Dear Topic Editor,

Thank you very much for all your comments and suggestions. We gave our manuscript for correction of the language and each time, before send it back to the reviewers, checked carefully and make sure that everything is correct. However, with all our commitment and attention we are aware that in some places we can miss language mistake, which is a big problem for us. We agree that even this time we also have made every effort and all our attention when checking the article in terms of language, because this is very important for us that our manuscript has been published. Below is a list of our correctness. All the mistakes and omissions have been fixed by us.

Below is my list of critiques, comments and suggestions. Unfortunately there were still many instances where the language needs improving. I have done my best, but there are more places. The **reasoning in the texts is sometimes not exact enough**. You should write in a way that the reader can always follow what exactly you are explaining. The number of errors (for example in the references) and inconsistencies gives a sloppy impression.

Thank you for the correctness. We also do our best, the text was checking by a native speaker, our colleague as you know, and by two other people, so we really don't know why so many gaps and language mistakes are still in there. We checked it again and believe that there are no more errors.

I noticed that there are (few?) differences between the manuscript versions (marked up and new version) you submitted. I hope there were really only few... Please explain. Sorry about that. Some of the changes were suggested by Jamie Shutler (the PI of the ESA project) who was used as a native speaker (the best one we could use). We informed you about this in the previous iteration of this editorial process. Indeed, after so many iterations, sometimes, it is difficult for us to know what has been changed in which of them. When we update our manuscript last time we must forgot to also update the mark up version. But right now both version are uniform.

In this study you discuss the uncertainty due to different parameterizations. However, the informed reader knows that generally there is a relatively large uncertainty in computing air-sea fluxes due to other factors as well. It is important to know how large the uncertainties are that you discuss in comparison to those others. Please add text to this issue.

This has been answered (affirmatively) under the next comment.

L56 The global interannual air-sea CO_2 fluxes variability can be vary about 60% due to different in pCO_2 and 35% by *k* (Courtney et al., 2016).

L65-71 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 choice of parameterization of wind speed may cause a difference in the results of *k*, even about 50% (Gregg et al., 2014; Couldrey et al., 2016). In this work we analyze solely the effect of choice between various published empirical wind driven gas transfer parameterizations. 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).

Also I am not quite satisfied with your response to a major critique by referee #1 (reproduced below). You'll have to discuss this in the manuscript, and possibly also the outcomes of the papers by Woolf et al.

----- Ref #1

Many of the mentioned gas transfer formulations have been developed for different wind products (e.g. NCEP, CCMP), whereas the authors only use one wind product. There are some major differences between wind products. I am not convinced that, if you would consider using a certain

transfer formulations in combination with the wind product it was initially calculated for, that you would still get the same difference in the results. I believe this aspect has to be thoroughly discussed.

-----Your previous answer:

A: We used only one wind product, an Altimeter Global Monthly Wind Field on a 1°x1° geographical grid from GlobWave L2P because at the time it was the only one available in the FluxEngine toolset (actually one could choose instead ASCAT Global Monthly Wind Field but... it was not yet ready). However we do not think it is a major problem because the point of the study was to constrain uncertainty caused by the choice of the gas transfer velocity parameterization. We did not want to repeat the analysis done within the same ESA project and presented in two submitted papers (Woolf et al., 2015a and Woolf at al., 2015b) which focused on other sources of uncertainty (the wind field is one of them).

The problem with the two submitted papers is they both got stuck in editorial process because (as we heard) of one recalcitrant reviewer, the same in both cases. We considered removing them from the manuscript. However, we believe the papers will be published sooner or later somewhere, therefore we decided we prefer to retain the citations. An additional reason is that otherwise it would be difficult to support the additional text we'll add now at the request of the editor, without the support of the citations (see below).

The additional text would make it precise what our manuscript is about (effect of the choice of parameterization formula) and what it is not, namely the other sources of uncertainty is measurement error in all the multiple physical variables used for calculation of gas flux (water and atmospheric CO2 partial pressures, wind speed, air and water temperatures temperature skin effects) as well as some usually not used because they are largely unknown (air bubble void fraction, surfactants, sea state parameters etc.).

Because the manuscript topic is only the former, the effect of wind climatologies is outside its focus. We have already stated before, that using FluxEngine we cannot change the wind climatology because there is only one implemented. If we decided to use FluxEngine we had no choice - it's as simple as that. Now, we additionally state that this is outside the scope of the paper. If we wanted to study this effect we would use different tools and create a separate paper. In fact we encourage anyone who thinks it would be publishable (we are not so sure) to write it himself/herself.

The changes:

L58-69 Sources of uncertainty include sampling coverage, the method of data interpolation, fugacity of CO_2 (fCO_2), the method used for normalization of fugacity data to a reference year in a world of ever increasing atmospheric CO_2 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 and 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 choice of wind speed parameterization may cause a difference in the results of k even about 50% (Gregg et al., 20154; Couldrey et al., 2016). In this work we analyze solely the effect of choice between various published empirical wind driven gas transfer parameterizations.

In the Acknowledgements I am missing those to all data sets used. As you have participated in the workshop in Kiel in 2015, you know that acknowledging SOCAT and its data providers is a hot issue

which was intensely debated. Please give them the right acknowledgement they deserve. You could not have done your research without these great data bases to which many scientists contributed. We added the SOCAT data provides into acknowledgments. However our memory of the discussion is that the SOCAT authors expected most of all referencing the SOCAT data papers. We had referenced all of them. The reason for that was exactly to give them back something that is useful in their career evaluations (acknowledgments do not help here).

L383 The authors are very grateful to those who have produced and made freely available the LDEO Flux Climatology base, FluxEngine software funded by European Space Agency, Surface Ocean CO₂ Atlas (SOCAT), GlobWave Project funded by European Space Agency, as well as Centre de Recherche et d'Exploitation Satellitaire (CERSAT) at Ifremere.

L17 delete: net

L17 CO2 (instead of: carbon) L17 fluxes

L17 North Atlantic Ocean

L16 We used a recently developed software toolbox, called the FluxEngine, to estimate the monthly air-sea CO₂ flux**es** for the extratropical **North Atlantic Ocean**, the European Arctic, and globally using several published quadratic and cubic wind speed parameterizations of the gas transfer velocity.

L20 delete: considered (it is certain that the region is a sink; you also show that) L21 delete: often (either it is characterized as region with strong winds or not; there is nothing in between)

L22 data (instead of: measurement)

L20 This region is a large oceanic sink of CO₂, and it is also a region characterized by strong winds, especially in winter but with good in situ data coverage.

L22 that the uncertainty in the parameterization is ... (the reader may have lost what uncertainty) L22 We show that the uncertainty in the parameterization is smaller in the North Atlantic and the Arctic than globally.

L24 parameterizations (instead of: functions)

L25 delete: Whereas

L25-26 ... respectively, when all parameterizations are considered.

L23-26 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 parameterization are considered.

L26 suggest (not: propose)

L26 specify what smaller uncertainty you are talking about

L27 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⁻¹) and lack of any seasonal changes in the direction of the flux direction within most of the region.

L29 Takahashi et al (2009)

L29 for (instead of: within)

L30-31 Change this sentence to: The annual fluxes using the two data sets differ by 8% in the North

Atlantic and 19% in the European Arctic.

L31 fluxes (not: flux)

L32 delete: climatology (this was already criticized by referee #2, but you haven't corrected it) L28 We also compare the impact of using two different *in situ* pCO₂ datasets (Takahashi et al. (2009) and SOCAT) 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 two data sets disagree with each other possibly due to insufficient spatial and temporal data coverage, especially in winter.

L40 ... of atmospheric CO2 ...

L37 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**₂ (Takahashi et al., 2002; Takahashi et al., 2009; Landschützer et al., 2014; Le Quéré et al., 2015).

L51 circulation (-s)

L51 ... or changes in meridional overturning circulations...

L57 "in -water fugacity data quality" This is an awkward term. Please use a more current one, or rewrite/describe.

L58 ... fugacity of CO₂ (fCO₂)...

L62 delete: Although (this sentence does not make sense when beginning with Although)

L69 The North Atlantic is one of the regions of the world ocean best covered by CO_2 fugacity measurements (Watson et al., 2011), the Arctic seas coverage is much poorer, especially in winter (Schuster et al., 2013).

L69 and (instead of: but)

L71 delete: implies or rephrase the sentence

L75-78 ... 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).

L75 ... the parameterizations were ...

L76 wind fields (instead of: wind distributions)?

L78 by (not: through)

L81 These regions were chosen as they are the areas for which many of the parameterization **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

L85 was (not: were)

L92 delete: These data are freely available for the scientific community to use. (is superfluous) L91 which was created within European Space Agency funded OceanFlux Greenhouse Gases project (<u>http://www.oceanflux-ghg.org</u>).

L93-94 producing (instead of: allowing the generation of the)

L99 ... producing monthly global gridded net air-sea fluxes...

L95 "Some Monthly composite" This doesn't make sense. Shouldn't this be: One monthly composite? (no capital for monthly)

L100 One monthly composite file includes the mean (first order moment), median, standard deviation and the second, third and fourth order moments.

L98 ...portal; an example of monthly EO input data includes: rain ... (no parentheses here and at end of sentence)

L103 Users can choose from all of the data available on the web portal; an example of 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_2 , SST, salinity.

L106 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.

L106 version (full)

L107 ... containing 6.3 and 10.1 million surface water CO2 fugacity values for versions 1.5 and 2.0, respectively, with a global coverage ...

L111 with (not: within)

L110-116 For some calculations we used, as an alternative, Surface Ocean CO₂ Atlas (SOCAT) **version** 1.5 and 2.0 (Sabine et al., 2013; Pfeil et al., 2013; Bakker et al., 2014) *p*CO₂ and associated SST data. SOCAT is a community driven dataset **containing 6.3 and 10.1 million surface water CO₂ 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.

L111 what is SSTfnd? This has not been defined earlier in the paper.

We changed SSTfnd to SStskin (this was used for both datasets) and explained what SSTskin means: **L116** 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 μ m)...

L111-112 Please briefly explain what kind of data Operational Sea Surface Temperature and Sea Ice Analysis (OSTIA) is?

We deleted sentence about OSTIA because, as we checked, we didn't use SSTfnd, only SSTskin, from ESA/ARC/(A)ATSR Global Monthly Sea Surface dataset, for both datasets.

L116-121 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 μ m) values were taken from 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).

L111-114 This sentence does not make sense. Does this mean that the SST data used in combination with the Takahashi data are from OSTIA and those with SOCAT from ARC/(A)ATSR? And if the answer is yes, why is that? Why not take the same SST data for both data sets? And the reader needs more info as to what kind of data sets these are.

For the second part I suggest:

... in the case of the SOCAT database, SST data were taken from the ARC/(A)ATSR Global Monthly Sea Surface dataset (Merchant et al., 2012).

We already explained and pointed out that which product were used

L117 Please give a reference or website for where the data can be found L117 It is not clear to me what σ 0 stands for. Is it for the sea roughness? Please clarify

L123-128 We used Earth Observation (EO) wind speed and sea roughness (σ_0 – **altimeter backscatter signal at the** K_u- band data from GlobWave L2P prodcuts) obtained from the European Space Agency (ESA). 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 and are publicity available at the Ifremere/CERSAT cloud (http://globwave.ifremer.fr/products/data-access).

Sigma0 (σ_0) is the altimeter backscatter signal (see Fangohr and Woolf 2007))

L120-124 This reads as if it is an advertisement. "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 windwave and swell data ..." There is certainly some useful info there, but this part needs to be rephrased.

L129-134 The aim of the project was to constrain a uniform, harmonized, quality controlled, multisensor set of satellite wind-wave data for using by different communities despite of *in situ* data. Wave data are collected from six altimeter missions (Topex/POSEIDON, Jason-1/22, CryoSAT, GEOSAT and GEOSAT Follow On) and from ESA Synthetic Aperture Radar (SAR) missions, namely ERS-1/22 and ENVISAT. All data come in netCDF-3 format.

L121 If using CNES, it must be defined

L128 GlobWave Project is funded by ESA and subsidised by Centre National d'Etudes Spatial (CNES).

L134 If you decide to write is so extensive, you may also mention that it is Pg is 1015 g. L141 We give all results of net CO_2 fluxes in the SI unit of Pg (Pg is 10^{15} g which is numerically identical to Gt).

L141 delete: also

L142 in (not: within)

L148 ... and the difference in CO_2 concentration (gm⁻³) in the sea water and its interface with the air (Land et al., 2013)

L149 with (not: to) L156 ... fugacity with the partial pressure...

L155 Equation (2) will then become: L162 Equation (2) will then became:

L161 In this study we ... L168 In this study we chose to...

L179 change to something like: ... where Scskin stands for the Schmidt number at the skin surface,

... L181

delete:

temperature

L186 where Sc_{skin} stands for the Schmidt numbers at the skin surface, a function of SST ([= (kinematic viscosity of water)/(diffusion coefficient of CO₂ 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 equations 4-8 the exponent of U10 looks very strange. It should be situated over U

We used U10 as it was specify inside the articles about choosing parameterizations. It is important to use U10 not U, because this means wind speed at 10 m heigh.

L186-187 delete: and it is provided as an option in the FluxEngine toolbox. (superfluous) L192 ... parameterization, which was developed as a test algorithm within of OceanFlux GHG Evolution project.

L190-191 delete: (one of the aims of the ongoing OceanFlux Evolution project is to develop a calibration for this algorithm) (this is irrelevant for the present study and only distracts)

L194 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.

L197 I suggest to order like this: global gridded monthly net CO2 air-sea fluxes

L202 Using the FluxEngine software, we have produced global gridded monthly net CO₂ air-sea fluxes and from these we have extracted the values for the two study regions, the extratropical North Atlantic Ocean...

L200 order: air-sea CO2 fluxes L205 ...mean air-sea CO₂ fluxes....

L202-204 "The area, as a whole, is a 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." These two sentences together don't make sense. The seasonal maps cannot show whether a region is a net source, this can only be demonstrated by an annual map. Moreover, this map does not demonstrate the area to be a net sink

If you mention features like the North Atlantic Drift and the East Greenland Current, you must show in the Figure where they are located. Not all readers know this exactly.

L207 The area, as a whole, is a sink of CO_2 (from blue to purple colored in the Fig. 1) but in some parts, close to North Atlantic Drift and East Greenland Current (Fig. 9), is net source (from yellow to red colored in the Fig. 1). At the seasonal maps one can see more variability affects by physical process or biological activity.

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.

L248 Southern Ocean

L249-250 I suggest: In the global case, the values for the backscatter and the wind-driven version of OCeanFlux were 44% and 52 % higher, respectively.

L253 The OceanFlux GHG parameterization results, for the backscatter and wind-driven version, in net air-sea CO_2 fluxes that are 38% and 47% higher for North Atlantic than the N2000 result and in the global case the values were 44% and 52% higher, respectively.

L256 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.

Towards the end of the paragraph (about L235-250), the number of values and where they belong to, is confusing. Please rewrite so that always is clear which value is higher or lower and where it belongs to. Be precise in what you write.

L221-227 This effect is especially visible between formulas with different powers of U_{10} . Figure 4 shows the difference in the air-sea CO₂ 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.

L229-258 Figure 5 shows the monthly values of air-sea CO₂ fluxes for the five parameterizations (eq. 4-8) for the North Atlantic and the European Arctic. The regions are sinks of CO₂ 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 6 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. The results show that the annual North Atlantic net air-sea CO₂ 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 CO₂ sink the values are, respectively, -1.30 Pg C and -2.15 Pg C. Table 1 as well as Figure 6 shows the same data "normalized" to the N2000 data (divided by value), this allows us to visualize the relative differences. In the case of the North Atlantic using the "quadratic" W2014 and H2006 parameterizations results in a net air-sea flux that is, respectively, 4% and 5% higher in absolute value than the equivalent N2000 result, while the "cubic" WMcG1999 and McG200 results in values that are up to 28% and 44% than N2000 results. The respective values for the Arctic are 3% for W2014 and 4% for H2006 as well as 28% for WMcG1999 and 44% for McG2001. In the case of global net air-sea CO_2 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, in net air-sea CO₂ fluxes that are 38% and 47% higher for North Atlantic than the N2000 result and in the global case the values were 44% and 52% higher,

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

L252 I suggest: All the above results were obtained with the Takahashi et al. (2009) pCO2 climatology.

L260 All the above results were obtained with the Takahashi et al. (2009) pCO_2 climatology and for comparison...

L269 I think you mean "range" instead of "variation".

L278 ... as this range is within the experimental.....

L269 "within the experimental uncertainty" What is this uncertainty? This is important for assessing the magnitude of the range (see above).

We had already a long discussion about that in previous iterations of the editorial process. This is exactly why we wanted the Nightingale "gray literature" citation because it refers to this: "For gas transfer of CO2 over the oceans the relationships proposed in Nightingale et al. (2000), Sweeney et al. (2007), Ho et al. (2006), and Wanninkhof et al. (2009) are recommended. They are very similar and fall within the overall uncertainty of DT measurements." DT means dual tracer. Nightingale (and also Ho and Wanninkhof who co-chaired the session) state here that the databases their parameterizations are based on have experimental uncertainty large enough to make it impossible to decide between their formulas. The sources of the uncertainty are described in the report. We believe that our sentence is good representation of that discussion and its conclusions and we refer the reader to the document itself. We did not include "dual tracer" in the sentence because the other experimental method (eddy covariance) described in the report has even larger errors (uncertainties) so logically mentioning only DT (as in the sentence cited above could be misleading for a reader.

L273-275 "The differences between these and the quadratic parameterization are large and although the quadratic functions are supported by several lines of evidence (see Garbe et. al., 2014 for discussion." I don't understand this when placed against the previous sentence. Moreover, the sentence is an anacoluthon. There might be an error here: instead of quadratic you mean cubic here, right? The word "although" appears wrongly used.

L282-287 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 lead to net air-sea CO_2 fluxes that are considerably larger in absolute values, by up to 33% in the North Atlantic and more than 50% globally.

L282 changes (+s)

L283 Change to: ... example in the South Atlantic, the annual mean wind speed is 8.5 m s-1 (I cannot imagine that the mean wind speed can be more precise than given by one decimal) L284 ... and the CO2 sink (south of 45°) decreases significantly after 1990 ... L285-286 Change this to: ...speeds; this may cause higher CO2 concentrations (and higher pCO2) in

surface water due to enhanced vertical mixing of CO2-rich deep waters (Le Quéré et al., 2007). (I don't know what biological has to do with this; either explain better or skip) Please note that the

CO2 has increased to normal levels again in the 2000s (Landschützer et al 2015, Science). Any comments?

L291-295 ...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⁻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 pCO_2) 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).

We did not want to get too deep into the effect of NAO on North Atlantic CO2 fluxes (an interesting topic one of us [JP] is deeply interested in but far from the scope of their paper about the effect of parameterization choice on net flux volumes). A change about 2000 is probably NAO related because NAO values peaked around 1995.

The biological factors are of curse related to seasonal primary production changes. In oligotrophic regions pCO2 increases in summer due to water temperature changes. In regions of high phytoplankton concentration, pCO2 decreases in summer due to increased primary production. It is a well know effect (see for example Figure 7 in Takahashi et al. 2003).

L295 we added "(seasonal changes in primary production)".

L287 which flux difference is meant here? Do you just mean the air-sea CO2 flux here? L287 Change to: ... in the Southern Ocean is strongly dependent on the choice ... L287-288 gas transfer parameterizations, I guess?

L288-289 "This is more surprising, for North Atlantic, given that at least some of the older parameterizations were developed using a smaller range of winds than can exist in the North Atlantic" I do not understand this sentence. Please rephrase. Which are the older

parameterizations? Please mention them, the reader should not have to guess.

L290-291 "After analysis of this unexpected fact, using the formula multiplied by the different wind distribution, we have found two reasons for this." This is not a good sentence. Actually, I think it can be deleted. Just write that there may be two reasons for this fact.

L296-300 Takahashi et al. (2009) also indicate that the air-sea CO₂ fluxes difference in the Southern Ocean is strongly **dependent on** the choice of the gas transfer parameterizations and wind speed. Smaller difference in the North Atlantic, than globally, 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 can exist in the North Atlantic. There may be two reasons for this.

L292-293 Change to (as the second was not clear enough)... the cubic parameterization implies higher air-sea fluxes for high winds, whereas the quadratic ones lead to higher fluxes for weaker winds.

L301 the cubic parameterization implies higher air-sea fluxes for high winds, whereas the quadratic ones lead to higher fluxes for weaker winds.

L310 being larger than that in the North Atlantic.

L310 The Southern Ocean

L311 than the North Atlantic (including also the Arctic Seas) which seems to

L320-322 ...**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.

L312 "The other reason" Please repeat here the other reason for what, because the reader may

have lost it due to the long previous explanation of reason one.

L322 The other reason of smaller relative differences between the parameterizations in the North Atlantic than globally,...

L319 A. Watson, University of Exeter

L330 ... observations (A. Watson, University of Exeter- personal communication)...

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]

L326-327 "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." Why is this your conclusion? There is no reason to be in favour of the one or the other relationship. In the previous discussion about the quadratic and cubic relationships you have not shown or evidenced that one or the other is the "correct" one. So it may also be that Fangohr and Woolf do not overestimate and show that the cubic relationship is not so bad after all. We deleted this.

L330 SOCAT is not a climatology so should not be called that way. If the FluxEngine software generates a climatology from SOCAT, this should be mentioned again. L330-340 It does not become clear here whether the changes in the Arctic are or are not caused by processing of the data through the FluxEngine software. It should be clearly stated that this may be the case.

L338-340 Please note that Yasunaka et al is not independent of SOCAT as it contains a significant part of the SOCAT data. Actually, also the Takahashi and SOCAT data base are not independent. Most data from Takahashi are also in SOCAT. This info should be included in the manuscript somewhere.

L342-356 Using both Takahashi et al. (2009) climatology and SOCAT datasets (Fig. 7) results in similar annual net air-sea CO_2 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 datasets contains more recent data). One have to remember that at present most of data from Takahashi et al. (2009) are included in SOCAT, so differences in the European Arctic, may be due to the underlying sparse data coverage and possible interpolation artifacts (Goddijn-Murphy et al., 2015) as well as processing of the data through the FluxEngine. 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. This discrepancy makes us treat the net air-sea CO_2 fluxes 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, new data, not included in the SOCAT versions we used, have been available to the recent analysis by Yasunaka et al. (2016). The observed inwater pCO_2 data (Fig. 3 in Yasunaka et al., 2016), especially since 2005, show clearly an annual cycle compatible with the SOCAT seasonal flux variability.

L352-353 Change to: We explain the smaller North Atlantic variability to be a combination of, firstly, higher than global average ...

L368 We explain the smaller North Atlantic variability to be a combination of, firsty, higher than global average...

L355 repeated L372 ...repeated...

L372 What is "correcting the native-speaker verification of our English"? This does not seem to be correct English. Please just write what your colleague did: correct the manuscript for English language.

L383-392 The publication has been financed from the funds of the Leading National Research Centre (KNOW) received by the Centre for Polar Studies for the period 2014-2018; OceanFlux Greenhouse Gases Evolution, a project funded by the European Space Agency, ESRIN Contract No. 4000112091/14/I-LG; and GAME "Growing of Marine Arctic Ecosystem", funded by Narodowe Centrum Nauki grant DEC-2012/04/A/NZ8/00661. We would also like to thank Jamie Shutler for important advice on the FluxEngine and for correct the manuscript for English language. The authors are very grateful to those who have produced and made freely available the LDEO Flux Climatology base, FluxEngine software funded by European Space Agency, Surface Ocean CO₂ Atlas (SOCAT), GlobWave Project funded by European Space Agency, as well as Centre de Recherche et d'Exploitation Satellitaire (CERSAT) at Ifremere.

There are many errors in the references. Below I listed some, but there are more. Please go through the references and correct all errors. Please also comply with the format of Ocean Science: http://www.ocean-science.net/Copernicus_Publications_Reference_Types.pdf Words in a title of a reference should not be written in capitals. L443-446 Rödenbeck, Buitenhuis, Metzl - please check this reference, as there are several errors in it. L466-468 please check this reference, as there are several errors in it. L475 report L483-486 Use initials L529, 534 use doi format, not http L537 Nojiri ; Deep-Sea (hyphen) All have been done from the list above.

Table 1 fluxes (not: fluxed) Figures 1,2,3,4 seasons not with capitals Note that there is a difference between the captions in the Figure list and those below the Figures at the end of the manuscript. Figure 1 caption: delete: combine ; delete (L597): in L597 Takahashi et al (2009) Figure 2 L602 Takahashi et al (2009) L602 delete: in L607 delete: in Figure 4 L612 between a cubed and a squared parameterization (... L613 delete: in Figure 5

L616 monthly values of CO2 air-sea fluxes L617 the North Atlantic ; the European Arctic L622 parameterizations (+s)

L625 Comparison of monthly air-sea CO2 fluxes calculated with different ...

L623-656 Figure 1. Seasonal and annual mean air-sea fluxes of CO_2 (mg C m⁻² day⁻¹) 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.

Figure 2. Seasonal and annual pCO_2 values (µatm) in surface waters of the North Atlantic, estimated using 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 3. Wind speed distribution U_{10} (ms⁻¹) in the North Atlantic used to determine the relationship between gas transfer velocity and air-sea CO₂ 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 masks.

Figure 4. Differences maps for the air-sea CO_2 fluxes (mg C m⁻² day⁻¹) in the North Atlantic, between a cubed and a squared parameterization (Wanninkhof and McGillis 1999 and Wanninkhof 2014) 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 5. Monthly values of CO_2 air-sea fluxes (Pg month⁻¹) for the five parameterizations (eq. 4-8) a) the North Atlantic, b) the European Arctic.

Figure 6. Annual air-sea fluxes of CO_2 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 European Arctic, d) the Southern Ocean. Average values for all parameterization and standard deviations are marked as vertical gray lines.

Figure 7. Comparison of monthly 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) a) the North Atlantic, b) the European Arctic.

Figure 8. Different k660 parameterizations as a function of wind speed.

L340 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.

Effect of gas-transfer velocity parameterization choice on <u>air-sea</u> CO₂ air-sea fluxes in the North Atlantic and the European Arctic 3

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7 8

11 Abstract12

13 The oceanic sink of carbon dioxide (CO_2) 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 14 15 research. One of the sources of the uncertainty within this calculation is the parameterization chosen for the CO_2 gas transfer velocity. We used a recently developed software toolbox, called the 16 17 FluxEngine, to estimate the monthly-net <u>air-sea CO₂ carbon air-sea</u> fluxes for the extratropical 18 North Atlantic Ocean, the European Arctic, and globally using several published quadratic and cubic wind speed parameterizations of the gas transfer velocity. The aim of the study is to constrain 19 the uncertainty caused by the choice of parameterization in the North Atlantic. This region is 20 21 considered a large oceanic sink of CO₂, and it is also a region -often characterised by strong winds, especially in winter but with good in situ data measurement coverage. We show that this the 22 23 uncertainty in the parameterization is smaller in the North Atlantic and the Arctic than globally. It is 24 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 functions with quadratic wind dependence. 25 Whereas tThis uncertainty becomes 46%, 44% and 65% respectively, if when you consider all 26 27 parameterization are considered of the parameterizations studied. We suggestpropose that this smaller uncertainty (5% and 4%) is caused by a combination of higher than global average wind 28 speeds in the North Atlantic (> 7 ms⁻¹) and lack of any seasonal changes in the direction of the flux 29 direction within most of the region. We also compare the impact of using two different in situ partial 30 31 pressure of CO₂ (pCO₂) datasets (Takahashi et al. (2009) and SOCAT) forwithin the flux 32 calculation. The annual fluxes using the two data sets differ by Differences in these pCO2 data in turn cause differences in the annual net flux values of 8% in 8% in 18% in 19% in 33 34 the European Arctic. The seasonal fluxes in the Arctic computed from two-climatology datasets disagree a sets are opposite to one another, with each other- possibly due to insufficient spatial and 35 36 temporal data coverage, especially in winter.

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39 1. Introduction

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The region of extratropical North Atlantic <u>Ocean</u>, 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 <u>atmospheric CO₂</u>
(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).

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49 The trend and variations in the North Atlantic CO₂ sinks has been intensively studied since 50 observations have shown it appears to be decreasing (Lefèvre et al., 2004). This decrease on inter-51 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 52 53 of these changes are the result of long-term changes, decadal changes in atmospheric forcing,namely the North Atlantic Oscillation (Gonzalez-Davila et al., 2007; Thomas et al., 2008; Gruber 54 55 2009; Watson et al., 2009) or changes in meridional overturning circulations (Pèerez et al., 2013). 56 Recent assessments of the Atlantic and the Arctic net sea-air CO₂ fluxes (Schuster et al., 2013) and 57 the global ocean net carbon uptake (Wanninkhof et al., 2013) show that the cause is still unknown.

58

59 To study the rate of the ocean CO₂ sink-and especially its long-term trend, one needs to first 60 constrain the total-uncertainty in the flux calculation. The global interannual air-sea CO₂ fluxes variability can be vary about 60% due to different in pCO_2 and 35% by k (Courtney et al., 2016). 61 Sources of uncertainty include sampling coverage, the method of data interpolation, fugacity of CO₂ 62 63 (fCO₂)in-water fugacity data quality, the method used for normalization of fugacity data to a 64 reference year in a world of ever increasing atmospheric CO₂ the measurement uncertainty in all the parameters used to calculate the fluxes (including partial pressure in water and air, bulk and skin 65 66 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 67 68 fraction, surfactant effects etc.) as well as the choice of gas transfer velocity k parameterization 69 formula 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 70 71 product provides an additional source of uncertainty in gas transfer velocity, even by 10% - 40% 72 and choice of wind speed parameterization may cause a difference in the results of k, even about 73 50% (Gregg et al., 20154; Couldrey et al., 2016). In this work we have analyze solely the effect of 74 choice between - chosen to analyze various published empirical wind driven gas transfer 75 parameterizations. TAlthough the North Atlantic is one of the regions of the world ocean best 76 covered by CO_2 fugacity measurements (Watson et al., 2011), the Arctic seas coverage is much 77 poorer, especially in winter (Schuster et al., 2013). 78

79 One of the factors influencing the value of the calculated net air-sea gas flux is the choice of the 80 formula for the gas transfer velocity. Within the literature there are many different parameterizations 81 to choose from and, but most depend on a cubic or quadratic wind speed relationship. The choice of 82 the appropriate parameterization is not trivial as indicated by the name of an international meeting whichthat focussed on this e-topic implies ("k conundrum" workshop, COST-735 Action organized 83 meeting in Norwich, February 2008). The conclusions from this meeting have been incorporated 84 into a recent review book chapter (Garbe et al., 2014). This paper concentrates on quantifying the 85 uncertainty caused by the choice of the gas transfer velocity parameterization in the North Atlantic 86 87 and the European Arctic. These regions were chosen as they are the areas for which many of the 88 parameterization wereas originally derived. They are also regions with wind fields distributions skewed towards higher winds (in comparison to the global average) enabling the effect of stronger 89 winds on the net flux calculations to be investigated bythrough using published gas transfer velocity 90 91 formulas.

- 92
- 93 2. Methods94
- 95 2.1 Datasets

97 We calculated net air-sea CO₂ fluxes using a set of software processing tools called the 98 'FluxEngine' (Shutler et al., 2016), which wasere created within European Space Agency funded 99 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 100 101 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 102 103 alternatively there is a version that can be run through a web interface. Within the online web 104 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 105 106 you to select several different air-sea flux parameterizations, as well as input data, producing 107 allowing the generation of the monthly global gridded net air-sea fluxes products with 1° x 1° spatial resolution. The output consists of twelve NNetCDF files (one file per month). Some One monthly 108 109 Monthly composite file includes the mean (first order moment), median, standard deviation and the 110 second, third and fourth order moments. There is also information (meta data) about origin of data 111 inputs. Users can choose from all of the data available on the web portal; -(an example of monthly EO input data includes: rain intensity, wind speed and direction, % of sea ice cover from monthly 112 model data, ECMWF air pressure, whitecapping (Goddijn-Murphy et al., 2011), two options for 113 114 monthly <u>elimatology datasets</u> of pCO_2 , SST, salinity). The user then needs to choose the different 115 components and structure of the net air-sea gas flux calculation and choose the transfer velocity 116 parameterization. 117 For the calculations, we used pCO_2 and salinity values from Takahashi et al. (2009) climatology 118

which is based on more than 3 million measurements of surface water pCO_2 in open-ocean 119 environments during non El Nino conditions. For some calculations we used, as an alternative, 120 Surface Ocean CO₂ Atlas (SOCAT) ver-sion 1.5 and 2.0 (Sabine et al., 2013; Pfeil et al., 2013; Bakker et al., 2014) pCO₂ and associated SST data. SOCAT is a community driven dataset 121 containing respectively 6.3 and 10.1 million surface water CO₂ fugacity values for version 1.5 and 122 123 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 124 the climatologies were calculated for year 2010 within the FluxEngine toolset. The SSTskin 125 (defined within Group for High Resolution SST (GHRSST) as temperature of the surface measured 126 by an infrared radiometer operating at the depth of ~10-20 μ m) fnd-values were taken -from the 127 Advance Along Track Scanning Radiometer (ESA/ARC/(A)ATSR) Global Monthly Sea Surface 128 dataset (Merchant et al., 2012)from Operational Sea Surface Temperature and Sea Ice Analysis 129 130 (OSTIA) (Donlon et al., 2011), and in the case of both SOCAT databasesets, -and while SST skin 131 data that we use come from ARC/(A)ATSR Global Monthly Sea Surface dataset (Merchant et al., 132 2012). Both data sets have been preprocessed in the same way for use with the FluxEngine (Shutler 133 et al., 2016).

134

135 We used Earth Observation -(EO) wind speed and sea roughness (σ_0 –altimeter backscatter signal in 136 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 137 138 available AAltimeters (spanning multiple space agencies) and from ESA Synthetic Aperture Radar 139 (SAR) data and are publicity available at the Ifremere/CERSAT cloud (http://globwave.ifremer.fr/products/data-access).- GlobWave Project is <u>an initiative</u> funded by 140 ESA and subsidised by Centre National d'Etudes Spatial -(CNES). The aim of the project wasis to 141 142 constrain a uniform, harmonized, quality controlled, multi-sensor set of satellite wind-wave data for 143 using by different communities despite of in situ data.to improve the uptake of satellite derived 144 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 145 146 wave data. Wave data areis collected from six-both altimeters missions (ERS-1, ERS-2, ENVISAT,

- 147 Topex/POSEIDON, Jason-1/2, Jason-2, CryoSAT, GEOSAT and GEOSAT Follow On) and from
- 148 ESA Synthetic Aperture Radar (SAR) missions, namely ERS-1/2, ERS-2 and ENVISAT. All data
- 149 come in netCDF-3 format.
- 150

All analyses were performed using global data within the FluxEngine software. From the gridded product (1° x 1°) we extracted the extratropical North Atlantic (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 CO₂ (upward ocean-toatmosphere gas fluxes) are positive and sinks (downward atmosphere-to-ocean gas fluxes) are negative. We give all results of net CO₂ fluxes in the SI unit of Pg (<u>Pg is 10¹⁵ g</u> which is numerically identical to Gt).

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160

159 2.2. *k* parameterizations

161 The flux of CO₂ at the interface of air and the sea is controlled by wind speed, sea state, sea 162 surface temperature (SST) and other factors. We estimate the net air-sea flux of CO₂ (*F*, mg C m⁻² 163 day⁻¹) as the product of gas transfer velocity (*k*, ms⁻¹) and also the difference in CO₂ concentration 164 (gm⁻³) <u>inwithin</u> the sea water and its interface with the air (Land et al., 2013). The concentration of 165 CO₂ in sea water is the product of its solubility (α , gm⁻³ µatm⁻¹) and its fugacity (*fCO*₂, µatm). 166 Solubility is in turn, a function of salinity and temperature. Hence F is defined as:

$$\mathbf{F} = k \left(\alpha_{\rm W} f \mathbf{CO}_{\rm 2W} - \alpha_{\rm S} f \mathbf{CO}_{\rm 2A} \right) \tag{1}$$

where the subscripts denote values in water (W) and the air-sea interface (S) and in the air (A). We can exchange fugacity <u>withto</u> the partial pressure (their values differ by <0.5 % over the temperature range considered) (McGillis et al., 2001). So equation (1) now becomes:

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191 192 193 $\mathbf{F} = k \left(\alpha_{\rm W} p \mathbf{CO}_{2\rm W} - \alpha_{\rm S} p \mathbf{CO}_{2\rm A} \right) \tag{2}$

176 One can also ignore the differences between the two solubilites, and just use the waterside solubility 177 $| \alpha_W$. Equation (2) will-<u>be represented</u> then <u>became</u>as: 178

$$F = k \alpha_{\rm W} \left(p \rm CO_{2W} - p \rm CO_{2A} \right) \tag{3}$$

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

183 In this studywork we chose to analyze the air-sea gas fluxes using five different gas transfer 184 parameterizations (k). All of them are wind speed parameterizations, but differ in the formula used:

$$k = \sqrt{(660.0) \text{ Sc}_{skin}} * (0.212 \text{ U}_{10}^2 + 0.318 \text{ U}_{10})$$
(Nightingale et al., 2000), (4)

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

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

(Wanninkhof and McGillis, 1999),

194
195
$$k = \sqrt{(660.0/ \text{ Sc}_{skin}) * 0.251 \text{ U}_{10}^2}$$
 (7)
196 (Wanninkhof, 2014),
197

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

201 where $\underline{Sc_{skin}}$ stands for the the subscripts are Schmidt numbers at the skin surface ($\underline{Sc_{skin}}$), a function 202 of SST ([= (kinematic viscosity of water)/(diffusion coefficient of CO₂ in water)]), 660.0 is the 203 Schmidt number for carbon dioxide corresponding to values of carbon dioxide at at 20 °C 204 temperature in seawater, U_{10} is the wind speed 10 m above the sea surface.

205

206 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 207 algorithm within of OceanFlux GHG Evolution project. and it is provided as an option in the 208 209 FluxEngine toolbox. This parameterization separates contributions from direct- and bubble-210 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 211 212 calibrated. (one of the aims of the ongoing OceanFlux Evolution project is to develop a calibration 213 for this algorithm). We used two versions of this parameterization: wind driven direct transfer 214 (using the U10 wind fields) and radar backscatter driven direct transfer (using mean wave square 215 slope) as described in Goddijn-Murphy et al. (2012).

- 216217 3. Results
- 217 218

219 Using the FluxEngine software, we have produced $\frac{-\text{net CO}_2}{2}$ -global gridded monthly griddednet CO₂ 220 air-sea fluxes and from these we have extracted the values for the two study regions, the 221 extratropical North Atlantic Ocean and separately for its subset -, the European Arctic seas. Figure 1 222 shows maps of the monthly mean $-CO_2$ -air-sea CO_2 fluxes for the North Atlantic, calculated with Nightingale et al. (2000) (hereafter called N2000) k parameterization and the Takahashi et al. (2009) 223 224 climatology for the whole year and for each season. The area, as a whole, is a sink of CO₂ (from 225 blue to purple colored in the Fig. 1) but even the seasonal maps show that in some parts, regions 226 close to North Atlantic Drift and East Greenland Current (Fig. 9), isare net sources source (from 227 yellow to red colored in the Fig. 1). - At T the seasonal maps one can show-see even more variability 228 affects by physical process or biological activity. -For example, the areas close to the North Atlantic 229 Drift And East Greenland current are sinks of CO₂ in the summer (likely due to the growth of phytoplankton) while the southern most areas of the region become CO₂ sources in summer and 230 231 autumn (which is likely to be due to the effect of sea-water temperature changes). Much of this 232 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). 233 234 However, the flux is proportional to the product of $\Delta p CO_2$ and k. In most parameterizations k is a 235 function of wind speed (eqs. 4-8). The mean 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 236 world ocean (7 m s⁻¹; Courtney et al., 2016), with mean values higher than 10 m s⁻¹ in many regions 237 of the study area in all seasons except for the summer (with highest values in winter). This is 238 239 important because the air-sea flux depends not only on average wind speed but also on its 240 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 air-sea <u>CO₂</u> fluxes calculated using 241 two example parameterizations: -one proportional to U_{10}^{3} (eq. 6) and one to U_{10}^{2} (eq. 7), namely 242 Wanninkhof and McGillis (1999) and Wanninkhof (2014). It can be seen that the "cubic" function 243 results in higher absolute air-sea flux values when compared to the "quadratic" function in the 244 245 regions of high winds, and lower absolute air-sea flux values in weaker winds.

246

Figure 5 shows the monthly values of CO_2 -air-sea CO_2 fluxes for the five parameterizations (eq. 4-8) for the North Atlantic and the European Arctic. The regions are sinks of CO_2 in every month,

249 although August is close to neutral for the North Atlantic. The results using cubic parameterizations 250 (eqs. 6 and 8) are higher in absolute values, respectively by up to 30% for Wanninkhof and 251 McGillis (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. 252 253 (2006) (hereafter called H2006) (eqs. 5 and 7) resulted in fluxes within 5% of N2000. Annual net 254 fluxes for the North Atlantic and the European Arctic and global (included for comparison) are 255 shown in Table 1. In addition to the five parameterizations Figure -6 presents results for both of the 256 OceanFlux GHG Evolution formulas (using wind and radar backscatter data). The mean and 257 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., 258 259 2014) but they cannot be directly compared due to different parameterization choices and 260 methodologies. The results show that the annual North Atlantic net air-sea CO₂ sink, 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 261 case of global net air-sea CO₂ sink the values are, respectively, -1.30 Pg C and -2.15 Pg C. Table 1 262 as well as Figure 6 shows the same data "normalized" to the N2000 data (divided by value), this 263 264 allows us to visualize the relative differences. In the case of the North Atlantic using the "quadratic" Wanninkhof (2014) and Ho et al. (2006) parameterizations results in a net air-sea flux that is, 265 respectively, 4% and 5% higher in absolute value than the equivalent N2000 result, while the 266 267 "cubic" WMcGanninkhof and McGillis (1999) and McGillis et al. (2001) results in values that are up to 28% and 44% than N2000 results.- The respective values for the Arctic are 3% for W2014 268 and, 4% for H2006quadratic as well as 28% for WMcG1999 and 44% for eubic functionsMcG2001. 269 270 In the case of global net air-sea CO_2 fluxes the equivalent values are 8% (W2014) and 9% (H2006) 271 higher than the N2000 result for the quadratic functions as well as 33% (WMcG1999) and 65% 272 (McG2001) for cubic ones. The OceanFlux GHG parameterization results, for the backscatter and 273 wind driven versions, -in net air-sea CO₂ fluxes that are 38% and 47% higher for North Atlantic than the N2000 result (for the backscatter and wind driven versions respectively and in the global 274 case the values were 44% and 52% higher, respectively.). The spread of the Arctic values was lower 275 276 than the Atlantic ones (see Table 1). On the other hand, the values for the Southern Ocean were 277 slightly higher than for the North Atlantic but lower than the global ones, with the exception of the 278 OceanFlux GHG parameterizations. In the case of global values the values were, 44% and 52% 279 respectively.

281 All the above results were obtained with used the Takahashi et al. (2009) pCO_2 climatology and -282 F for comparison, we have also calculated the air-sea \underline{CO}_2 fluxes using the re-analysed SOCAT 283 version 1.5 and 2.0 data (Goddijn-Murphy et al., 2015). Figure 7 shows the results using the N2000 284 k parameterization for all three of the datasetselimatologies (Takahashi et al. (2009) and both SOCAT). In the case of the North Atlantic Ocean study area, although the monthly values show 285 286 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 287 Takahashi et al. (2009) and SOCAT v-1.5 and -0.41 Pg C for SOCAT v-2.0. In the case of the 288 European Arctic the situation is very different, with Takahashi et al. (2009) and SOCAT dataset 289 290 derived climatologies resulting in inverse seasonal variability but with annual net air-sea CO₂ fluxes 291 results that are similar: -0.102 Pg C for Takahashi et al. (2009), -0.085 Pg C for SOCAT v_-1.5 and -292 0.088 Pg C for SOCAT v-2.0.

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294 4. Discussion295

Our results show that using the three "quadratic" parameterizations (Nightingale et al., 2000; Ho et al., 2006 and Wanninkhof, -2014) results in air-sea flux 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 case (Fig. 6). This result <u>above</u> confirms that at present, these different parameterizations are 300 interchangeable for the North Atlantic as this rangevariation is within the experimental uncertainty 301 (Nightingale, 2015). The three parameterizations were derived using different methods and data 302 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), 303 and global ocean ¹⁴C inventories in the case of Wanninkhof (2014). The differences between these 304 305 and the quadratic and cubic parameterization are large, e and instead of although the quadratic functions that are supported by several lines of evidence (see Garbe et. al., 2014 for discussion), the 306 307 cubic function are not completely refuted by the available observation. Therefore, it is important to 308 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 more than 50% 309 310 globally.

312 The above results imply smaller relative differences between the parameterizations in the North 313 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 314 example in the South Atlantic, the annual mean of-wind speed is within 8.548 m s⁻¹ (Takahashi et 315 al., 2009) and <u>sink of of the -CO₂ sink</u> (south of 45°) decreases significantly after 1990 with 316 increasing wind speeds; what can influencethis may cause higher CO2 concentration of (and higher 317 318 pCO_2) in surface water due to enhanced vertical mixing of CO_2 -rich deep waters (Le Quèrè et al., 319 2007).and biological activity (seasonal changes in primary production). (Le Quèrè et al., 2007). 320 Takahashi et al. (2009) also indicate that the air-sea CO₂ fluxes difference in the Southern Ocean 321 isare very strongly dependentee onto the choice of the gas transfer parameterizations and wind 322 speed. Smaller difference in the North Atlantic, than globally, are more surprising, given that at least 323 some of the older parameterizations (e.g. W2009 or WMcG1999) were developed using a smaller 324 range of winds than can exist in the North Atlantic. This is more surprising, for North Atlantic, 325 given that at least some of the older parameterizations were developed using a smaller range of winds than can exist in the North Atlantic. After analysis of this unexpected fact, using the formula 326 327 multiplied by the different wind distribution, we have There may be two reasons found two reasons for this. First, when comparing quadratic and cubic parameterizations (Fig. 8), the cubic 328 parameterization implyies higher air-sea fluxes for high winds, whereas theile quadratic ones lead to 329 higher fluxes for weaker winds. This difference can be presented in arithmetic terms. Let us assume 330 331 two functions of wind speed U, $F_1(U)$ quadratic and $F_2(U)$ cubic:

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

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

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337 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 winds 341 less U_x . U_x is the value of wind speed for which the two functions intersect. In the case of equations 342 (6) and (7), where a = 0.251 and b = 0.0283, they imply that $U_x = 8.87$ m s⁻¹. In fact all of the 343 functions presented in Fig. 8 produce very similar values for U_x , all of which are close to 9 m s⁻¹. 344 This value is very close to average wind speed in the North Atlantic (Fig. 3). This is one of the 345 346 reasons of the small relative difference in net air-sea fluxes. The spread of flux values for the 347 Southern Ocean seems to support this conclusion, being larger than that in the North Atlantic-one. 348 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. 349 350 The other reason of smaller relative differences between the parameterizations in the North Atlantic 351 than globally, is the lack of seasonal variation in the sign of the air-sea flux. In the case of seasonal 352 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 353 winds in winter and weak in summer), the air-sea fluxes partly cancel each other while the 354 difference between cubic and quadratic parameterizations add to each other due to simultaneous 355 356 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 357 358 communication) but we are unaware of any paper investigating it or even describing it explicitly.

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360 In addition to the five parameterizations described above, we calculated the air-sea fluxes using the OceanFlux GHG Evolution combined formula (Goddijn-Murphy et al., 2016), which parameterises 361 362 the contributions from direct and bubble-mediated gas transfer into separate components. The 363 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 364 mediated term of Fangohr and Woolf (2007) is overestimating the bubble component, implying the 365 366 need for a dedicated calibration effort. This question will be the subject of further studies in the 367 OceanFlux GHG Evolution project.

369 Although, uUsing both Takahashi et al. (2009) climatology and SOCAT datasets pCO₂ climatology (Fig. 7) results in similar annual net air-sea CO_2 fluxes in the North Atlantic, it should be noted that 370 they show different seasonal variations. This may have been caused by slightly different time 371 372 periods of the datasets (i.e. the SOCAT based datasetelimatology contains more recent data). One 373 have to remember that at present most of data from Takahashi et al. (2009) are included in SOCAT, 374 so <u>T</u>the differences, is much larger in the European Arctic, may be due to the underlying sparse 375 data coverage and possible interpolation artifacts (Goddijn-Murphy et al., 2015) as well as 376 processing of the data through the FluxEngine. The results are improved in Courtney et al. (2016) where modeled and observation data were compared and has been show the same relationships in 377 378 high-latitude zone. This discrepancy makes us treat the net air-sea CO₂ fluxes results from the 379 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, 380 new data, not included in the SOCAT versions we used, such data have been recently available to 381 the recent analysis by published (Yasunaka et al., (2016). The observed in-water pCO_2 data (Fig. 3) 382 383 in Yasunaka et al., 2016), especially since 2005, show clearly an annual cycle compatible with the SOCAT seasonal flux variability. 384

385386 5. Conclusions

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388 In this paper we have studied the effect of the choice of gas transfer velocity parameterization on the net CO₂ air-sea gas fluxes in the North Atlantic and the European Arctic using the recently 389 390 developed FluxEngine software. The results show that the uncertainty caused by the choice of the k391 formula is smaller in the North Atlantic and in the Arctic than it is globally. The difference in the annual net air-sea CO₂ fluxes caused by the choice of the parameterization is within 5% in the 392 393 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 Arctic 394 and 65% globally when comparing cubic and quadratic functions. In both cases the uncertainty in 395 the North Atlantic and the Arctic regions are smaller than the global case. We explain that the 396 397 smaller North Atlantic variability to be is the a -combination of, firstly, higher than global average wind speeds in the North Atlantic, close to 9 m s⁻¹, which is the wind speed at which most k 398 399 parameterization have similar values, and secondly the all-season CO₂ sink conditions in most 400 North Atlantic areas. We repeated the analysis using Takahashi et al. (2009) and a SOCAT pCO_2 401 derived climatology and find that although the seasonal variability in the North Atlantic is different.

- 402 the annual net air-sea CO_2 fluxes are within 8% in the North Atlantic and 19% in the European 403 Arctic. The seasonal flux calculated from the two *p*CO2 datasets in the Arctic have inverse seasonal 404 variations, indicating possible under sampling (aliasing) of the *p*CO₂ in this polar region and 405 therefore highlighting the need to collect more polar *p*CO₂ observations in all months and seasons.
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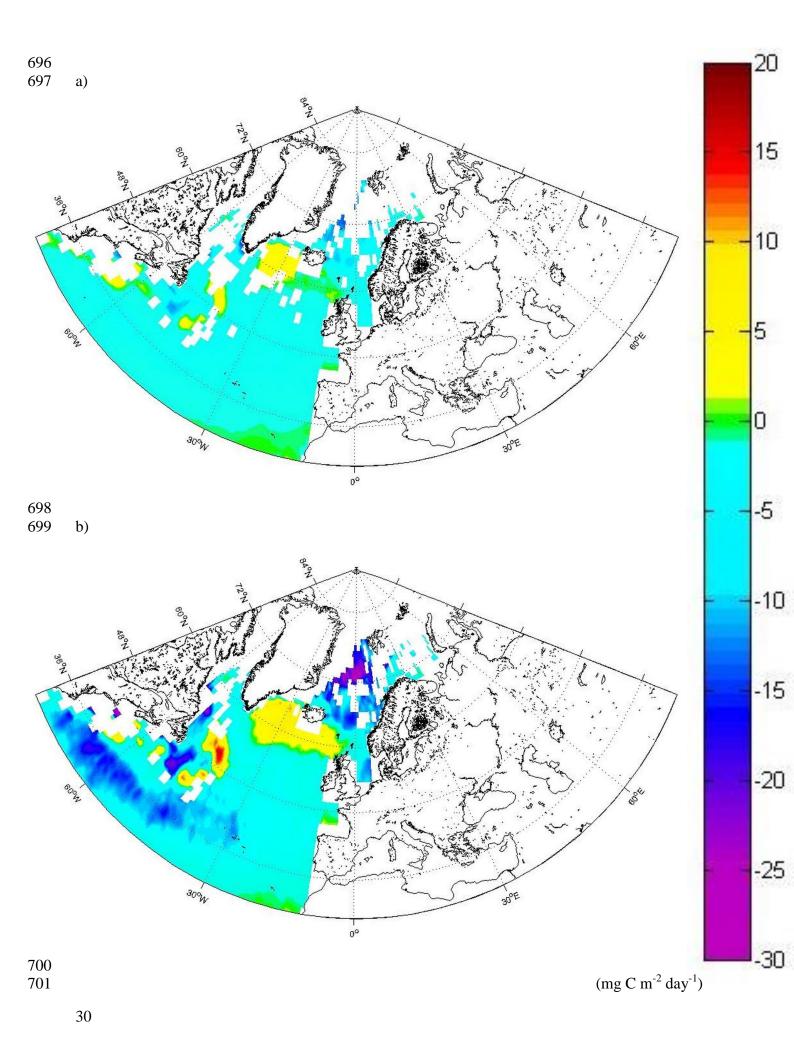
- Figure 1. Seasonal and annual mean air-sea fluxes of CO_2 (mg C m⁻² day⁻¹) in the North Atlantic, using Nightingale et al. (2000) *k* parameterization and Takahashi <u>et al.</u> (2009) climatology-<u>in</u> a) annual, b) DJF (<u>w</u>Winter), c) MAM (<u>s</u>Spring), d) JJA (<u>s</u>Summer), -e) SON (<u>a</u>Autumn). The gaps (white areas) are due to missing data, land and ice masks.
- 663 | Figure 2. Seasonal and annual $-pCO_2$ values (µatm) in surface waters of the North Atlantic, 664 | estimated using the Takahashi <u>et al.</u> (2009) climatology_in_a) annual, b) DJF (<u>w</u>Winter), c) MAM 665 | (<u>s</u>Spring), d) JJA (<u>s</u>Summer), e) SON (<u>a</u>Autumn). The gaps (white areas) are due to missing data, 666 | land and ice masks.
- 668 Figure 3. Wind speed distribution $-U_{10}$ (ms⁻¹) in the North Atlantic used to determine the 669 relationship between gas transfer velocity and air-sea CO₂ fluxes-in a) annual, b) DJF (<u>w</u>Winter), c) 670 MAM (<u>s</u>Spring), d) JJA (<u>s</u>Summer), e) SON (<u>a</u>Autumn). The gaps (white areas) are due to missing 671 data, land and ice masks.
- Figure 4. Differences maps for the air-sea CO₂ fluxes (mg C m⁻² day⁻¹) -in the North Atlantic,
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- 678Figure 5. Monthly values of CO_2 air-sea fluxes of CO_2 -(Pg month⁻¹) for the five parameterizations679(eq. 4-8) in a) the North Atlantic, b) the European Arctic.680
- 681 Figure 6. –Annual air-sea fluxes of CO_2 –for the five (eq. 4-8) parameterizations as well as for 682 backscatter (default) and wind driven OceanFlux_GHG parameterizations normalized to flux values 683 of Nightingale et al. (2000) *k* parameterization (see text)-in a) globally, b) the North Atlantic c) the 684 European Arctic, d) the Southern Ocean. Average values for all parameterization and standard 685 deviations are marked as vertical gray lines.–
- 687 | Figure 7. Comparison of monthly values fluxes of air-sea CO_2 fluxes calculated with different pCO_2 688 datasets (Takahashi et al., 2009, SOCAT v. 1.5 and 2.0) using the same k parameterization 689 | (Nightingale et al., 2000) in a) the North Atlantic, b) the European Arctic.
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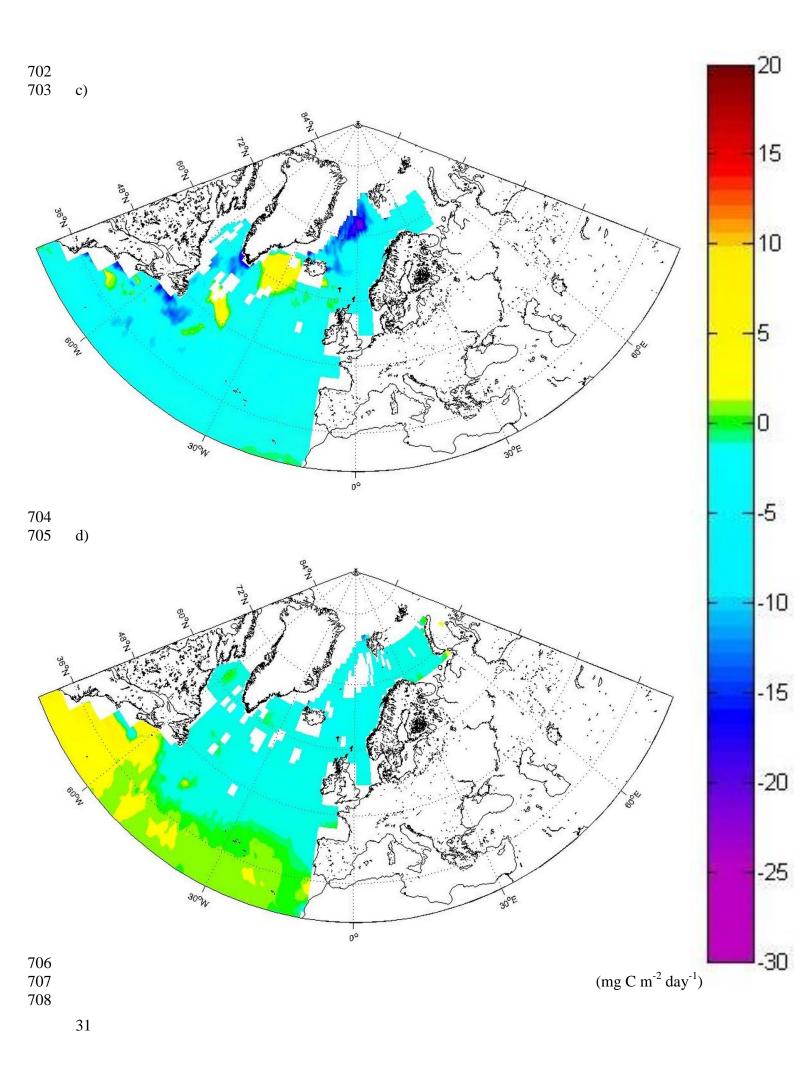
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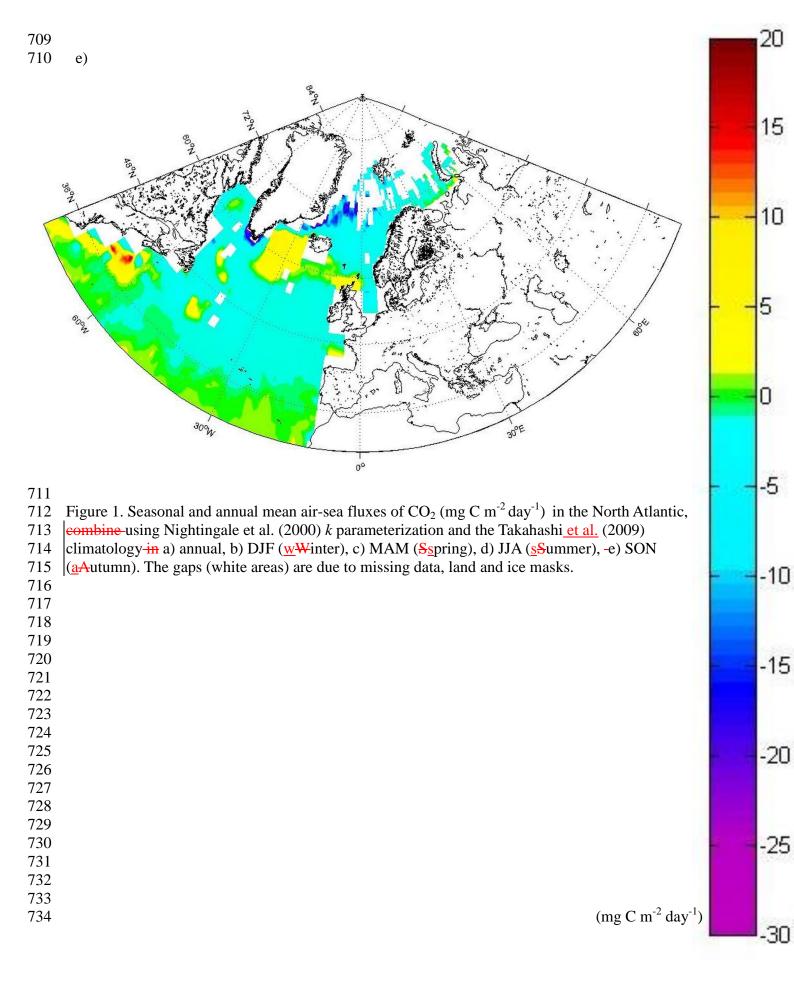
- 691 Figure 8. Different k660 parameterizations as a function of wind speed.
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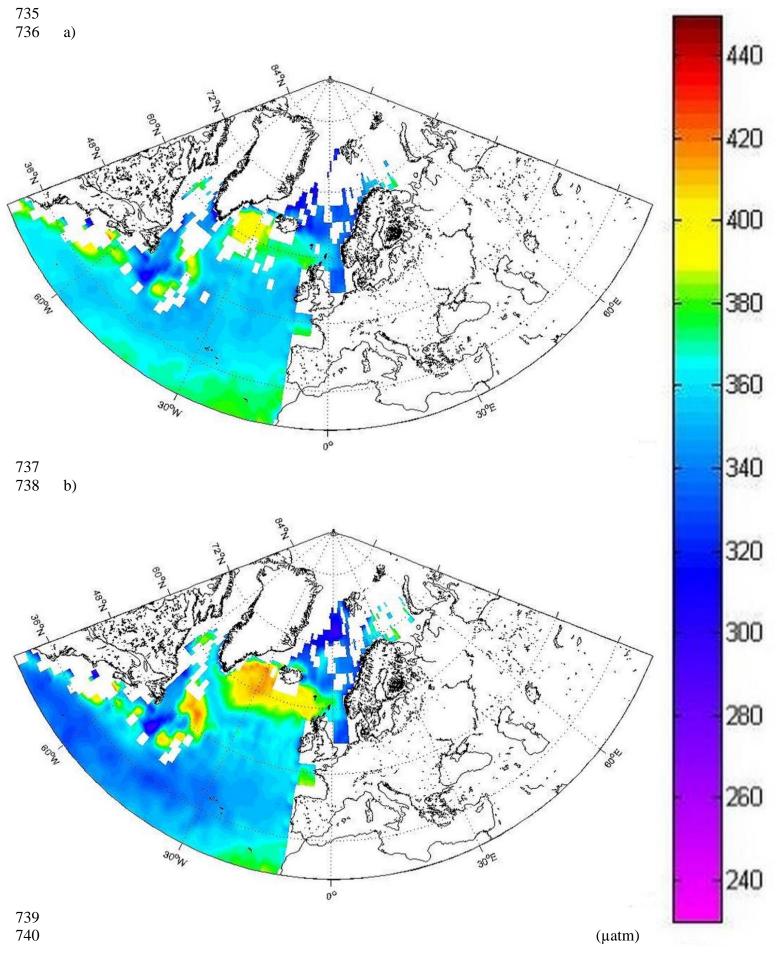
1	Global	Arctic	North Atlantic	Southern Ocean
Nightingale et al., 2000	-1.30	-0.102	-0.382	-0.72
	(1.00)	(1.00)	(1.00)	(1.00)
Ho et al., 2006	-1.42	-0.106	-0.402	-0.76
	(1.09)	(1.04)	(1.05)	(1.06)
Wanninkhof and McGillis, 1999	-1.73	-1.130	-0.490	-0.93
	(1.33)	(1.28)	(1.29)	(1.30)
Wanninkhof, 2014	-1.40	-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)

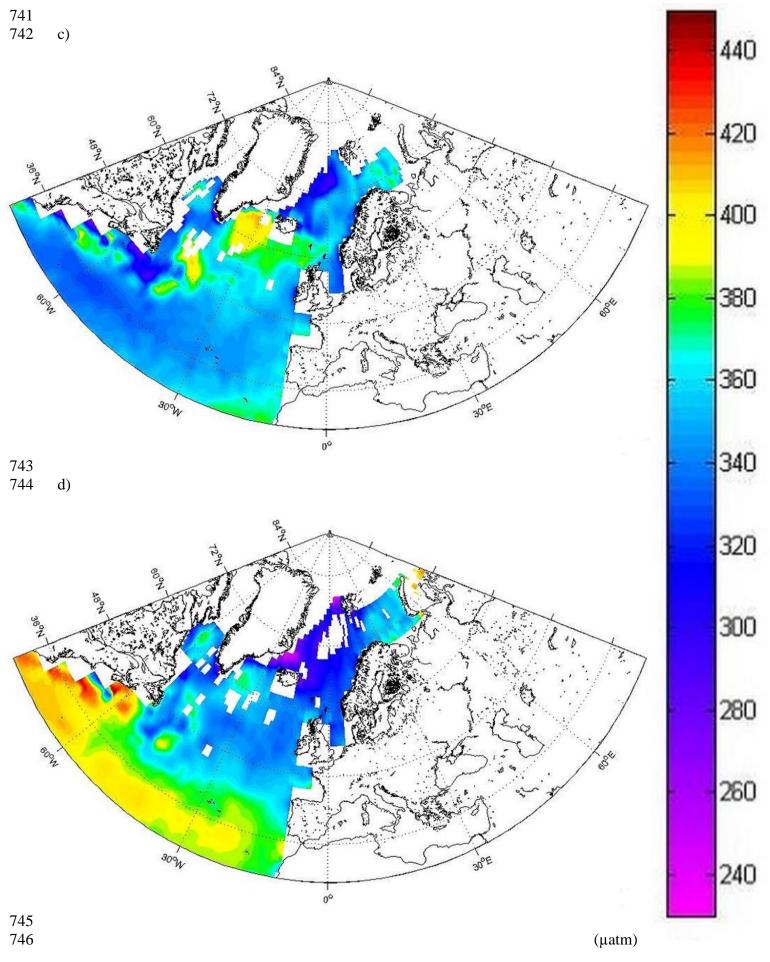
Table <u>1</u>. Annual air-sea $CO_{2_2}^2$ fluxes (in Pg) using different k parameterizations. The values in parentheses are fluxe<u>ds</u> normalized to Nightingale et al., 2000 (as in Fig. 6)











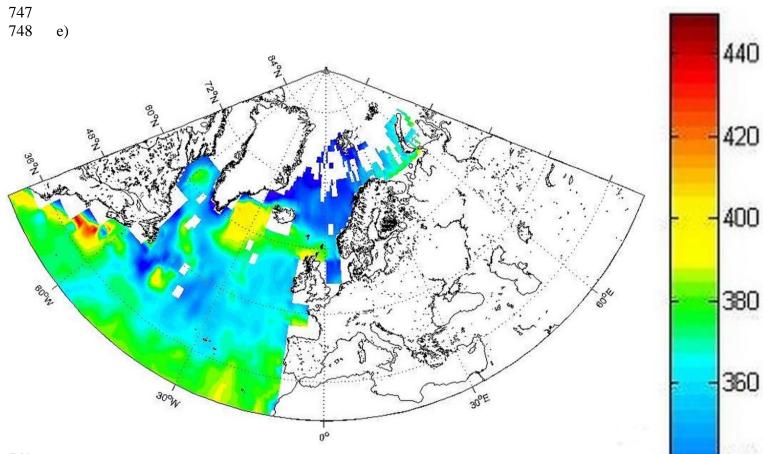
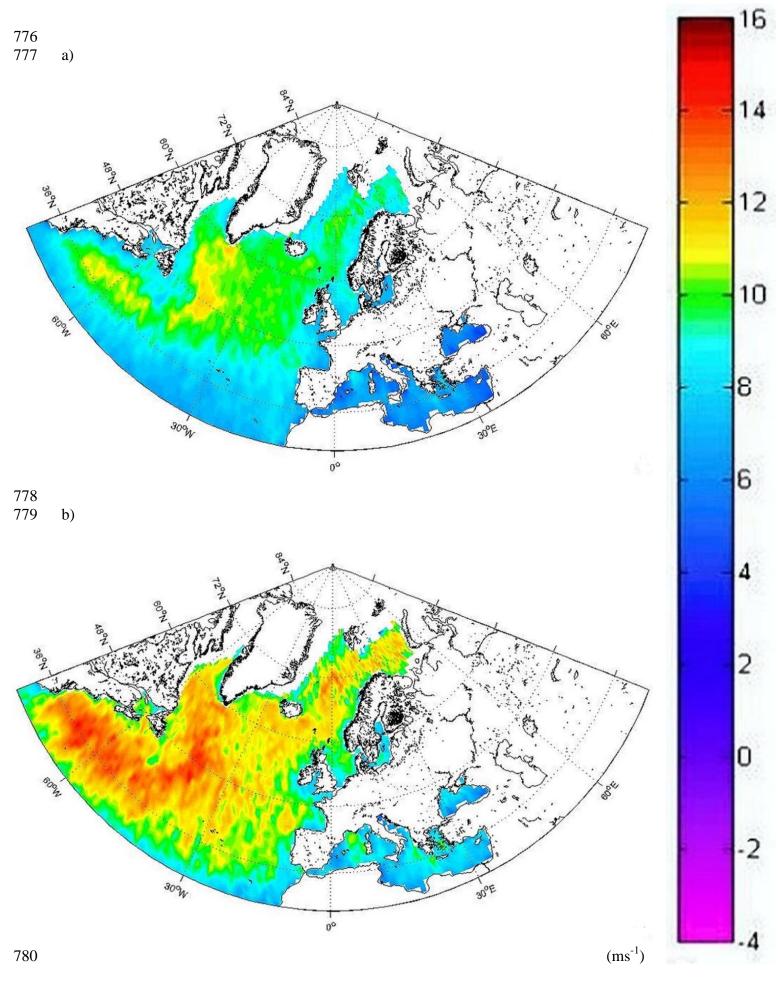


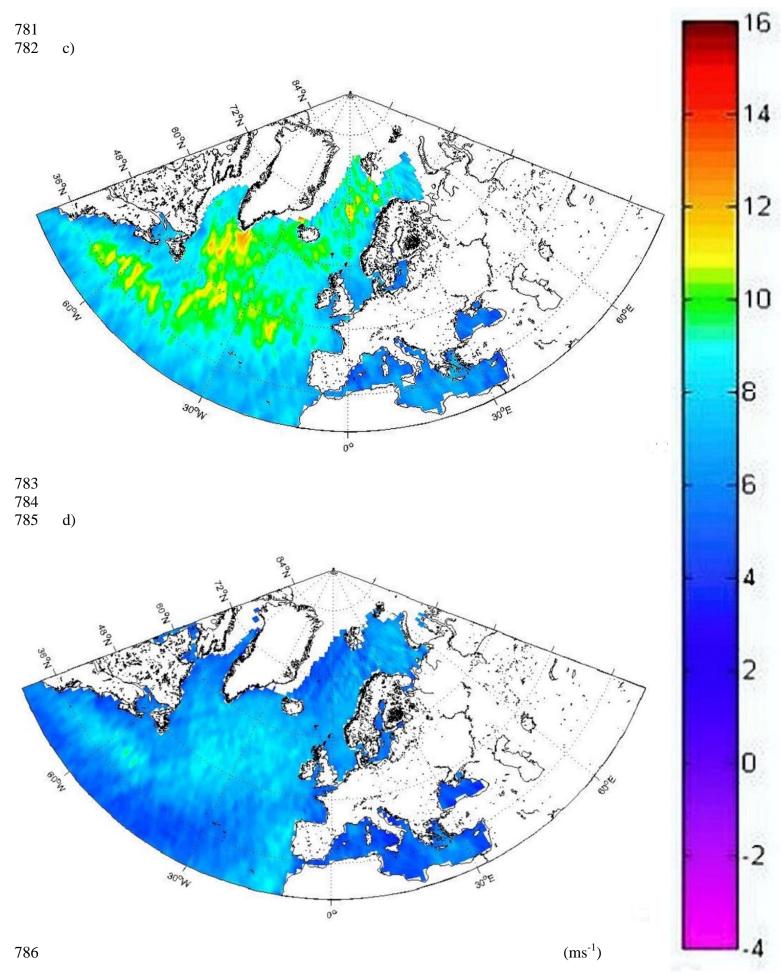
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753 missing data, land and ice masks.









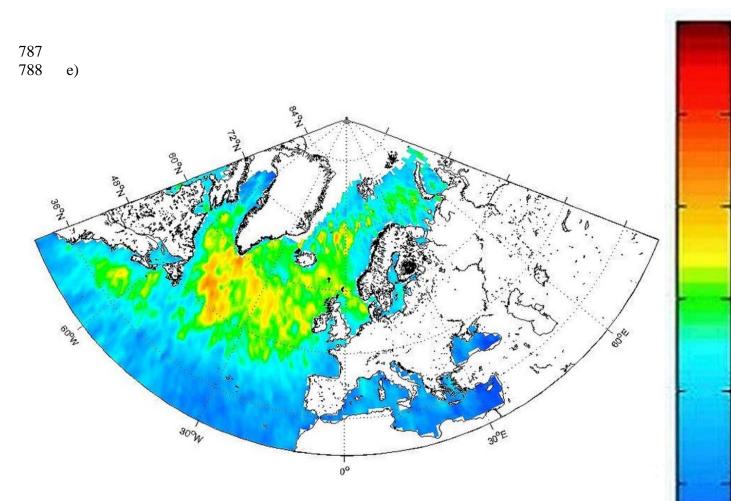
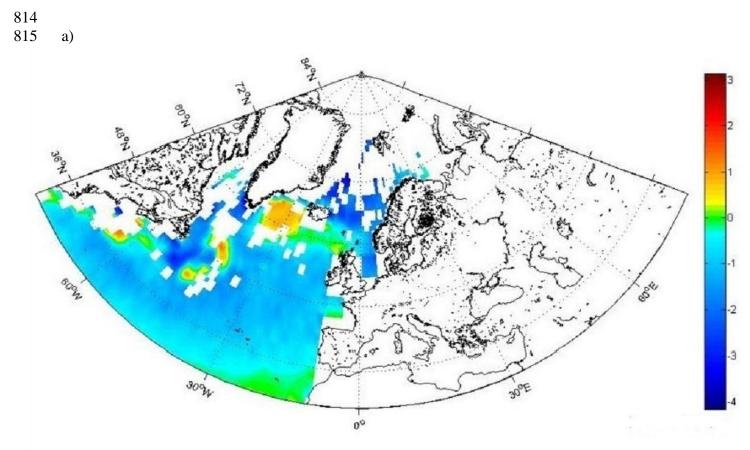
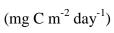


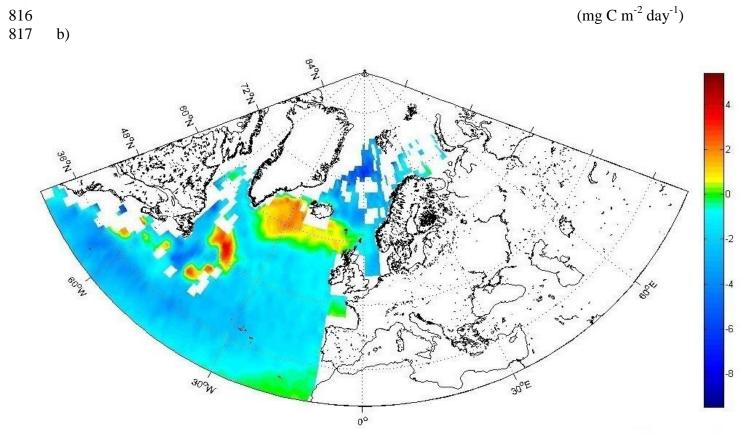
Figure 3. Wind speed distribution U_{10} (ms⁻¹)- in the North Atlantic used to determine the relationship between gas transfer velocity and air-sea CO₂ fluxes-in a) annual, b) DJF (<u>w</u>Winter), c) MAM (<u>s</u>Spring), d) JJA (<u>s</u>Summer), e) SON (<u>a</u>Autumn). The gaps (white areas) are due to missing data, land and ice masks.

(ms⁻¹)

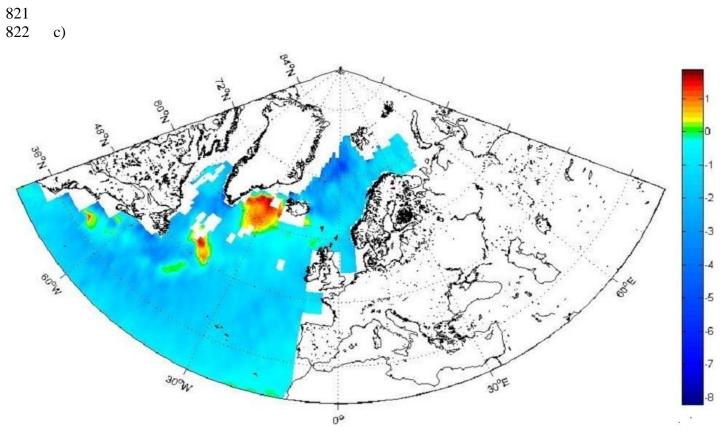


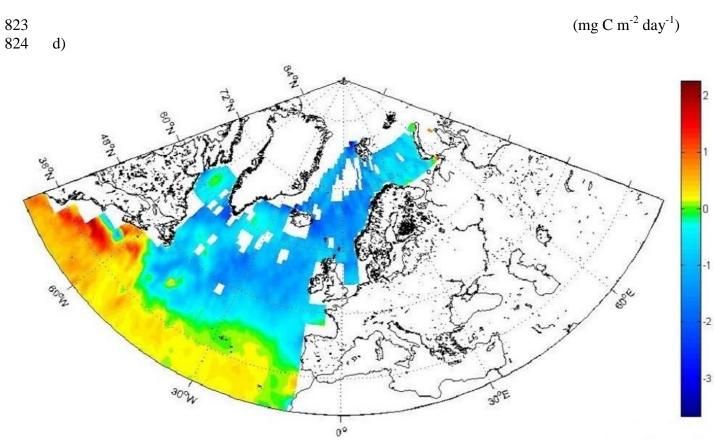


 $(mg C m^{-2} day^{-1})$



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

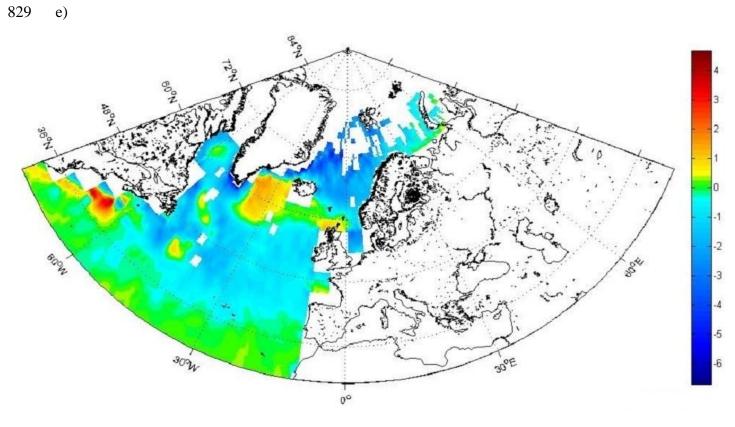
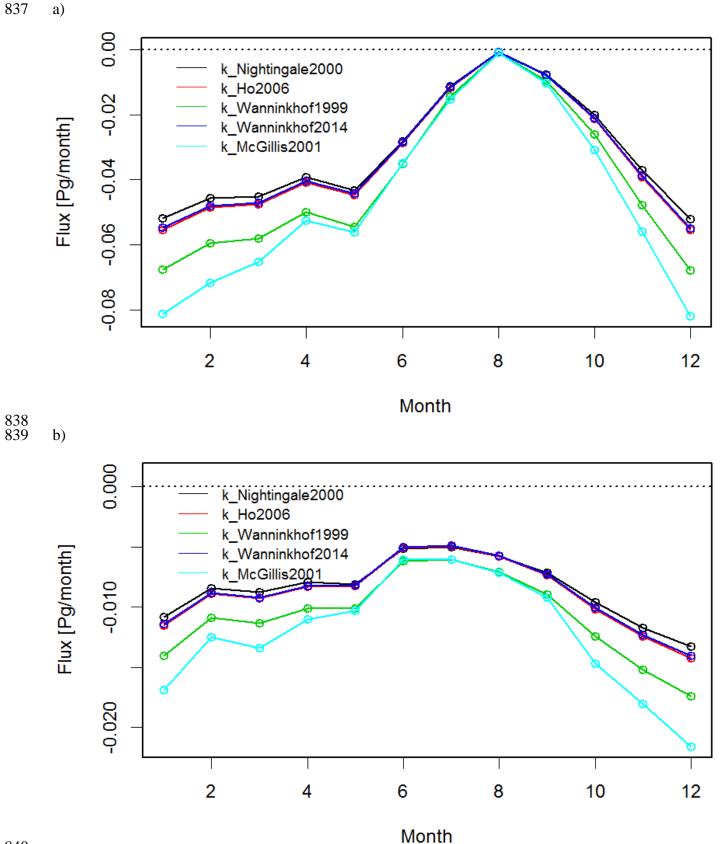


Figure 4. Differences maps for the air-sea CO_2 fluxes (mg C m⁻² day⁻¹) –in the North Atlantic, between a wind–cubed and <u>a</u> squared parameterizations (Wanninkhof and McGillis 1999 and Wanninkhof 2014) in-a) annual, b) DJF (<u>w</u>Winter), c) MAM (<u>s</u>pring), d) JJA (<u>s</u>bummer), e) SON (aAutumn). The gaps (white areas) are due to missing data, land and ice masks.-

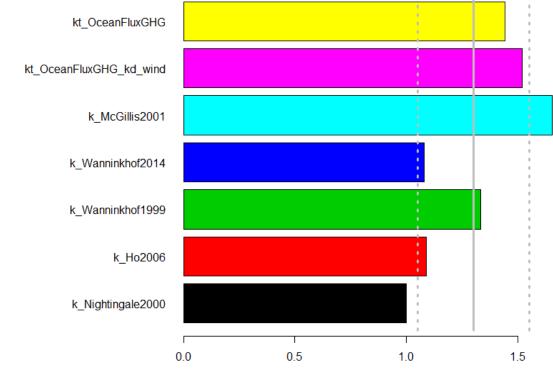
 $⁽mg C m^{-2} day^{-1})$





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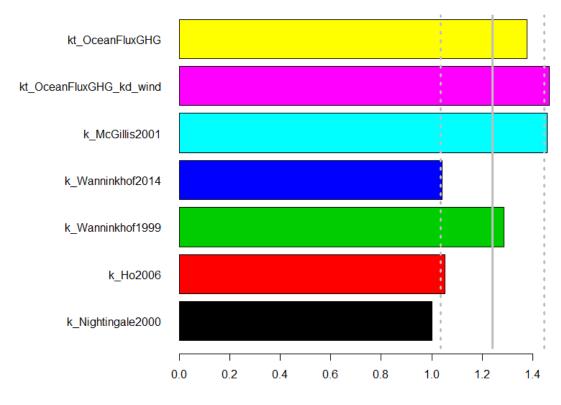
a)



Annual net global CO2 flux normalized to N2000

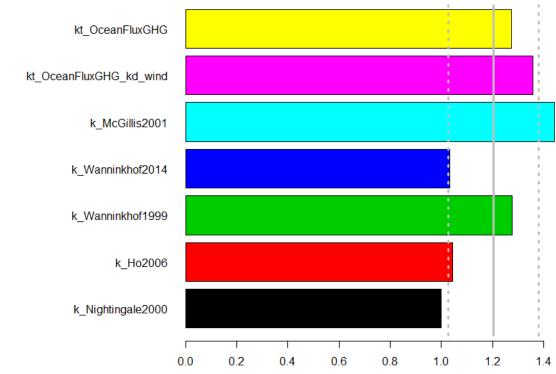
b)

Annual net North Atlantic CO2 flux normalized to N2000



853 c)

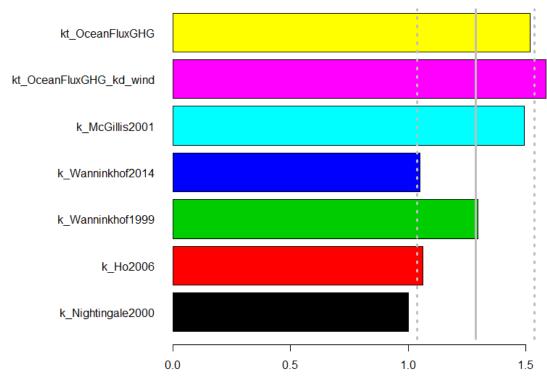
Annual net Arctic CO2 flux normalized to N2000

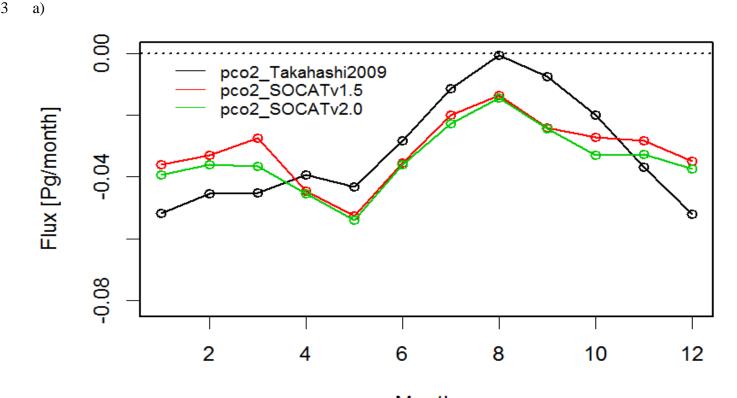


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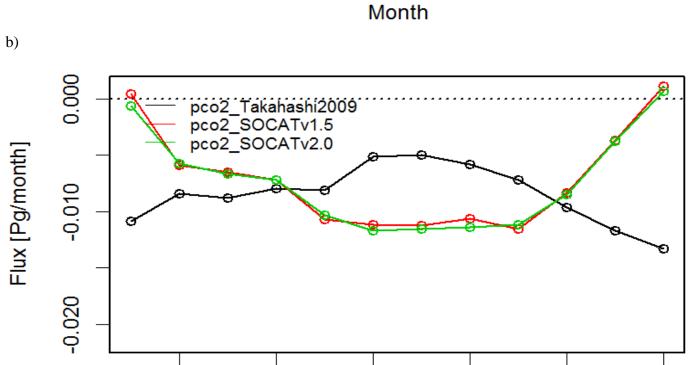
d)

Annual net Southern Ocean CO2 flux normalized to N2000





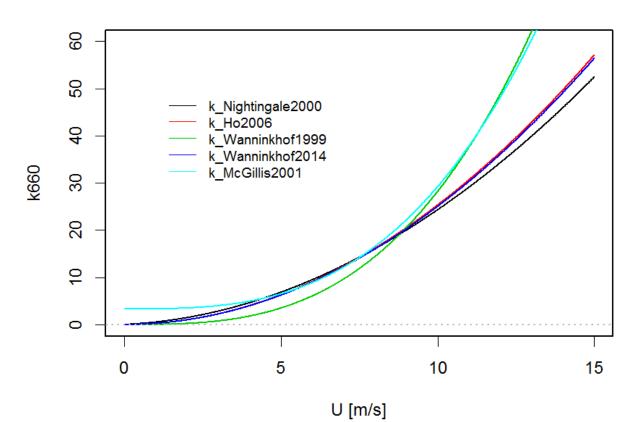
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Month

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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) the North Atlantic, b) the European Arctic.



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