 7 m s<sup>-1</sup>; Courtney et al., 2016), with mean values higher than 10 m s<sup>-1</sup> in many regions of the study area in all seasons except for the summer (with highest values in winter). This is important because the air-sea flux depends not only on average wind speed but also on its distribution (see also the Discussion below). This effect is especially visible between formulas with different powers of  $U_{10}$ . Figure 5 shows the difference in the air-sea  $CO_2$  fluxes calculated using two example parameterizations: one proportional to  $U_{10}^3$  (eq. 6) and one to  $U_{10}^2$  (eq. 7), namely Wanninkhof and McGillis (1999) (hereafter called WMcG1999) and Wanninkhof (2014) (hereafter called W2014). It can be seen that the "cubic" function results in higher absolute air-sea flux values when compared to the "quadratic" function in the regions of high winds, and lower absolute air-sea flux values in weaker winds.

Figure 6 shows the monthly values of air-sea CO<sub>2</sub> fluxes for the five parameterizations (eq. 4-8) for the North Atlantic and the European Arctic. The regions are sinks of CO<sub>2</sub> in every month, although

August is close to neutral for the North Atlantic. The results using cubic parameterizations (eqs. 6 and 8) are higher in absolute values, respectively by up to 30% for WMcG1999 and 55% for McGillis (2001) (hereafter called McG2001), in comparison to the "quadratic" of N2000 (eq. 4). The other two "quadratic" parameterizations W2014 and Ho et al. (2006) (hereafter called H2006) (eqs. 5 and 7) resulted in fluxes within 5% of N2000. Annual net fluxes for the North Atlantic and the European Arctic and global (included for comparison) are shown in Table 1. In addition to the five parameterizations Figure 7 presents results for both of the OceanFlux GHG Evolution formulas (using wind and radar backscatter data). The mean and standard deviations of the parameterization ensemble are shown as grey vertical lines. The standard deviation 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. Annual net fluxes for the North Atlantic, Southern and global ocean as well as for the European Arctic are shown in Table 1. The results show that the annual North Atlantic net air-sea CO<sub>2</sub> sink, depending on the formula used, varies from -0.38 Pg C for N2000 to -0.56 Pg C for McG2001. In the case of global net air-sea CO<sub>2</sub> sink the values are, respectively, -1.30 Pg C and -2.15 Pg C, respectively. Table 1 as well as Figure 7 shows the same data "normalized" to the N2000 data (divided by value), which this allows us to visualize the relative differences (in Table 1 values in parentheses). -In the case of the North Atlantic using the "quadratic" W2014 and H2006 parameterizations results in a net air-sea flux that areis, respectively, 4% and 5% higher in absolute values, respectively, than the equivalent N2000 result, while the "cubic" WMcG1999 and McG2000 results in values that are up to 28% and 44% higher, respectively, than N2000 results, for this regions. The respective values for the Arctic are 3%- for W2014 and 4% for H2006, as well as 28% for WMcG1999 and 44% for McG2001 than N2000. In the case of global net air-sea CO<sub>2</sub> fluxes the equivalent values are 8% (W2014) and 9% (H2006) higher than the N2000 result for the quadratic functions as well as 33% (WMcG1999) and 65% (McG2001) for cubic ones. The OceanFlux GHG parameterization results, for the backscatter and wind-driven versions, results in net air-sea CO<sub>2</sub> fluxes higher for North Atlantic Ocean than the N2000 that are 38% and 47%, respectively, higher for North Atlantic than the N2000 result and in the global case the values, for those two versions, were 44% and 52% higher, respectively, than N2000 results. The spread of the Arctic values was lower than that of the Atlantic ones (see Table 1). On the other hand, the values for the Southern Ocean were slightly higher than for the North Atlantic but lower than the global ones, with the exception of the OceanFlux GHG parameterizations.

All the above results were obtained with the Takahashi et al. (2009) pCO<sub>2</sub> climatology and for comparison, we have also calculated the air-sea CO<sub>2</sub> fluxes using the re-analysed SOCAT versions 1.5 and 2.0 data (which were converted to climatologies using methodology described in Goddijn-Murphy et al., 2015). Figure 8 shows the results using the N2000 k parameterization for all three of the datasets (Takahashi et al. (2009) and both SOCAT versions). In the case of the North Atlantic Ocean 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 et al. (2009)), the annual values are similar: -0.38 Pg C for both Takahashi et al. (2009) and SOCAT v1.5 and -0.41 Pg C for SOCAT v2.0. In the case of the European Arctic the situation is very different, with Takahashi et al. (2009) and SOCAT dataset derived climatologies resulting in inverse seasonal variability but with annual net air-sea CO<sub>2</sub> fluxes results that are similar: -0.102 Pg C for Takahashi et al. (2009), -0.085 Pg C for SOCAT v1.5 and -0.088 Pg C for SOCAT v2.0.

#### 4. Discussion

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 (Table 1, values in parentheses). This discrepancy is smaller than the

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9% difference identified for the global case (<u>Table 1 and Fig. 7</u>). This <u>result above</u> confirms that at present, these different parameterizations are interchangeable for the North Atlantic as this range is 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 <sup>14</sup>C inventories in the case of Wanninkhof (2014). The differences between the quadratic and cubic parameterization are large, and instead of the quadratic functions that are supported by several lines of evidence (see Garbe et. al., 2014 for discussion), the cubic function are not completely refuted by the available observation. Therefore, it is important to notice that a choice of one of the available cubic functions may leads to net air-sea CO<sub>2</sub> fluxes that are considerably larger in absolute values, by up to 33% in the North Atlantic <u>Ocean</u> and more than 50% in the global oceanglobally.

The above results imply smaller relative differences between the parameterizations in the North Atlantic Ocean than in the global oceanglobally. 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, the annual mean wind speed is 8.5 m s<sup>-1</sup> (Takahashi et al., 2009), which is lower values of wind speed than in the North Atlantic (9 m s<sup>-1</sup>) and of the CO<sub>2</sub>-sink (south of 45°) and the range of seasonal changes in the air-sea CO<sub>2</sub> fluxes are from -0.05 to +0.05 Pg C yr<sup>-1</sup> with differences between the parameterizations lower than in the North Atlantic (Le Quèrè et al., 2007; Takahashi et al., 2009). decreases significantly after 1990 with increasing wind speeds; this may cause higher CO2 concentration (and higher pCO2) in surface water due to enhanced vertical mixing of CO2-rich deep waters (Le Quèrè et al., 2007) and biological activity (seasonal changes in primary production). Takahashi et al. (2009) also indicate that the air-sea CO<sub>2</sub> fluxes difference in the Southern Ocean is strongly dependent on the choice of the gas transfer parameterizations and wind speed. Smaller differences in the North Atlantic Ocean, than in the global oceanly, are more surprising, given that at least some of the older parameterizations (e.g. W2009 or WMcG1999) were developed using a smaller range of winds than what occurs can exist in the North Atlantic. -There may be two reasons for this. First, when comparing quadratic and cubic parameterizations (Fig. 9), the cubic parameterization implies higher air-sea fluxes for high winds, whereas the quadratic ones lead to higher fluxes for weaker 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  $\Delta F$  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 winds less than  $U_x$ .  $U_x$  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 that  $U_x = 8.87$  m s<sup>-1</sup>. In fact all of the functions presented in Fig. 9 produce very similar values for  $U_x$ , all of which are close to 9 m s<sup>-1</sup>. This value is very close to average wind speed in the North Atlantic (Fig. 4). This is one of the reasons of the small relative difference in net air-sea fluxes. The spread of flux values for the Southern Ocean seems to support this conclusion, being larger than that in the North Atlantic. The Southern Ocean has on average stronger winds than the North Atlantic (including also the Arctic Seas) which seems to have the smallest spread of flux values for different parameterizations. The

other reason of smaller relative differences between the parameterizations in the North Atlantic than in the global ocean-globally, is the lack of seasonal variation in the sign of the air-sea flux. 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 adds to each other due to simultaneous changes in the sign of both fluxes itself and the U -  $U_x$  term. This effect of seasonal variation has been suggested to us based on available observations (A. Watson, University of Exeter- personal communication) but we are unaware of any paper investigating it or even describing it explicitly.

In addition to the five parameterizations described above, we calculated the air-sea fluxes using the OceanFlux GHG Evolution combined formula-which is based on knowledge that air-sea exchange is enhanced by air-entraining wave breaking and bubble-mediated transfer, especially for the less soluble gases than CO<sub>2</sub>. Goddijn-Murphy et al. (2016) assume a linear wind relationship for dimethyl sulphide (DMS) and an additional bubble-mediated term for less soluble gases, parameterized with whitecap coverage. (Goddijn-Murphy et al., 2016), which parameterises the contributions from direct and bubble mediated gas transfer into separate components. The resulting air-sea fluxes are higher in absolute terms, than all of the quadratic functions considered in this study, and are closer in value to cubic parameterization. This may mean that the bubble mediated term of Fangohr and Woolf (2007) is overestimating the bubble component, implying the need for a dedicated calibration effort. This question will be the subject of further studies in the OceanFlux GHG Evolution project.

Using both Takahashi et al. (2009) climatology and SOCAT datasets (Fig. 8) results in similar annual net air-sea CO2 fluxes in the North Atlantic; however it should be noted that they show different seasonal variations. This may have been caused by slightly different time periods of the datasets as(i.e. the SOCAT-based dataset contains more recent data). It should be noted that a significant part of the One have to remember that at present most of data from Takahashi et al. (2009) are included in SOCAT, so the differences, in the European Arctic, may be due to the sparse data the underlying sparse data coverage and possible interpolation artifacts (Goddijn-Murphy et al., 2015) or to as well as processing of the data through the FluxEngine. A recent paper (Courtney et al., 2016) using even more high latitude data than were available in the versions of SOCAT we used, shows similar seasonal pattern as SOCAT. The results are improved in Courtney et al. (2016) where modeled and observation data were compared and has been show the same relationships in high latitude zone. Still, Tthis discrepancy makes us treat the net air-sea CO<sub>2</sub> fluxes results from the Arctic with much less confidence than the values for the whole North Atlantic. It is impossible to decidee inlare withinin this study which dataset is more accurate as only new data can settle this. However, new data, not included in the SOCAT version we used, have been available to the recent analysis by Yasunaka et al. (2016). The observed in-water pCO<sub>2</sub> data (Fig. 4 in Yasunaka et al., 2016), especially since 2005, show clearly an annual cycle compatible with the SOCAT seasonal flux variability.

# 5. Conclusions

In this paper we have studied the effect of the choice of gas transfer velocity parameterization on the net  $CO_2$  air-sea gas fluxes in the North Atlantic and the 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  $CO_2$  fluxes 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 is up to 46% different for the North Atlantic, 36% for the Arctic and 65% globally when comparing cubic and quadratic functions. In both cases the uncertainty in the North Atlantic and the Arctic regions are smaller than the global case. We explain the smaller North Atlantic variability to be a combination of, firstly, higher than global average wind speeds in the North Atlantic, close to 9 m s<sup>-1</sup>, which is the wind speed at which most *k* parameterization have similar values, and secondly the all-season CO<sub>2</sub> sink conditions in most North Atlantic areas. We repeated the analysis using Takahashi et al. (2009) and SOCAT *p*CO<sub>2</sub> derived climatology and find that although the seasonal variability in the North Atlantic is different the annual net air-sea CO<sub>2</sub> fluxes are within 8% in the North Atlantic and 19% in the European Arctic. The seasonal flux calculated from the two *p*CO<sub>2</sub> datasets in the Arctic have inverse seasonal variations, indicating possible under sampling (aliasing) of the *p*CO<sub>2</sub> in this polar region and therefore highlighting the need to collect more polar *p*CO<sub>2</sub> observations in all months and seasons.

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## References

Bakker, D. C. E., Pfeil, B., Smith, K., Hankin, S., Olsen, A., Alin, S. R., Cosca, C., Harasawa, S., Kozyr, A., Nojiri, Y., O'Brien, K. M., Schuster, U., Telszewski, M., Tilbrook, B., Wada, C., Akl, J., Barbero, L., Bates, N. R., Boutin, J., Bozec, Y., Cai, W.-J., Castle, R. D., Chavez, F. P., Chen, L., Chierici, M., Currie, K., De Baar, H. J. W., Evans, W., Feely, R. A., Fransson, A., Gao, Z., Hales, B., Hardman-Mountford, N. J., Hoppema, M., Huang, W.-J., Hunt, C. W., Huss, B., Ichikawa, T., Johannessen, T., Jones, E. M., Jones, S. D., Jutterstrom, S., Kitidis, V., Kortzinger, A., Landschützer, P., Lauvset, S. K., Lefèvre, N., Manke, A. B., Mathis, J. T., Merlivat, L., Metzl, N., Murata, A., Newberger, T., Omar, A. M., Ono, T., Park, G.-H., Paterson, K., Pierrot, D., Ríos, A. F., Sabine, C. L., Saito, S., Salisbury, J., Sarma, V. V. S. S., Schlitzer, R., Sieger, R., Skjelvan, I., Steinhoff, T., Sullivan, K. F., Sun, H., Sutton, A. J., Suzuki, T., Sweeney, C., Takahashi, T., Tjiputra, J., Tsurushima, N., van Heuven, S. M. A. C., Vandemark, D., Vlahos, P., Wallace, D. W. R., Wanninkhof, R., and Watson, A. J.: An update to the Surface Ocean CO<sub>2</sub> Atlas (SOCAT version 2), Earth Syst. Sci. Data, 6: 69-90, doi:10.5194/essd-6-69-2014, 2014.

Couldrey, M. P., Oliver, K. I. C., Yool, A., Halloran, P. R., Achterberg, E. P.: On which timescale do gas transfer velocities control North Atlantic CO<sub>2</sub> flux variability?, Global Biogechem. Cycles,

Donlon, C. J., Martin, M., Stark, J., Roberts-Jones, J., Fiedler, E., and Wimmer, W.: The Operational Sea Surface Temperature and Sea Ice Analysis (OSTIA) system, Remote Sens. Environ., 116, 140-158, doi: 10.1016/j.rse.2010.10.017, 2011.

Fangohr, S. and Woolf, D. K.: Application of new parameterizations of gas transfer velocity and their impact on regional and global marine CO2 budgets, J. Marine Syst., 66, 195-203, doi:10.1016/j.jmarsys.2006.01.012, 2007.

Garbe, C. S., Rutgersson, A., Boutin, J., de Leeuw, G., Delille, B., Fairall, C. W., Gruber, N., Hare, J., Ho, D. T., Johnson, M. T., Nightingale, P. D., Pettersson, H., Piskozub, J., Sahlée, E., Tsai, W., Ward, B., Woolf, D. K., and Zappa, C. J.: Transfer across the air-sea Interface, in: Ocean-atmosphere interactions of gases and particles, edited by: Liss, P. S. and Johnson, M. T., Earth Sys. Sci., Springer, Berlin, Heidelberg, 55–111, 2014.

Goddijn-Murphy, L., Woolf, D. K., Callaghan, A. H.: Parameterizations and algorithms for oceanic whitecap coverage, J. Phys. Oceanogr., 41, 742-756, doi:10.1175/2010JPO4533.1, 2011.

Goddijn-Murphy, L. M., Woolf, D. K., and Marandino, C.: Space-based retrievals of air-sea gas transfer velocities using altimeters: Calibration for dimethyl sulfide, J. Geophys. Res., 117, C08028, doi: 10.1029/2011JC007535, 2012.

Goddijn-Murphy, L. M., Woolf, D. K., Land, P. E., Shutler J. D., Donlon, C.: The OceanFlux Greenhouse Gases methodology for deriving a sea surface climatology of CO<sub>2</sub> fugacity in support of air-sea gas lux studies, Ocean Sci., 11, 519-541, doi: 10.5194/os-11-519-2015, 2015.

Goddijn-Murphy, L., Woolf, D. K., Callaghan, A. H., Nightingale, P. D., and Shutler, J. D.: A reconciliation of empirical and mechanistic models of the air-sea gas transfer velocity, J. Geophys. Res. Oceans, 121,818-835, doi:10.1002/2015JC011096, 2016.

González-Dávila, M., Santana-Casiano, J. M., and González-Dávila, E. F.: Interannual variability of the upper ocean carbon cycle in the northeast Atlantic Ocean, Geophys. Res. Lett., 34, L07608, doi: 10.1029/2006GL028145, 2007.

Gregg, W. W., Casey, N. W., Rosseaux, C. S.: Sensitivity of simulated global ocean carbon flux estimates to forcing by reanalysis products, Ocean Model., 80, 24-35, doi: 10.1016/j.ocemod.2014.05.002, 2014.

477 Gruber, N.: Carbon cycle: Fickle trends in the ocean, Nature, 458, 155-156, doi: 10.1038/458155a, 2009.

Halloran, P. R., Booth, B. B., Jones, C. D., Lambert, F. H., McNeall, D. J., Totterdell, I. J., and Völker, C.: The mechanisms of North Atlantic CO<sub>2</sub> uptake in a large Earth System Model ensemble, Biogeosciences, 12, 4497–4508, doi: 10.5194/bg-12-4497-2015, 2015.

Ho, D. T., Law, C. S., Smith, M. J., Schlosser, P., Harvey, M., and Hill, P.: Measurements of air-sea gas exchange at high wind speeds in the Southern Ocean: Implications for global parameterizations, Geophys. Res. Lett., 33, 16611, doi: 10.1029/2006/GL026817, 2006.

Landschützer, P., Gruber, N., Bakker, D. C. E., Schuster, U., Nakaoka, S., Payne, M. R., Sasse, T. P., and Zeng, J.: A neural network-based estimate of the seasonal to inter-annual variability of the Atlantic Ocean carbon sink, Biogeosciences, 10, 7793-7815, doi: 10.5194/bg-10-7793-2013, 2013.

Landschützer, P., Gruber, N., Bakker, D. C. E., Schuster, U.: Recent variability of the global ocean

- 494 carbon sink, Global Biogeochem. Cycles, 28, 927–949, doi: 10.1002/2014GB004853, 2014.
- Le Quéré, C., Rödenbeck, C., Buitenhuis, E. T., Conway, T. J., Langenfelds, R., Gomez, A., Labuschagne, C., Ramonet, M., Nakazawa, T., Metzl, N., Gillett, N., Heimann, M.: Saturation of the Southern Ocean CO<sub>2</sub> sink due to recent climate change, Science 316, 1735-1738, doi:10.1126/science.1136188, 2007.
- Le Quéré, C., Moriarty, R., Andrew, R. M., Peters, G. P., Ciais, P., Friedlingstein, P., Jones, S. D., 501 502 Sitch, S., Tans, P., Arneth, A., Boden, T. A., Bopp, L., Bozec, Y., Canadell, J. G., Chini, L. P., 503 Chevallier, F., Cosca, C. E., Harris, I., Hoppema, M., Houghton, R. A., House, J. I., Jain, A. K., 504 Johannessen, T., Kato, E., Keeling, R. F., Kitidis, V., Klein Goldewijk, K., Koven, C., Landa, C. 505 S., Landschützer, P., Lenton, A., Lima, I. D., Marland, G., Mathis, J. T., Metzl, N., Nojiri, Y., Olsen, A., Ono, T., Peng, S., Peters, W., Pfeil, B., Poulter, B., Raupach, M. R., Regnier, P., 506 507 Rödenbeck, C., Saito, S., Salisbury, J. E., Schuster, U., Schwinger, J., Séférian, R., Segschneider, J., Steinhoff, T., Stocker, B. D., Sutton, A. J., Takahashi, T., Tilbrook, B., van der Werf, G. R., 508 509 Viovy, N., Wang, Y.-P., Wanninkhof, R., Wiltshire, A., and Zeng, N.: Global carbon budget 2014, 510 Earth Syst. Sci. Data, 7, 47–85, doi: 10.5194/essd-7-47-2015, 2015.
- Lefèvre, N., Watson, A. J., Olsen, A., Rios, A. F., Perez, F. F., Johannessen, T.: A decrease in the sink for atmospheric CO<sub>2</sub> in the North Atlantic, Geophys. Res. Lett., 31, L07306, doi: 10.1029/2003GL018957, 2004.
- McGillis, W. R., and Edson, J. B., Hare, J. E., Fairall, C. W.: Direct covariance air-sea CO<sub>2</sub> fluxes, J. Geophys. Res., 106, C8 16729-16745, 2001.
- Merchant, C. M., Embury, O., Rayner, N. A., Berry, D. I., Corlett, G. K., Lean, K., Veal, K. L., Kent,
  E. C., Llewellyn-Jones, D. T., Remedios, J. J., and Saunders, R.: A 20 year independent record of
  sea surface temperature for climate from Along-Track Scanning Radiometers, J. Geophys. Res.,
  117, C12, doi:10.1029/2012JC008400, 2012.
- Nightingale, P. D., Malin, G., Law, C. S., Watson, A. J., Liss, P. S., Liddicoat, M. I., Boutin, J., and Upstill-Goddard, R. C.: In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, Global Biogeochem. Cycles, 14, 373-387, 2000.
- Nightingale, P. D., Relationship between wind speed and gas exchange over the ocean: which parameterisation should I use? Report from Discussion Session at SOLAS Open Science conference in Kiel, <a href="http://goo.gl/TrMQkg">http://goo.gl/TrMQkg</a>, 2015.
- Orr, J. C., Maier-Reimer, E., Mikolajewicz, U., Monfray, P., Sarmiento, J. L., Toggweiler, J. R., Taylor, N. K., Palmer, J., Gruber, N., Sabine, C. L., Le Quéré, C., Key, R. M., Boutin, J.: Estimates of anthropogenic carbon uptake from four three-dimensional global ocean models, Global Biogeochem. Cycles, 15, 43-60, doi: 10.1029/2000GB001273, 2001.
- Pérez, F. F., Mercier, H., Vázquez-Rodríguez, M., Lherminier, P., Velo, A., Pardo, P. C., Rosón, G., and Ríos, A. F.: Atlantic Ocean CO<sub>2</sub> uptake reduced by weakening of the meridional overturning circulation, Nat. Geosci., 6, 146-152, doi: 10.1038/NGEO1680, 2013.
- Pfeil, B., Olsen, A., Bakker, D. C. E., Hankin, S., Koyuk, H., Kozyr, A., Malczyk, J., Manke, A.,
  Metzl, N., Sabine, C. L., Akl, J., Alin, S. R., Bates, N., Bellerby, R. G. J., Borges, A., Boutin, J.,
  Brown, P. J., Cai, W.-J., Chavez, F. P., Chen, A., Cosca, C., Fassbender, A. J., Feely, R. A.,
  González-Dávila, M., Goyet, C., Hales, B., Hardman-Mountford, N., Heinze, C., Hood, M.,

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511

515

518

523

527

536

- 545 Hoppema, M., Hunt, C. W., Hydes, D., Ishii, M., Johannessen, T., Jones, S. D., Key, R. M.,
- Körtzinger, A., Landschützer, P., Lauvset, S. K., Lefèvre, N., Lenton, A., Lourantou, A., 546
- 547 Merlivat, L., Midorikawa, T., Mintrop, L., Miyazaki, C., Murata, A., Nakadate, A., Nakano, Y.,
- Nakaoka, S., Nojiri, Y., Omar, A. M., Padin, X. A., Park, G.-H., Paterson, K., Perez, F. F., Pierrot, 548
- 549 D., Poisson, A., Ríos, A. F., Santana-Casiano, J. M., Salisbury, J., Sarma, V. V. S. S., Schlitzer, R., Schneider, B., Schuster, U., Sieger, R., Skjelvan, I., Steinhoff, T., Suzuki, T., Takahashi, T., 550
- 551 Tedesco, K., Telszewski, M., Thomas, H., Tilbrook, B., Tjiputra, J., Vandemark, D., Veness, T.,
- 552 Wanninkhof, R., Watson, A. J., Weiss, R., Wong, C. S., and Yoshikawa-Inoue, H.: A uniform,
- 553 quality controlled Surface Ocean CO<sub>2</sub> Atlas (SOCAT), Earth Syst. Sci. Data, 5, 125-143, doi:
- 554 10.5194/essd-5-125-2013, 2013.
- 555 556 Sabine, C. L., Hankin, S., Koyuk, H., Bakker, D. C. E., Pfeil, B., Olsen, A., Metzl, N., Kozyr, A., Fassbender, A., Manke, A., Malczyk, J., Akl, J., Alin, S. R., Bellerby, R. G. J., Borges, A., 557
- Boutin, J., Brown, P. J., Cai, W.-J., Chavez, F. P., Chen, A., Cosca, C., Feely, R. A., González-558 559 Dávila, M., Goyet, C., Hardman-Mountford, N., Heinze, C., Hoppema, M., Hunt, C. W., Hydes,
- 560 D., Ishii, M., Johannessen, T., Key, R. M., Körtzinger, A., Landschützer, P., Lauvset, S. K.,
- Lefèvre, N., Lenton, A., Lourantou, A., Merlivat, L., Midorikawa, T., Mintrop, L., Miyazaki, C., 561
- 562 Murata, A., Nakadate, A., Nakano, Y., Nakaoka, S., Nojiri, Y., Omar, A. M., Padin, X. A., Park,
- 563 G.-H., Paterson, K., Perez, F. F., Pierrot, D., Poisson, A., Ríos, A. F., Salisbury, J., Santana-
- 564 Casiano, J. M., Sarma, V. V. S. S., Schlitzer, R., Schneider, B., Schuster, U., Sieger, R., Skjelvan,
- 565 I., Steinhoff, T., Suzuki, T., Takahashi, T., Tedesco, K., Telszewski, M., Thomas, H., Tilbrook, B.,
- 566 Vandemark, D., Veness, T., Watson, A. J., Weiss, R., Wong, C. S., and Yoshikawa-Inoue, H.:
- Surface Ocean CO<sub>2</sub> Atlas (SOCAT) gridded data products, Earth Syst. Sci. Data, 5, 145-153, doi: 567
- 568 10.5194/essd-5-145-2013, 2013.

572

578

583

- 570 Schuster, U., and Watson, A. J.: A variable and decreasing sink for atmospheric CO<sub>2</sub> in the North 571 Atlantic, J. Geophys. Res., 112, C11006, doi: 10.1029/2006JC003941, 2007.
- 573 Schuster, U., McKinley, G. A., Bates, N., Chevallier, F., Doney, S. C., Fay, A. R., González-Dávila, M., Gruber, N., Jones, S., Krijnen, J., Landschützer, P., Lefèvre, N., Manizza, M., Mathis, J., 574
- 575 Metzl, N., Olsen, A., Rios, A. F., Rödenbeck, C., Santana-Casiano, J. M., Takahashi, T.,
- 576 Wanninkhof, R., and Watson, A. J.: An assessment of the Atlantic and Arctic sea-air CO<sub>2</sub> fluxes,
- 577 1990–2009, Biogeosciences, 10, 607–627, doi: 10.5194/bg-10-607-2013, 2013.
- 579 Shutler, J. D., Piolle, J-F., Land, P. E., Woolf, D. K., Goddijn-Murphy, L., Paul, F., Girard-Ardhuin, 580 F., Chapron, B., and Donlon, C. J.: FluxEngine: a flexible processing system for calculating airsea carbon dioxide gas fluxes and climatologies, J. Atmos. Ocean. Tech., doi:10.1175/JTECH-D-581
- 582 14-00204.1, 2016.
- 584 Sweeney, C., Gloor, E., Jacobson, A. R., Key, R. M., McKinley, G., Sarmiento, J. L. and Wanninkhof, R.: Constraining global air-sea gas exchange for CO<sub>2</sub> with recent bomb 14C 585
- 586 measurements, Global Biogeochem. Cycles, 21, doi:10.1029/2006GB002784, 2007. 587
- 588 Takahashi, T., Sutherland, S. C., Sweeney, C., Poisson, A., Metzl, N., Tilbrook, B., Bates, N., 589 Wanninkhof, R., Feely, R. A., Sabine, C., Olafsson, J., and Nojiri, Y.: Global sea-air CO<sub>2</sub> flux
- 590 based on climatological surface ocean pCO<sub>2</sub>, and seasonal biological and temperature effects,
- 591 Deep-Sea Res., Pt. II, 49, 1601-1622, 2002.
- 593 Takahashi, T., Sutherland, S. C., Wanninkhof, R., Sweeney, C., Feely, R. A., Chipman, D. W., Hales,
- B., Friederich, G., Chavez, F., Sabine, C., Watson, A., Bakker, D. C. E., Schuster, U., Metzl, N., 594
- Yoshikawa-Inoue, H., Ishii, M., Midorikawa, T., Nojiri, Y., Körtzinger, A., Steinhoff, T., 595

- Hoppema, M., Olafsson, J., Arnarson, T. S., Tilbrook, B., Johannessen, T., Olsen, A., Bellerby, R., Wong, C. S., Delille, B., Bates, N. R., and de Baar, H. J. W.: Climatological mean and decadal change in surface ocean *p*CO<sub>2</sub> and net sea-air CO<sub>2</sub> flux over the global oceans, Deep-Sea Res. Pt. II, 56, 554–577, doi: 10.1016/j.dsr2.2008.12.009, 2009.
- Talley, L. D.: Closure of the global overturning circulation through the Indian, Pacific, and Southern Oceans: schematics and transports, Oceanography 26(1), 80–97, doi:10.5670/oceanog.2013.07, 2013.
- Thomas, H., Friederike Prowe, A. E., Lima, I. D., Doney, S. C., Wanninkhof, R., Greatbatch, R. J., Schuster, U., and Corbière, A.: Changes in the North Atlantic Oscillation influence CO<sub>2</sub> uptake in the North Atlantic over the past 2 decades, Global Biogeochem. Cycles, 22, GB4027, doi:10.1029/2007GB003167, 2008.
- Wanninkhof, R.: Relationship between wind speed and gas exchange over the ocean revisited, Limnol. Oceanogr. Methods, 12, 351–362, doi: 10.4319/lom.2014.12.351, 2014.
- Wanninkhof, R., and McGillis, W. R.: A cubic relationship between air-sea CO<sub>2</sub> exchange and wind speed, Geophys. Res. Lett., 26, 1889-1892, 1999.
- Wanninkhof, R., Park, G.-H., Takahashi, T., Sweeney, C., Feely, R., Nojiri, Y., Gruber, N., Doney, S.
  C., McKinley, G. A., Lenton, A., Le Quéré, C., Heinze, C., Schwinger, J., Graven, H.,
  Khatiwala, S.: Global ocean carbon uptake: magnitude, variability and trends, Biogeosciences,
  10, 1983-2000, doi: 10.5194/bg-10-1983-2013, 2013.
- Watson, A. J., Schuster, U., Bakker, D. C. E., Bates, N. R., Corbière, A., González-Dávila, M.,
  Friedrich, T., Hauck, J., Heinze, C., Johannessen, T., Körtzinger, A., Metzl, N., Olafsson, J.,
  Olsen, A., Oschlies, A., Padin, X. A., Pfeil, B., Santana-Casiano, J. M., Steinhoff, T., Telszewski,
  M., Rios, A. F., Wallace, D. W., Wanninkhof, R.: Tracking the variable North Atlantic sink for
  atmospheric CO<sub>2</sub>, Science, 326(5958), 1391-1393, doi: 10.1126/science.1177394, 2009.
- Watson, A. J., Metzl, N., Schuster, U.: Monitoring and interpreting the ocean uptake of atmospheric CO<sub>2</sub>, Philos. T. R. Soc. A, 369, 1997–2008, doi: 10.1098/rsta.2011.0060, 2011.
- Woolf, D. K.: Parameterization of gas transfer velocities and sea-state dependent wave breaking. Tellus B, 57, 87–94, 2005.
- Woolf, D. K., Shutler, J. D., Goddijn-Murphy, L., Donlon, C. J., Nightingale, P. D., Land, P. E., Torres, R., Chapron, B., Piolle, J-F., Herledan, S., Hanafin, J., Girard-Ardhuin, F., Ardhuin, F., Prytherch, J., Moat, B., and Yelland, M.: Key uncertainties in the contemporary air-sea flux of carbon dioxide: an OceanFlux study, submitted 2015a.
- Woolf, D. K., Goddijn-Murphy, L. M., Shutler, J. D., Land, P. E., Donlon, C. J., Prytherch, J., Yelland, M. J., Nightingale, P. D., Torres, R., Chapron, B., Piolle, J-F., Herledan, S., Hanafin, J., Girard-Ardhuin, F., Ardhuin F., and Moat, B.: Sources and types of uncertainty in the contemporary air-sea flux of carbon dioxide: an OceanFlux study, submitted 2015b.
- Yasunaka, S., Murata, A., Watanabe, E., Chierici, M., Fransson, A., van Heuven, S., Hoppema, M., Ishii, M., Johannessen, T., Kosugi, N., Lauvset, S. K., Mathis, J. T., Nishino, S., Omar, A. M., Olsen, A., Sasano, D., Takahashi, T., Wanninkhof, R.: Mapping of the air–sea CO<sub>2</sub> flux in the Arctic Ocean and its adjacent seas: Basin-wide distribution and seasonal to interannual

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647 | variability, Polar Sci., 1–12, doi:10.1016/j.polar.2016.03.006, 2016.

- Figure 1. Seasonal and annual mean air-sea fluxes of CO<sub>2</sub> (mg C m<sup>-2</sup> day<sup>-1</sup>) in the North Atlantic,
- using Nightingale et al. (2000) k parameterization and Takahashi et al. (2009) climatology 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.
- 652
- Figure 2. Surface ocean currents in the Arctic (sources:
- 654 http://www.grida.no/graphicslib/detail/ocean-currents-and-sea-ice-extent\_4aa6, author: Philippe
- Rekacewicz, UNEP-GRID,—Arendal, Norway). North Atlantic Drift forming the Norwegian
- 656 Atlantic Current in the Arctic Ocean.

- Figure 3. Seasonal and annual  $pCO_2$  values ( $\mu$ atm) in surface waters of the North Atlantic, estimated using the Takahashi et al. (2009) climatology a) annual, b) DJF (winter), c) MAM
- 660 (spring), d) JJA (summer), e) SON (autumn). The gaps (white areas) are due to missing data, land
- and ice masks.

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- Figure 4. Wind speed distribution  $U_{10}$  (ms<sup>-1</sup>) in the North Atlantic used to determine the relationship between gas transfer velocity and air-sea CO<sub>2</sub> fluxes 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
- 666 masks.

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- Figure 5. Differences maps for the air-sea CO<sub>2</sub> fluxes (mg C m<sup>-2</sup> day<sup>-1</sup>) in the North Atlantic,
- between a cubed and a squared parameterization (Wanninkhof and McGillis 1999 and Wanninkhof
- 670 2014) a) annual, b) DJF (winter), c) MAM (spring), d) JJA (summer) e) SON (autumn). The gaps
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- Figure 6. Monthly values of CO<sub>2</sub> air-sea fluxes (Pg month<sup>-1</sup>) for the five parameterizations (eq. 4-8)
- a) the North Atlantic, b) the European Arctic.

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- 676 Figure 7. Annual air-sea fluxes of CO<sub>2</sub> for the five (eq. 4-8) parameterizations as well as for
- backscatter (default) and wind driven OceanFlux GHG parameterizations normalized to flux values
- of Nightingale et al. (2000) k parameterization (see text) a) globally, b) the North Atlantic c) the
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- deviations are marked as vertical gray lines.

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- Figure 8. Comparison of monthly air-sea CO<sub>2</sub> fluxes calculated with different pCO<sub>2</sub> datasets
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- al., 2000) a) the North Atlantic, b) the European Arctic.

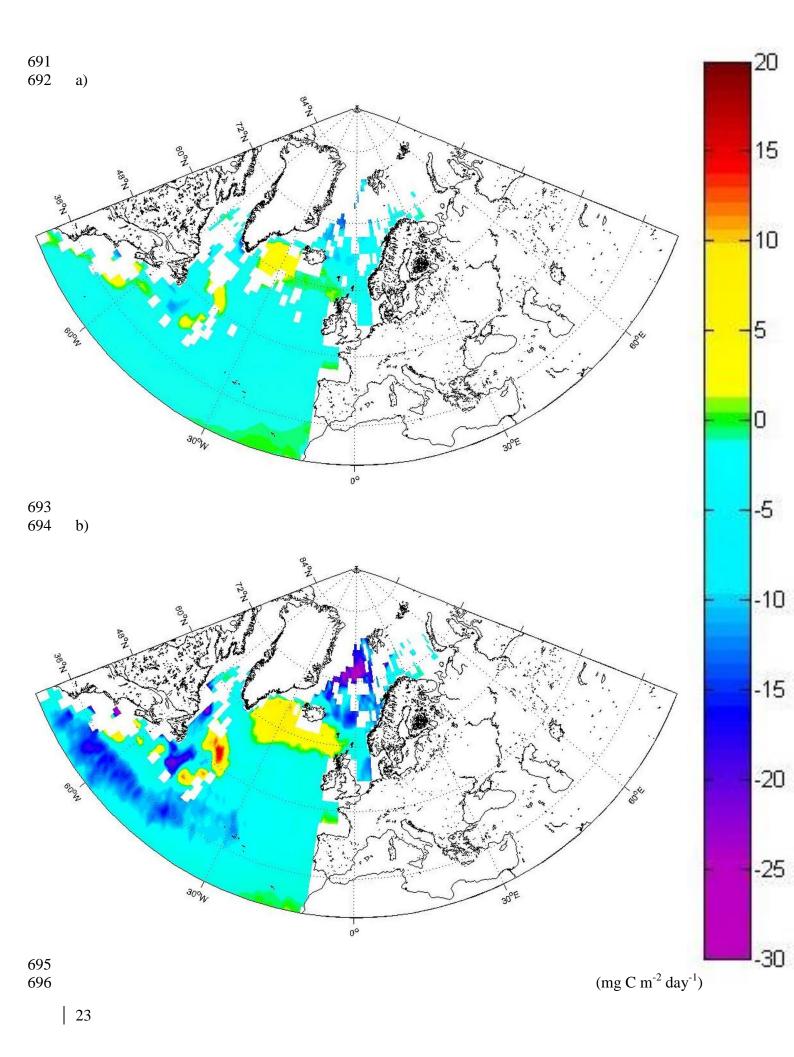
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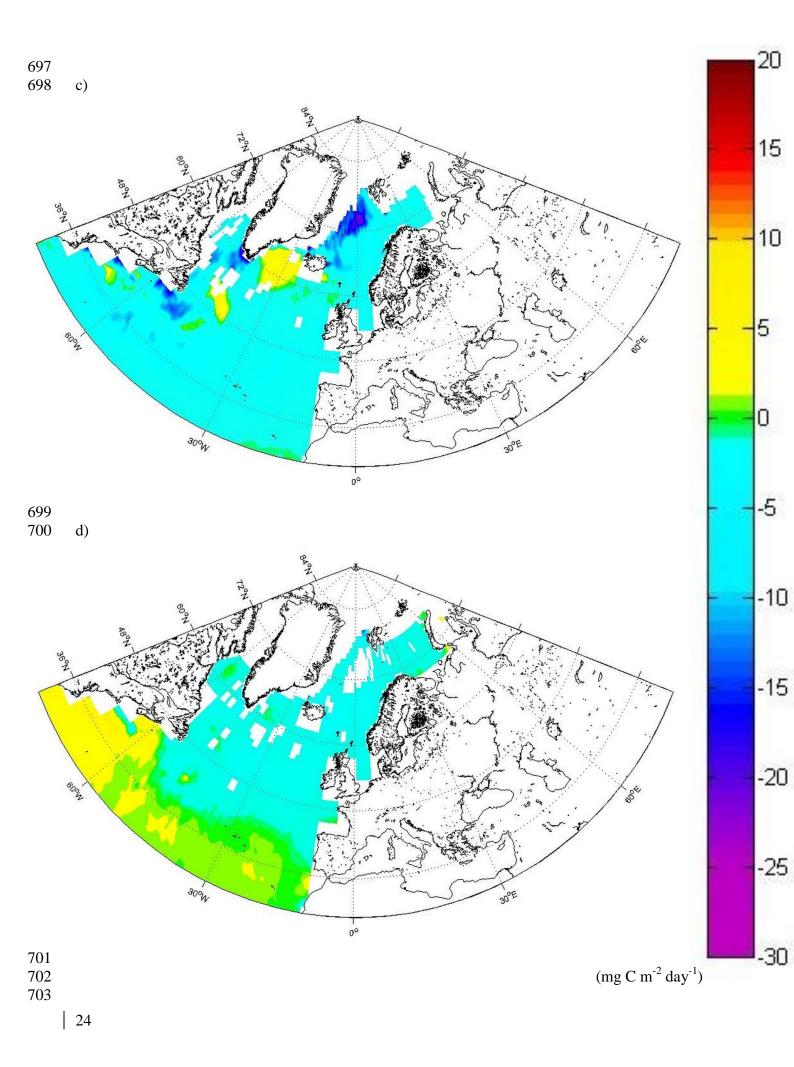
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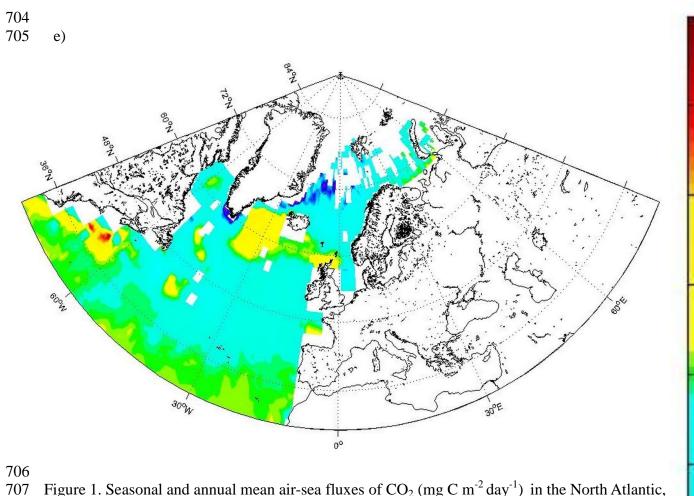
Figure 9. Different k660 parameterizations as a function of wind speed.

Table 1. Annual air-sea  $CO_2$  fluxes (in Pg) using different k parameterizations. The values in parentheses are fluxes normalized to Nightingale et al., 2000 (as in Fig. 7)

	Global	Arctic	North Atlantic	Southern Ocean
Nightingale et al., 2000	-1.30	-0.102	-0.382	-0.72
	(1.00)	(1.00)	(1.00)	(1.00)
Ho et al., 2006	-1.42	-0.106	-0.402	-0.76
	(1.09)	(1.04)	(1.05)	(1.06)
Wanninkhof and McGillis, 1999	-1.73 (1.33)	- <u>10</u> .130 (1.28)	-0.490 (1.29)	-0.93 (1.30)
Wanninkhof, 2014	-1.40	-0.105	-0.398	-0.76
	(1.08)	(1.03)	(1.04)	(1.05)
McGillis et al., 2001	-2.15	-0.147	-0.557	-1.08
	(1.65)	(1.44)	(1.46)	(1.49)
OceanFlux GHG wind driven	-1.98	-0.138	-0.560	-1.14
	(1.52)	(1.36)	(1.47)	(1.58)
OceanFluxGHG backscatter	-1.88	-0.130	-0.526	-1.09
	(1.44)	(1.27)	(1.38)	(1.51)







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 $(mg C m^{-2} day^{-1})$ 

Figure 1. Seasonal and annual mean air-sea fluxes of  $CO_2$  (mg C m<sup>-2</sup> day<sup>-1</sup>) in the North Atlantic, using Nightingale et al. (2000) k parameterization and the Takahashi et al. (2009) climatology 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.

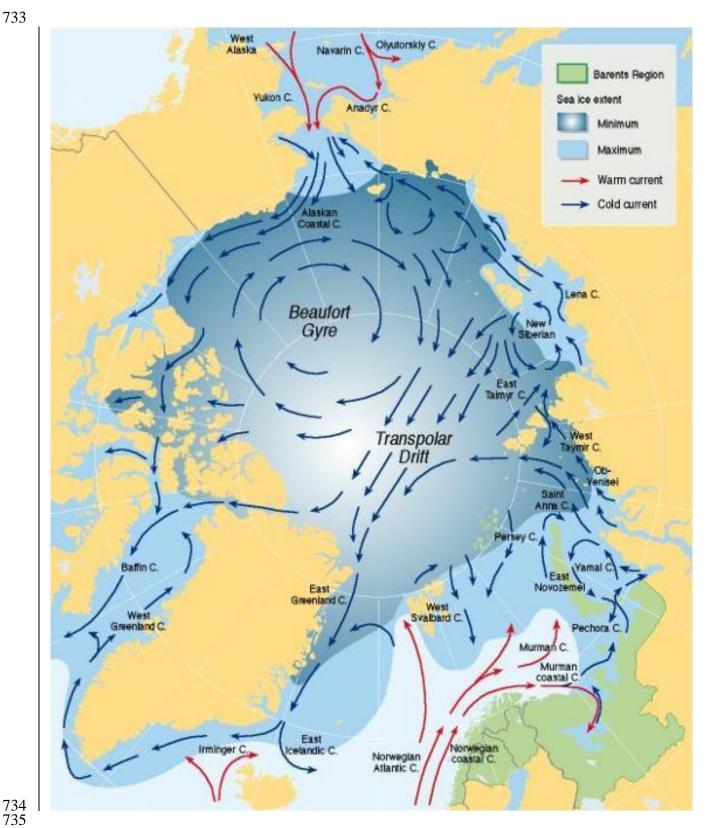
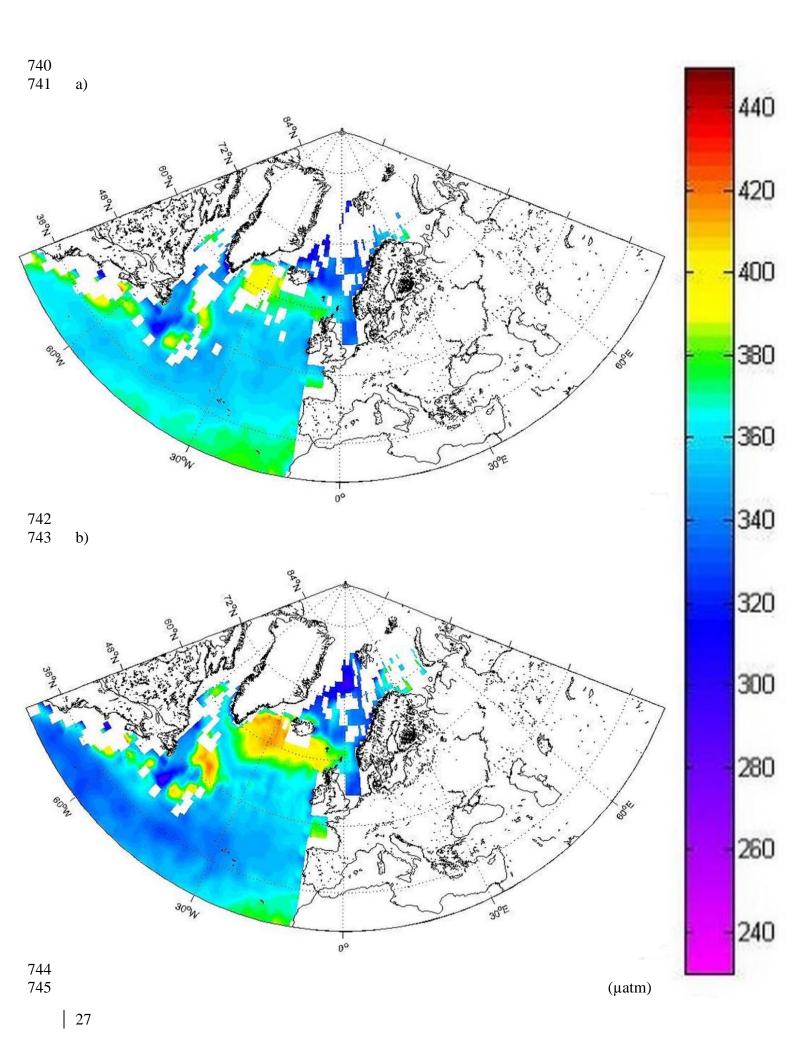
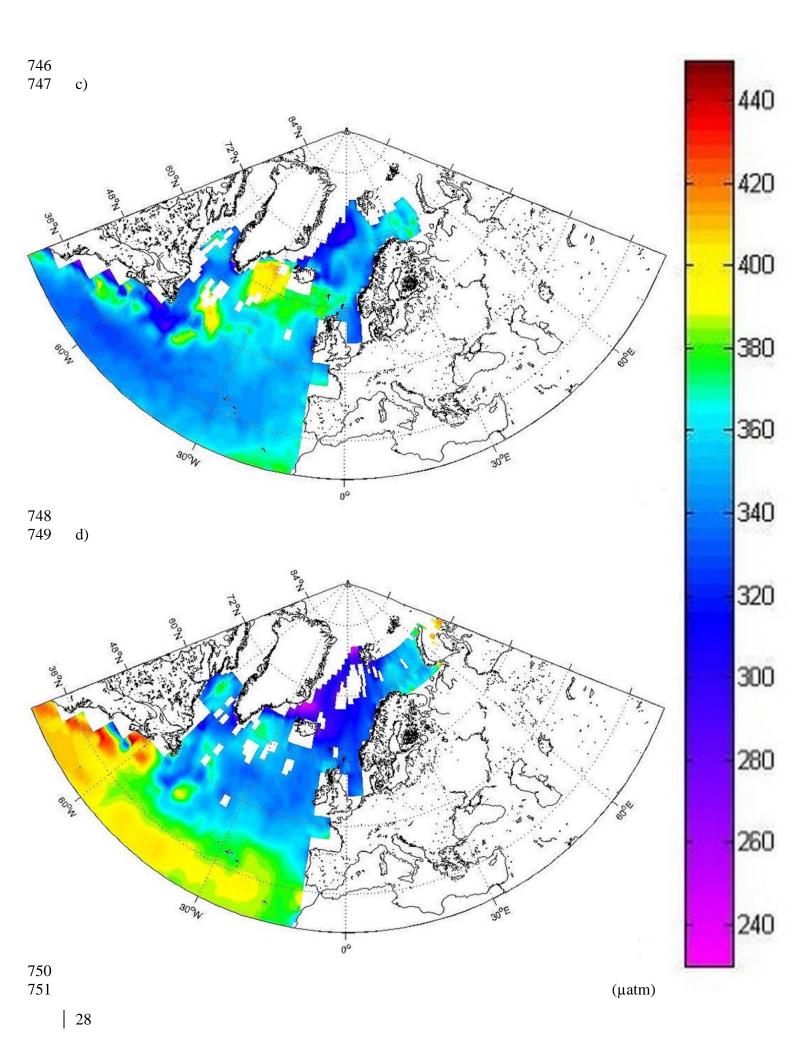


Figure 2. Surface ocean currents in the Arctic (sources: http://www.grida.no/graphicslib/detail/ocean-currents-and-sea-ice-extent\_4aa6, author: Philippe Rekacewicz, UNEP-GRID,- Arendal, Norway). North Atlantic Drift forming the Norwegian Atlantic Current in the Arctic Ocean.

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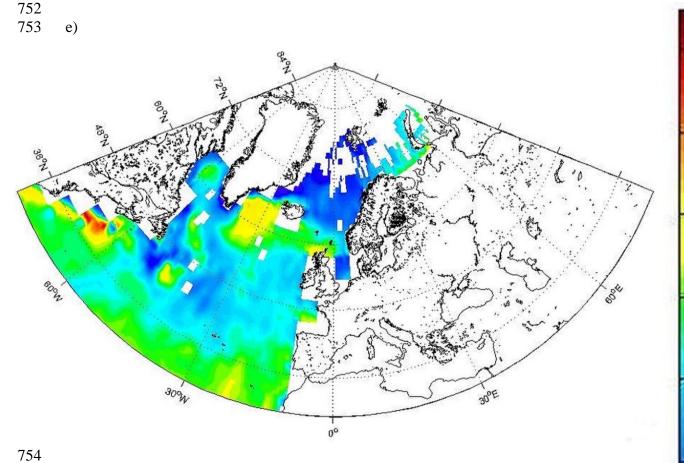
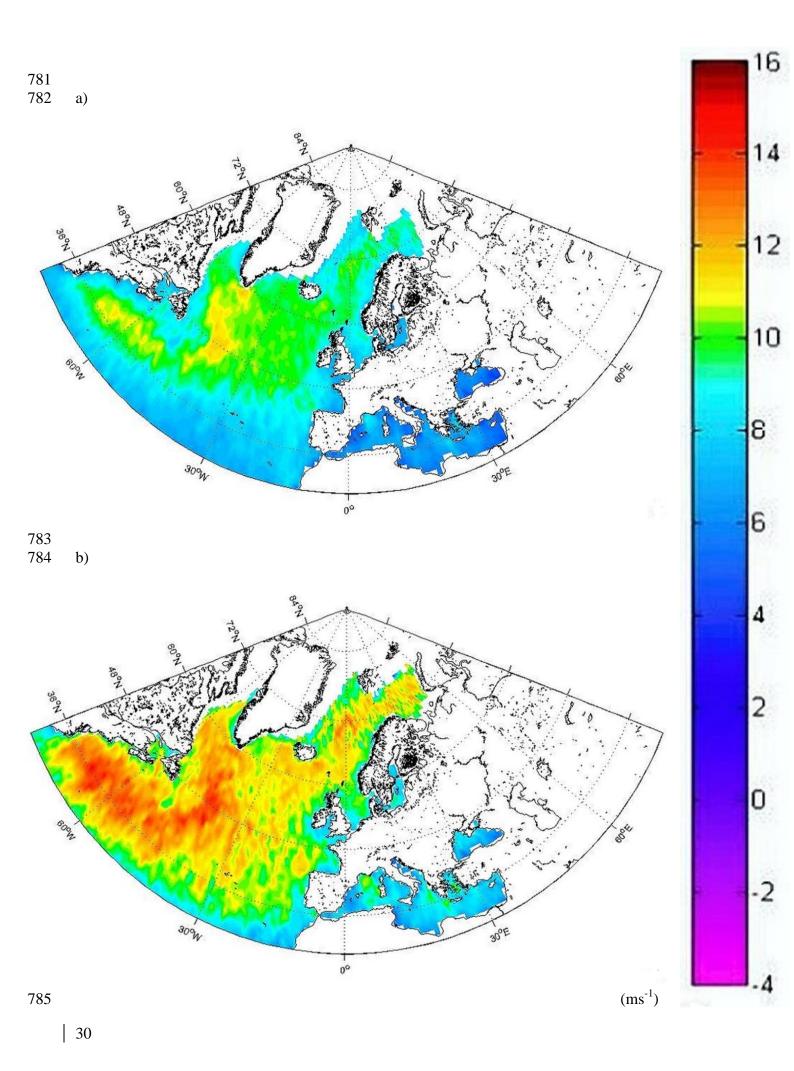
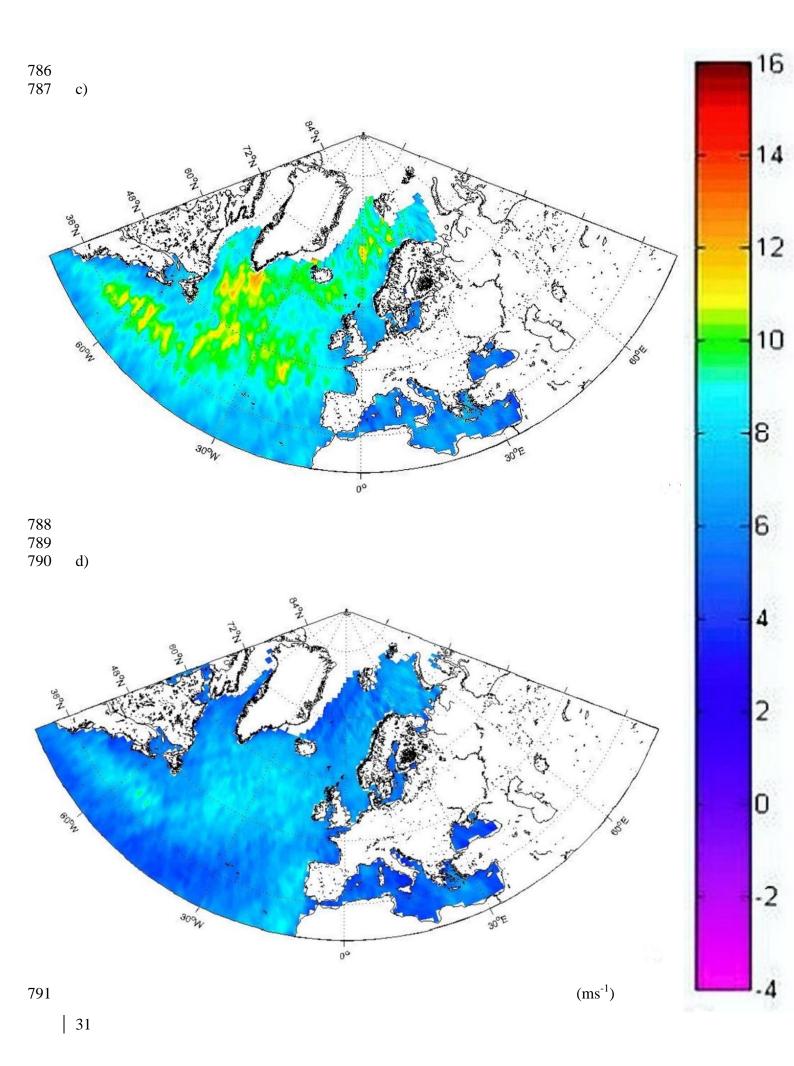
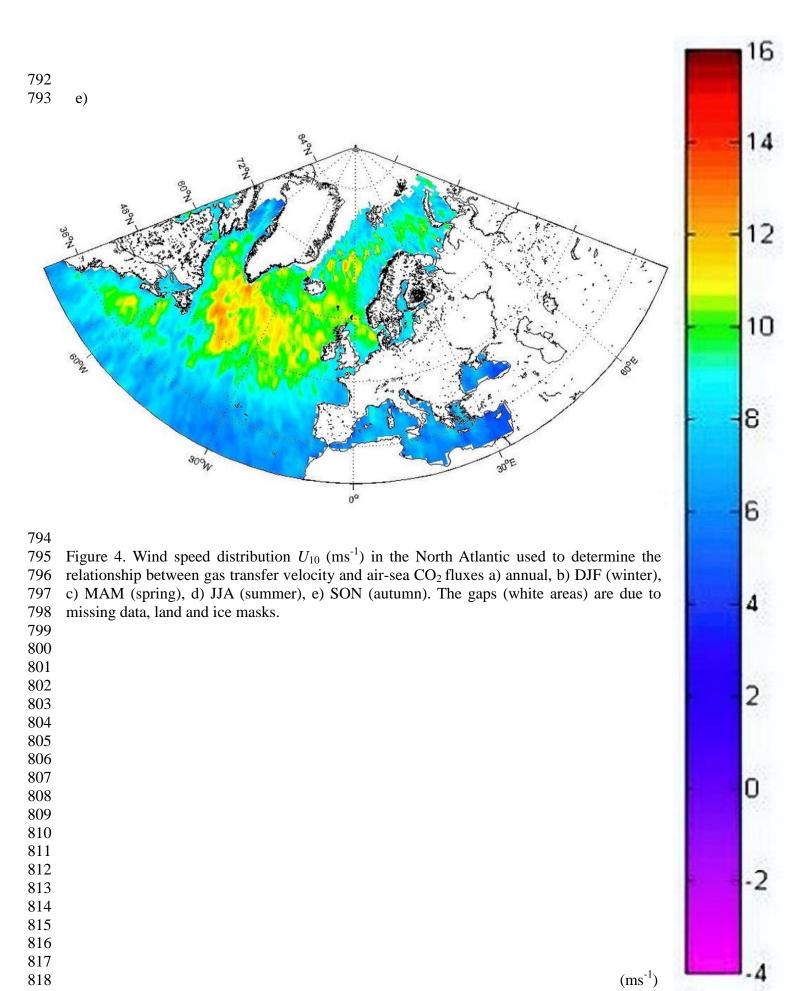


Figure 3. Seasonal and annual  $pCO_2$  values ( $\mu$ atm) in surface waters of the North Atlantic, estimated using Takahashi et al. (2009) climatology a) annual, b) DJF (winter), c) MAM (spring), d) JJA (summer), e) SON (autumn). The gaps (white areas) are due to missing data, land and ice masks.

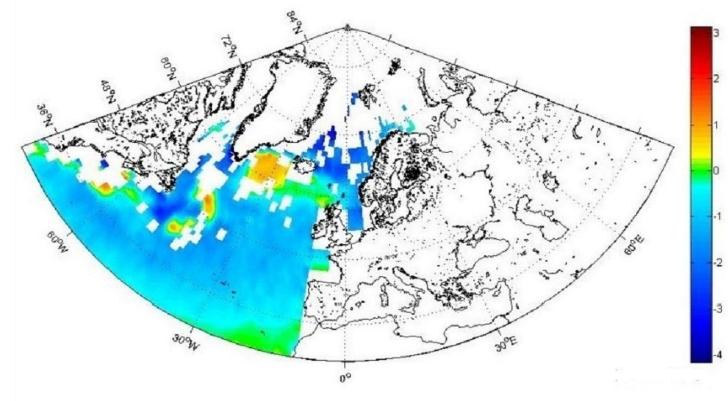




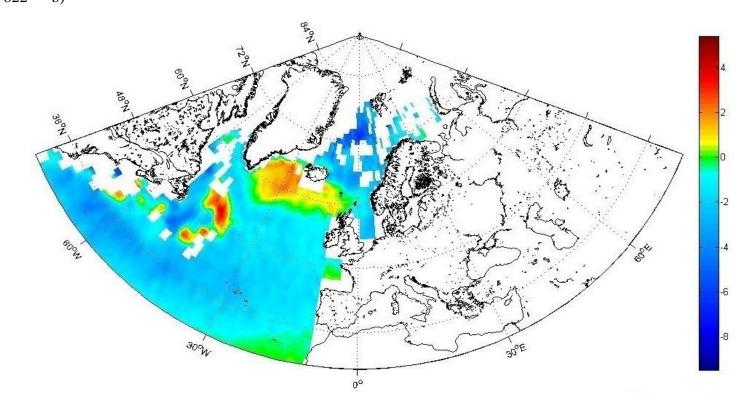




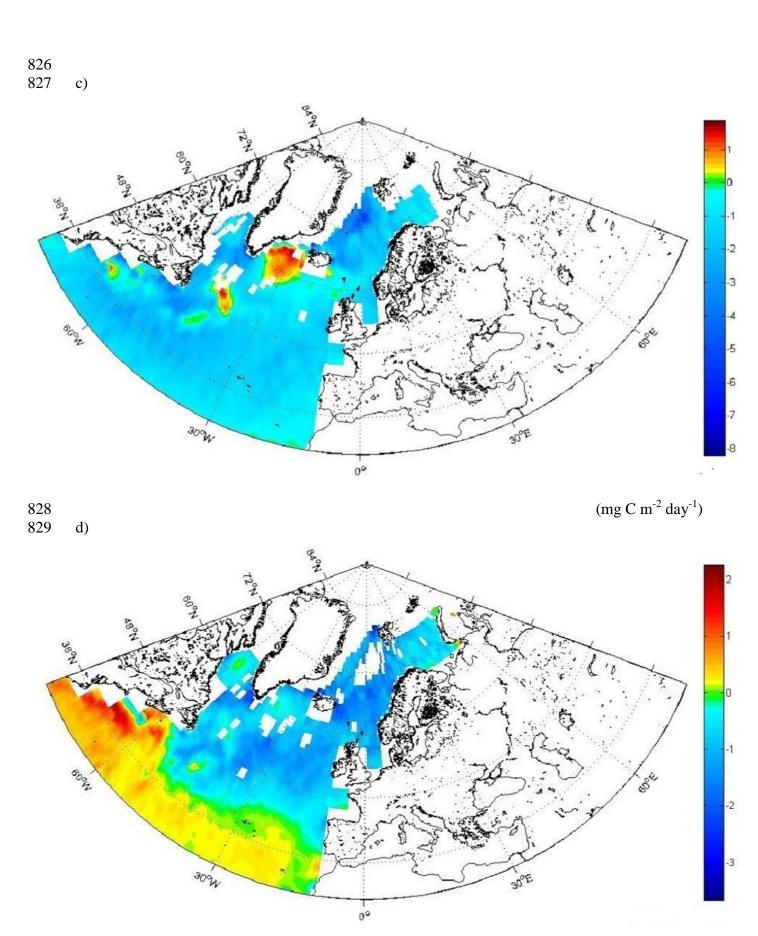
819 820 a)



 $(mg C m^{-2} day^{-1})$ 821 822 b)



823 824 825 (mg C m<sup>-2</sup> day<sup>-1</sup>)



(mg C m<sup>-2</sup> day<sup>-1</sup>) 831 832



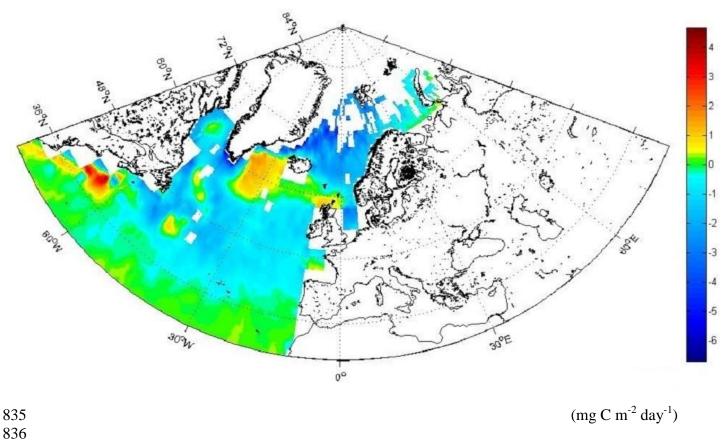
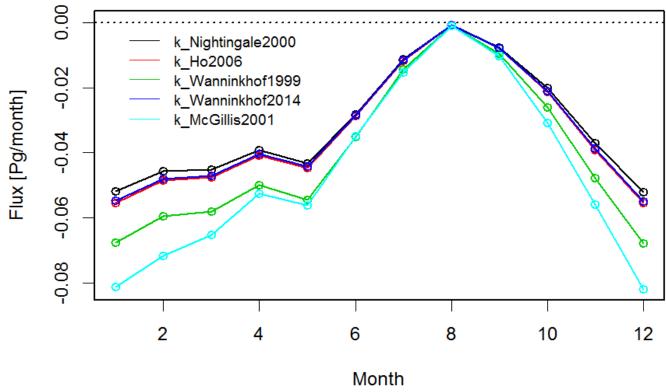


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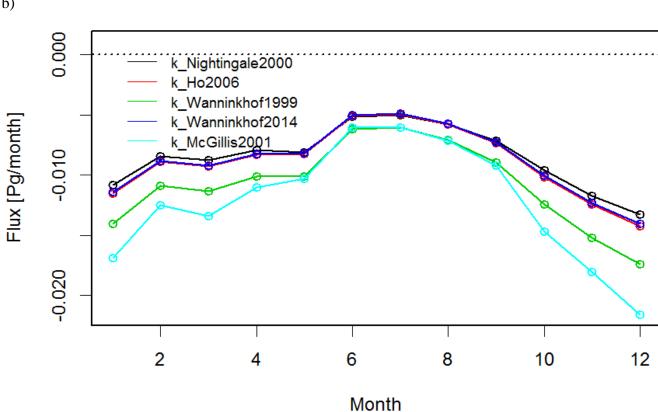
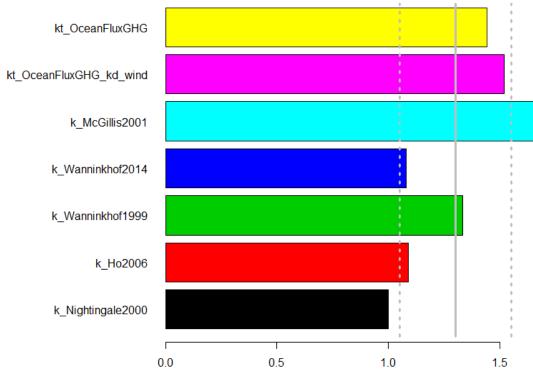


Figure 6. Monthly values of  $CO_2$  air-sea fluxes (Pg/month) for the five parameterizations (eq. 4-8) a) the North Atlantic, b) the European Arctic.

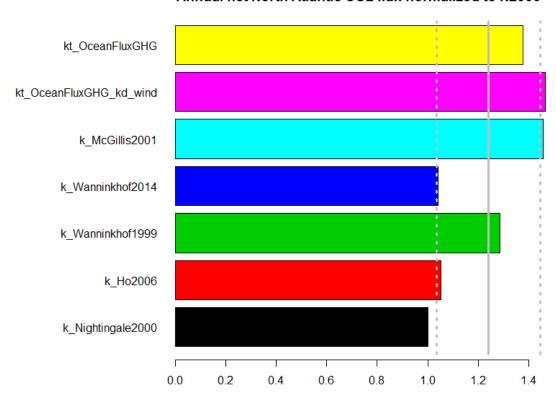
850 a)

# Annual net global CO2 flux normalized to N2000

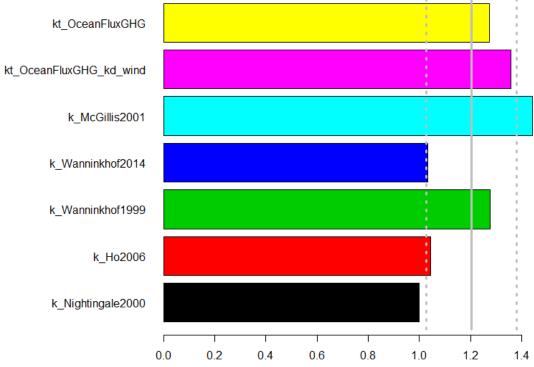


853 b)

## Annual net North Atlantic CO2 flux normalized to N2000



#### Annual net Arctic CO2 flux normalized to N2000



860 d)

#### Annual net Southern Ocean CO2 flux normalized to N2000

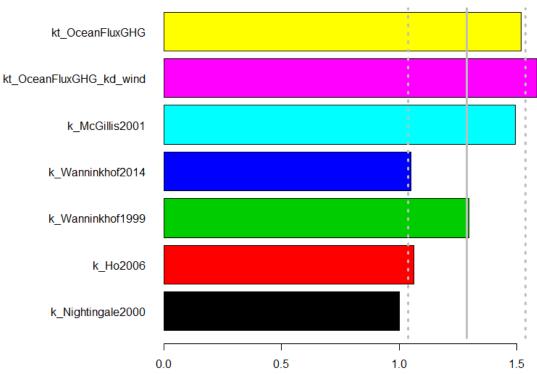
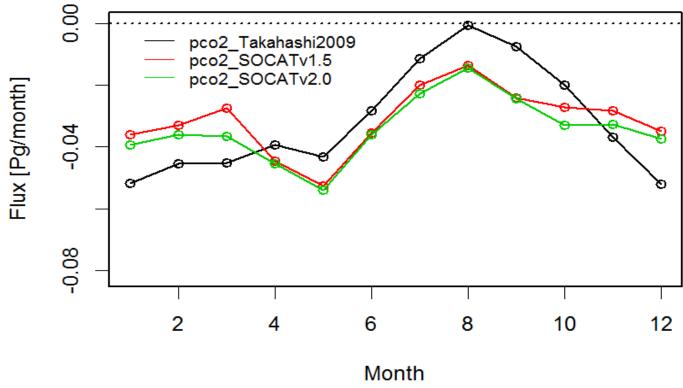


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869 870 b)

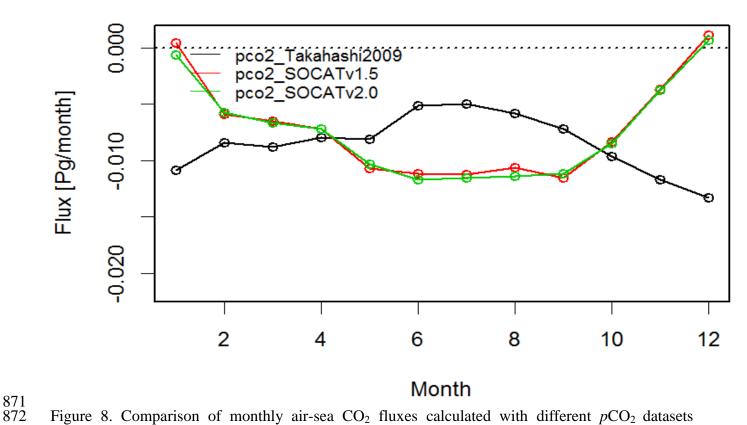
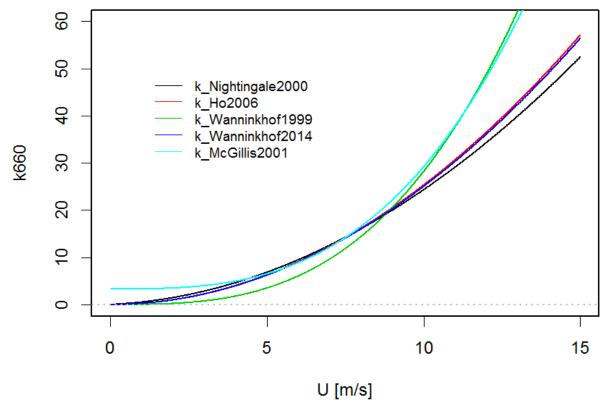


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878 Figure 9. Different k660 parameterizations as a function of wind speed.