Interactive comment on "Variability of North Atlantic CO_2 fluxes for the 2000-2017 period" by Zhaohui Chen et al.

Zhaohui Chen et al.

<u>Author responses to comments of the 2 referees.</u>

Reviewer comments are listed in italics.

We thank both the reviewers for their detailed comments and suggestions on improvements to the manuscript. Below we list our responses to both sets of reviewer comments.

REVIEWER 1

The authors present an improved atmospheric inversion data assimilation model (GCL) and apply it to the investigation of mean, variability, and trends of North Atlantic air-sea carbon fluxes. Specifically, the advancements made to the inverse model within involve multiple representations of prior ocean fluxes as well as sensitivity experiments assessing the different priors and related flux uncertainties from three different schemes. Additionally, comparisons are made to previous estimates of North Atlantic carbon fluxes as well as estimates from observation-based pCO_2 products and global ocean models. Overall, I found this manuscript well organized, concise, and novel. I would support its publication but have a few suggestions that I believe that would improve the overall strength of the paper.

Reviewer 1: *Main comments*

I highly suggest including additional observation-based products in your analysis (Figure 3 specifically). You use Takahashi et al 2009 in the uncertainty analysis section but can't utilize it for long term mean/variability because it is of course only a climatology. You include 2 products (one by Landschützer and one by Rödenbeck) but there are more available and I highly suggest including them in the comparison to improve your message. Given that your ensemble of inverse models and of GOBMs are much larger than 2, it is worth making the effort to include more pCO_2 products as well. See Denvil-Sommer et al. 2019, Gregor et al. 2019, Iida et al. 2015, and Zeng et al. 2015 for starters.

Response: We agree with the reviewer's suggestion, that inclusion of additional observation-based products would strengthen our analysis. Accordingly we have sourced the following additional global pCO_2-based air-sea CO_2 flux products, and include them in our analyses. The specific data product names, or model versions are appended within parentheses following each published source.

(1) Denvil-Sommer et al. 2019 (product LSCE-FFNN-v1); (2) Iida et al. 2015 (JMA); (3) Zeng et al. 2015 (NIES); (4) Gregor et al. 2019 (CSIR-ML6); (5) Chau et al. 2020 (CMEMS); and (6) Watson et al. 2020.

The above six data products all account for inter-annually varying air-sea CO_2 fluxes (as do our previously included data products from Landschützer et al. (2016) and Rödenbeck et al. (2013)). Updates of our results, accounting for the inclusion of these six additional flux products, are presented in the revised manuscript, and in relevant sections of our Response to Reviewers. Updated results included in this Response to Reviewers include Figure 1, Figure 3 and revised sections of Tables 2, 3, and 4, that now include the additional pCO_2-based flux products.

Updated results for Figure 1 of our study: We present below the original version of Figure 1 (three flux products), along with a revised version (eight flux products).

- Figure 1A: Original version of Figure 1 for the three flux products from Landschützer et al. 2016, Rödenbeck et al. 2014, and Takahashi et al. 2009.
- Figure 1B : Spread-based prior ocean flux uncertainty for an extended set of eight flux products that only include the data products that represent inter-annually varying fluxes (i.e., the climatological product of Takahashi et al. 2009, is not included in the calculations for Figure 1B).

We note that the main features and magnitudes of the spread-based prior flux uncertainty are consistent across the two figures, for example, highest levels of flux uncertainty are associated with the sub-polar North Atlantic region for the winter (DJF) and spring (MAM) months. This similarity is not unexpected given the common underlying pCO_2 database (i.e., SOCAT; Bakker et al., 2016, 2020) used for many of the products. Additional details on these flux products can be found in their related publications and in Friedlingstein et al. (2020).

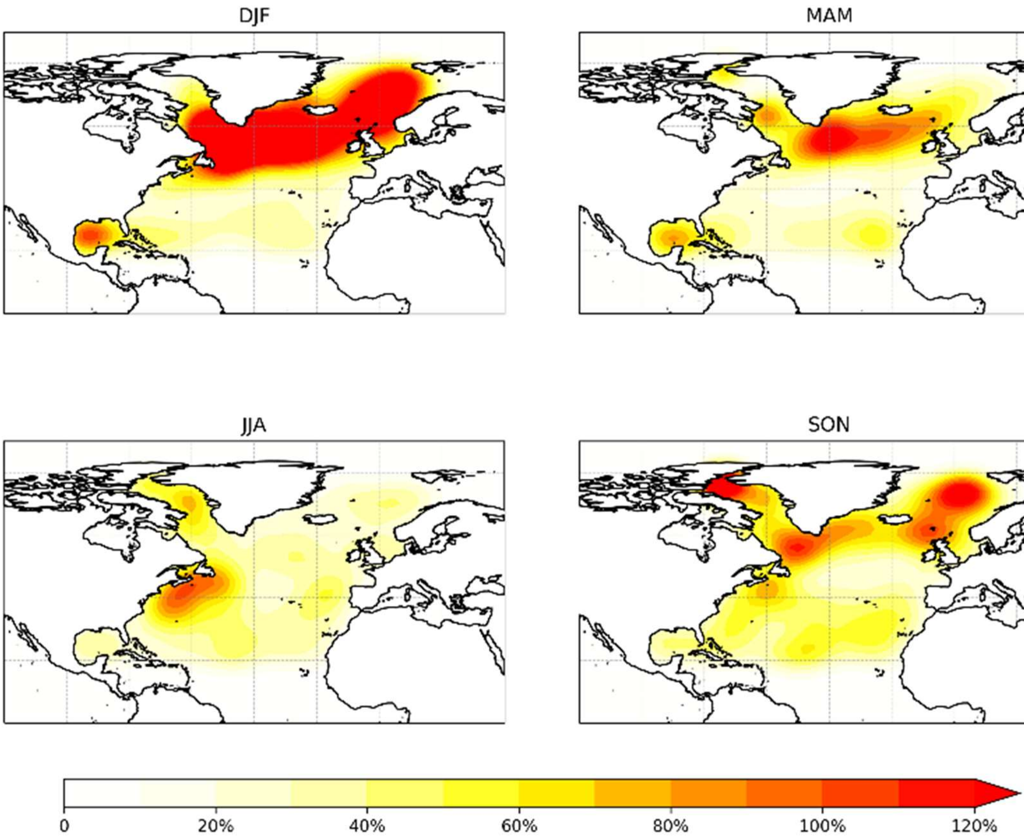

Figure 1A: Original version of Figure 1 from manuscript, derived from three flux products (Landschützer et al. 2016, Rödenbeck et al. 2013, and Takahashi et al. 2009).

Original caption: "Distribution of the spread-based prior ocean flux uncertainty (U3) (annual average for the year 2003). It is represented here as a percentage of the prior ocean flux. The percentage shown for each grid-cell is derived from the ratio of spread-based prior ocean uncertainty divided by the prior ocean flux value at that grid cell. DJF represents the monthly average for December, January, February; MAM for March, April, May; JJA for June, July, August; SON for September, October, November. "

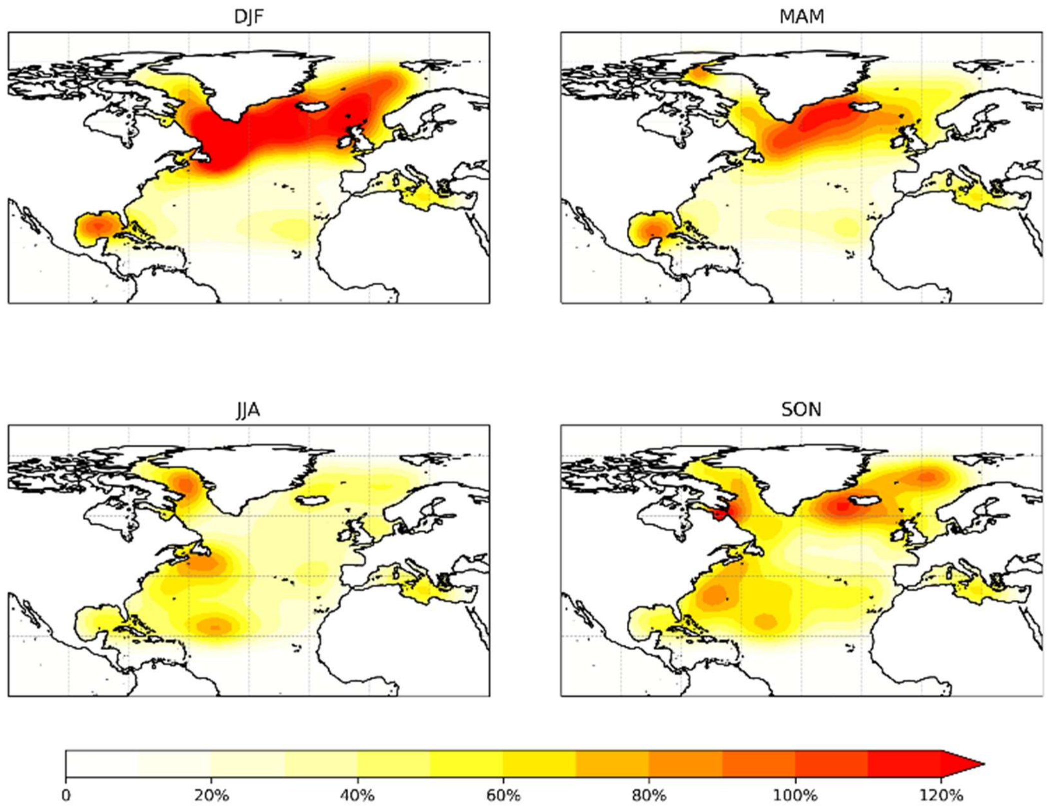

Figure 1B: Revised version of Figure 1.

Distribution of the spread-based prior ocean flux uncertainty (year 2003) calculated from the following 8 flux air-sea CO_2 flux products: (1) Denvil-Sommer et al. 2019 (product LSCE-FFNN-v1); (2) Iida et al. 2015 (JMA); (3) Zeng et al. 2015 (NIES); (4) Gregor et al. 2019 (CSIR-ML6); (5) Chau et al. 2020 (CMEMS); (6) Watson et al. 2020; (7) Landschützer et al. 2016; (8) Rödenbeck et al. 2013.

Reviewer 1: *Additionally, if Landschützer's product is used as the prior for the GCL inverse method, is it fair to use it as an independent comparison? If the data assimilation method is trying to "fit" or "correct" the GCL inverse model to the pCO_2 from that product then I would not consider it an independent comparison.*

Response: In the GEOSChem-LETKF formulation employed in this study, the data assimilated (that the flux estimates are "corrected" by) are the atmospheric observations of CO_2 (described in section 2.4 of our manuscript, and represented by y in the LETKF model equations of the Appendix). The posterior estimates of surface CO_2 fluxes (specified via the analysis state equations A6 and A12) are dependent on the differences between the atmospheric observations (y) and the transport model derived atmospheric concentrations (Hx^b), and the atmospheric observations. The posterior flux estimates also do depend on the prior flux estimates (here Landschutzer et al. 2016), which are represented via the specification of the background state x^b, however we do not consider the Landschutzer et al. (2016) product as an independent product for comparison. One aim of the comparison of the GEOSChem-LETKF posterior fluxes to the Landschutzer product is to assess how much the posterior flux estimates vary from the prior specification following assimilation of the atmospheric CO_2 measurements. For example, in section 3.1, Table 1, we compare the representation of prior and posterior fluxes for three separate representations of the prior ocean flux (namely Landschützer et al. 2016, Rödenbeck et al. 2013, and Takahashi et al. 2009). In our discussion of the multi-year analyses

(2000-2017) of section 3.2, we have noted in the caption of Figure 2 that the prior flux used is that of Landschutzer et al. (2016). We will clarify this in more detail in the discussion of section 3.2 as well.

Reviewer 1: *Section 2.5 could use more discussion/explanation. To my understanding, while the inverse model itself is not new, this method of specifying prior CO_2 fluxes and using them to create more robust flux uncertainties seems to be a major improvement described in this manuscript. I'd be keen to see more explanation and discussion on that in this section.*

Response:

To address this reviewer comment we have expanded the discussion of the spread-based uncertainty scheme in Section 2.4, as listed below. This discussion also includes the extended list of pCO_2-based flux products used in our updated analyses.

"Many previous atmospheric inverse estimates of air–sea carbon fluxes have employed relatively simple characterizations of the prior ocean flux uncertainty, for example, based on a fixed proportion of the grid–scale or regional prior flux (Nassar et al., 2011, Liu et al., 2016, Feng et al., 2016). In Section 3.1, we employ both fixed flux uncertainties, and also present an alternative scheme derived from the ensemble spread of ocean CO_2 flux products, as described below.

The prior ocean flux distributions employed in atmospheric inversions are frequently derived from interpolations of the surface ocean pCO_2 database (e.g., SOCAT, Bakker et al., 2016) in combination with ocean–atmosphere gas exchange parameterizations. Uncertainties in the derived products stem from uncertainties in the input data (e.g., density of measurements), interpolation methods, and gas–transfer parameterizations (Landschutzer et al., 2013). However, some ocean regions, the North Atlantic in particular, have a higher density of pCO_2 measurements and more consistent flux estimates from pCO_2–based products (Schuster et al., 2013, Landschutzer et al., 2013). Here we exploit the recent expansion of pCO_2–based ocean flux products to outline a new specification of ocean prior flux uncertainty based on the ensemble-spread of the different flux products (the "spread–based" uncertainty scheme). Towards the development of the spread-based scheme, we have compiled a set of eight global gridded interannually varying ocean-atmosphere CO_2 flux products. These are Landschutzer et al., 2016, Rodenbeck et al., 2014, Denvil–Sommer et al., 2019, Iida et al., 2015, Zeng et al., 2015, Gregor et al., 2019, Chau et al., 2020, and Watson et al., 2020.

The spread–based prior flux uncertainty scheme uses a diagnostic derived from the variation among the set of ocean atmosphere carbon flux products (see Eq. (1)). This scheme specifies lower uncertainty levels where alternative prior flux representations are in accord (e.g., when well–constrained by availability of surface pCO_2 measurements), and higher uncertainty levels where the prior flux distributions differ significantly (typically in under–sampled regions or those of significant flux variability). This specification follows previously used methods to characterize uncertainties in ocean flux distributions (e.g., Bopp et al., 2013)."

Reviewer 1: *Lastly, I think the title could be more descriptive of the actual work you are presenting. Specifically mentioning inverse models or uncertainty or a comparison between approaches.*

Response: We propose to change the title of the study to "Variability of North Atlantic CO_2 fluxes for the 2000–2017 period estimated from atmospheric inverse analyses"

Reviewer 1: Minor comments

Reviewer 1: The end of the introduction could improve from a motivation statement. Why do this work? Who will use this? How will it impact the community and what are the broader impacts? Clearly your improvements on the uncertainty estimates would be beneficial to the community as a whole so make that case more clearly.

Response: To address this comment we have expanded the discussion of the spread-based uncertainty scheme in the Introduction as follows (section following line 55):

"Previous studies also note that estimates of carbon fluxes from the atmospheric inverse method are sensitive to the specification of the prior flux distribution and its associated uncertainty distribution (Carouge et al., 2010; Chatterjee et al., 2013; Peylin et al., 2013). While there have been recent studies evaluating the sensitivity of land–based carbon flux estimates to specification of the prior flux and its uncertainty, there has been far less examination of ocean flux estimates from inverse methods. Several global inverse model assessments of the past decade have relied on the climatological ocean–atmosphere CO_2 flux database of Takahashi et al. (2009) to specify prior ocean fluxes. In view of the limited information available on the temporal and spatial variability of ocean carbon fluxes from this climatological ocean database, these inverse analyses have adopted different approaches to the specification of prior uncertainty for ocean fluxes, ranging from uncertainties derived from a separate ocean model inversion (in the case of Nassar et al., 2011), to a specified percentage of the prior flux magnitude (Feng et al., 2016, Liu et al. 2016).

In this study we present a new long term estimate of North Atlantic air–sea CO_2 fluxes for recent decades (period 2000–2017) using atmospheric inverse methods. We focus in particular on the specification of prior ocean fluxes (including sensitivity of flux estimates to alternative prior flux distributions) and on their associated flux uncertainties. To our knowledge these influences on inverse estimates of North Atlantic CO_2 flux have not been assessed previously. We use the carbon cycle data assimilation system GEOSChem–LETKF (GCL, described further in Section 2) which combines the global atmospheric CO_2 transport model GEOS–Chem (Nassar et al., 2010) with the Localized Ensemble Transform Kalman Filter (LETKF) data assimilation system (Hunt et al., 2007; Miyoshi et al., 2007; Liu et al., 2019). In recent years several new global air-sea CO2 flux products have been developed based on mappings of ocean surface pCO_2 measurements (e.g., Landschutzer et al., 2016, Rodenbeck et al., 2014, Watson et al. 2020, and products reported in the intercomparison of Roedenbeck et al. 2015). These ocean flux distributions are frequently derived from interpolations of surface ocean pCO_2 measurements from the SOCAT database (Bakker et al., 2016) together with parameterizations of air–sea gas exchange. Following recent updates, the surface ocean pCO_2 database SOCATv2020 (https://www.socat.info/index.php/data-access/), now includes over 28 million surface ocean carbon measurements. The SOCAT database provides a valuable resource towards the development of bottom–up estimates of ocean–atmosphere CO_2 fluxes, and a compilation of these flux products is reported in the recent Global Carbon Budget (Friedlingstein et al., 2020). The increased range of global air-sea CO_2 flux products available (beyond the Takahashi et al. 2009 climatology) provides a valuable opportunity to develop an improved representation of air-sea CO_2 flux variability and a more robust characterization of the uncertainties associated with ocean carbon fluxes. In this study we employ some of the recently developed ocean CO_2 flux products to provide a new method of characterizing the prior ocean flux uncertainty used for atmospheric inverse analyses. The methodology is based on the ensemble spread of the multiple ocean flux products, and reflects underlying uncertainties in these products, such as those associated with sampling density of the surface measurements and interpolation method employed. It provides a spatially and temporally variable specification of prior flux uncertainty that will be of value to the inverse modeling community.
"

Reviewer 1: *Why is year 2003 selected for sensitivity tests on the prior flux uncertainty? Is three years of spin up sufficient? Is 2003 an anomalous year at all? With the dynamics at play in the North Atlantic basin it is important to consider how the selection of one year of focus can influence your analysis.*

Response: We have added more information about the selection of year 2003 in section 3.1, as follows:

"Sensitivity analyses are conducted for the year 2003, following a 3 year GEOSChem model spin-up, starting from January 1st, 2000; the length of spin-up was determined by recommendations on the duration required for stabilization of tropospheric CO_2 gradients (e.g., Gurney et al. 2002), and following methods used for previous GEOSChem CO_2 analyses (e.g., Nassar et al. 2010). The year 2003 was selected for sensitivity tests as the first viable year following spin-up. Analyses of inter-annual variability in Atlantic CO_2 (e.g., Landschutzer et al. 2013; Schuster et al. 2013) do not find 2003 to be an anomalous year for regional ocean fluxes."

Reviewer 1*: It could be clarified that when you move on from Section 3.1 you will only be using the U3 approach to specify uncertainty. Additionally, the same for your selection of*

Landschützer et al. 2017 as the prior for the GCL model as you transition to analysis in Section 3.2.
Response: We have added this clarification to Section 3.1, and have provided further justification for the selection of the U3 scheme using assessment metrics derived from model-observation differences at the NOAA network sites. This section now reads as follows:

"The U3 flux uncertainty specification is derived from the variation among a set of ocean–atmosphere carbon flux products (Eq. (1)). This scheme specifies lower uncertainty levels where alternative prior flux representations are in accord (e.g., when well constrained by availability of surface pCO_2 measurements, as in the subtropical North Atlantic), and higher uncertainty levels where the prior flux distributions differ significantly (typically in under-sampled regions or those of significant flux variability, such as the subpolar North Atlantic). We further assess the value of the U3 scheme using a metric of GCL modeled atmospheric CO_2 concentration; specifically, estimates of the model–observation mismatch for the year 2003 at the NOAA network station sites in the North Atlantic using the a posteriori fluxes associated with the sensitivity analyses of this section (Appendix Table A2). The results summarized in Table A2 indicates that scheme U3 provides the smallest magnitude model-observation mismatch for the individual North Atlantic sites and for the global network average. For the long term analyses of the remainder of this study, therefore, we use the U3 spread–based flux uncertainty scheme in preference to the fixed level flux uncertainty schemes used in many previous inverse analyses."

Reviewer 1: *Figure 2: I find it very interesting that the CTE is so anomalous in the NA subtropics but the CT model is more anomalous in the subpolar regions. It jumps out at you from this figure and you barely notice anything else. Would be worth further discussion as to why those are so different in their mean, IAV, and decadal variability. What do these other inverse methods use as prior flux inputs?*

Response:

Since our previous submission of this manuscript, the model results reported for the CTE model for this period (2000-2017) have been updated by the model investigators. Updates include a change in the ocean prior fluxes used for this model (to Roedenbeck et al. 2003), and the updated CTE model configuration is reported Global Carbon Budget database (Friedlingstein et al. 2020). In our revised analysis for this manuscript revision we use the updated CTE model results, and the behaviour of the CTE in the NA subtropics is now less anomalous.

We explained the anomalous from CT in L307:
A potential reason for the anomalous behaviour of the CT estimate in the North Atlantic is the underlying prior flux uncertainties used in the analysis which give a loose constraint on the prior ocean fluxes and allow the ocean fluxes deviate far from the prior because of the impaction from atmospheric CO_2 signals.

In addition, Peylin et al. (2013) have noted that significant variability in atmospheric inverse IAV estimates is a potential indicator of 'flux leakage', where significant variability of terrestrial carbon fluxes in combination with sparse atmospheric sampling can result in misattribution of carbon flux estimates between land and ocean. See additional discussion of this in our Response to Reviewer 2.

Reviewer 1: *Figure 3: could be cleaned up and simplified by reducing the y-label axis ticks and tick labels. Additionally, on this figure, if the trends in subplots e and f are not significant, consider making the filling color gray or something else to distinguish. They should be noted on the figure as well as in the table. Currently you only note that one of the GCL trends is significant and one is not but need to do this for all inverse models, pCO_2 products, and GOBMs as well.*

Response: We have addressed the reviewer comments on Figure 3. Changes made include: (i) changes to y-axis labels and ticks; (ii) inclusion of the 6 additional pCO_2-based flux products introduced in this revision and discussed above; (iii) changes to plot symbols in panels (e) and (f) to highlight the cases where the derived trends are significant. A revised version of Figure 3 is shown below.

In addition, we have also augmented the tables corresponding to Figure 3, to account for the additional pCO_2 based products. The revised sections of Tables 2, 3, and 4 concerning the additional pCO_2-based flux products are also listed below.

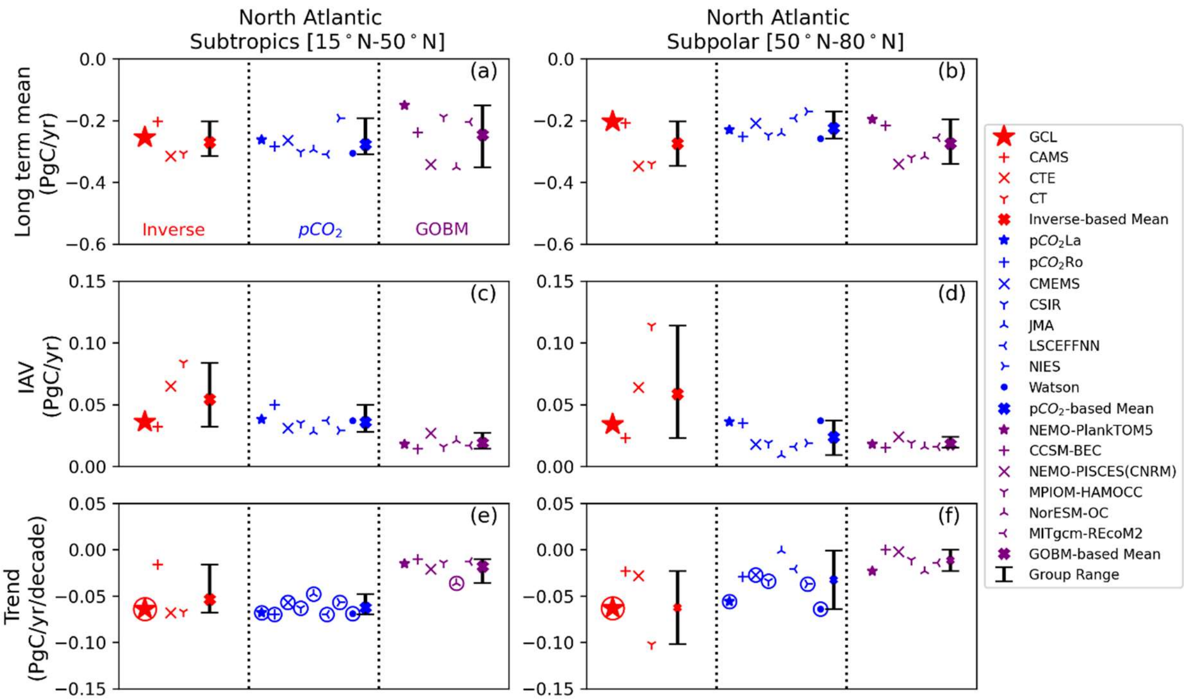

Figure 3. Comparison of CO_2 ocean flux metrics for the 2000−2017 period for North Atlantic subtropics (left panels) and subpolar regions (right panels). Metrics shown are the long term mean (panels (a) and (b)); interannual variability (IAV) (panels (c) and (d)); and long term trend (panels (e) and (f)). The GCL estimates (red stars) are shown in comparison to other atmospheric inverse analyses (red symbols), surface ocean pCO_2 products (blue) and global ocean biogeochemistry models (GOBMs, purple). Also shown are the estimated mean values from each sub–group of analyses (filled cross symbols) with their minimum–maximum range. Circled symbols in panel (e) and (f) indicate a statistically significant trend.

Table 2 revised subsection : Revised section on pCO_2-based flux products to include the six additional interannually varying flux data products.

Long term mean (PgC y^{-1})		
NA Subtropics (15ºN–50ºN)	NA Subpolar (50ºN–80ºN; eastern boundary at 20°E)	
Surface ocean pCO_2-based flux products		
-0.263	-0.23	pCO_2La (Landschutzer et al. 2016)
-0.284	-0.252	pCO_2Ro (Rodenbeck et al. 2013)
-0.264	-0.208	CMEMS (Chau et al. 2020)
-0.302	-0.248	CSIR (Gregor et al. 2019)
-0.295	-0.241	JMA (Iida et al. 2015)
-0.309	-0.192	LSCEFFNN (Denvil-Sommer et al. 2019)

-0.193	-0.171	NIES (Zeng et al. 2015)
-0.305	-0.259	Watson et al. (2020)
[-0.309, -0.193]	[-0.259, -0.171]	Range of all pCO_2–based representations

Table 3 revised subsection: Revised section on pCO_2-based flux products to include the six additional interannually varying flux data products**.**

Interannual Variability (IAV) ($PgC\ y^{-1}$)		
NA Subtropics (15ºN–50ºN)	NA Subpolar (50ºN–80ºN; eastern boundary at 20°E)	
Surface ocean pCO_2–based flux products		
0.038	0.036	pCO_2La (Landschutzer et al. 2016)
0.050	0.035	pCO_2Ro (Rodenbeck et al. 2013)
0.031	0.018	CMEMS (Chau et al. 2020)
0.035	0.020	CSIR (Gregor et al. 2019)
0.028	0.010	JMA (Iida et al. 2015)
0.037	0.017	LSCEFFNN (Denvil-Sommer et al. 2019)
0.029	0.020	NIES (Zeng et al. 2015)
0.037	0.040	Watson et al. (2020)
[0.029, 0.050]	[0.009, 0.037]	Range of all pCO_2–based representations (minimum to maximum)

Table 4 revised subsection: Revised section on pCO_2-based flux products to include the six additional interannually varying flux data products**.**

Trend ($PgC\ y^{-1}\ decade^{-1}$)		
NA Subtropics (15ºN–50ºN)	NA Subpolar (50ºN–80ºN; eastern boundary at 20°E)	
Surface ocean pCO_2-based flux products		
-0.068(S)	-0.056(S)	pCO_2La (Landschutzer et al. 2016)
-0.070(S)	-0.029	pCO_2Ro (Rodenbeck et al. 2013)
-0.057(S)	-0.027(S)	CMEMS (Chau et al. 2020)
-0.063(S)	-0.034(S)	CSIR (Gregor et al. 2019)
-0.048(S)	-0.001	JMA (Iida et al. 2015)
-0.070(S)	-0.021	LSCEFFNN (Denvil-Sommer et al. 2019)
-0.057(S)	-0.037(S)	NIES (Zeng et al. 2015)
-0.069(S)	-0.064(S)	Watson et al. (2020)

[-0.048, -0.070]	[-0.001,-0.064]	Range of all pCO_2–based representations (minimum to maximum)

Reviewer 1: *Throughout the paper, where you mention Figure 3, please add the subplot letter so that the reader can easily navigate to which subplot you are referencing (e.g. Line 268: "Fig 3e, Table 4)". Also, in the Table 2 caption you could reference "Figure 3 a,b" rather than just "Fig 3".*
Response: To address the above reviewer comments, we have made the following changes to the manuscript:
Table 2 caption: changed to "The metrics listed in this table are plotted in Fig. 3 a, b"
Table 3 caption: changed to "The metrics listed in this table are plotted in Fig.3 c, d"
Table 4 caption: changed to "The metrics listed in this table are plotted in Fig. 3 e, f"
Line 225 changed to "Fig. 3a"
Line 236 changed to "Fig. 3 c, d"
Line 269 changed to "Fig. 3 e"

Reviewer 1: *Section 3.2.2. should include further discussion and references explaining why the GOBMs have such low IAV as compared to the other products and inverse models.*
Response: In response to this reviewer comment have added the following discussion to section 3.2.2:

"Recent synthesis studies of global ocean carbon fluxes have noted that GOBMs underestimate the magnitude of IAV in comparison to estimates from pCO_2-based mappings and inverse analyses (DeVries et al.2019, Hauck et al.2020). An important driver of IAV is the variability in biological carbon export; the lower variability observed in the GOBMs could result from opposing changes in biological vs. circulation impacts on carbon export, which potentially reduces the sensitivity of the GOBM air-sea carbon fluxes to climate variability (Landschutzer et al. 2013, DeVries et al. 2019)."

Reviewer 1*: Line 225: Your first comparison is to Schuster et al. 2013 but that work is looking at a very different time period. While it can still be referenced and mentioned, highlighting comparisons that focus on the same decades of analysis is more appropriate.*
Response: We have rewritten this discussion to focus on the estimates from the recent pCO_2-based products and GOBMs, as summarised in our revised versions of Figure 3a and Table 2. We will note that the GCL posterior flux estimate of -0.203 ± 0.037 PgC y^{-1} is in agreement with estimates from the extended set of pCO_2-based products (range of [-0.259,-0.171] PgC y^{-1}), and from GOBMs (range of [-0.341,-0.197] PgC y^{-1}).

Reviewer 1: *Your summary statement on beginning on Line 303 could be expanded on. How is it more "robust"? Is it just smaller uncertainties and significant trends? Perhaps tie in reference to Figure 1 to discuss further.*
Response: We have rewritten sections of the manuscript Summary section to reflect our revised analyses with the extended eight-flux ensemble of air-sea CO_2 fluxes. This section of the Summary discussion now includes the following:

"Our GEOSChem–LETKF estimates also indicate statistically significant trends of increasing CO_2 uptake for the North Atlantic subtropical and subpolar region (estimated trend of -0.064 ± 0.007 and -0.063 ± 0.008 PgC y^{-1} decade⁻

[1] respectively). These trends are of comparable magnitude to those estimated from surface pCO_2–based flux products, but much larger than those derived from global ocean biogeochemistry models for the 2000-2017 period. Estimates of inter-annual variability and long term trends derived from our GEOSChem–LETKF analyses are generally more robust for the North Atlantic subtropics than for the subpolar region, and characterized by smaller uncertainty bounds. Limiting factors affecting estimates for the North Atlantic subpolar region include higher levels of uncertainty associated with specification of prior fluxes (Figure 1), and the observational uncertainty at the atmospheric measurement CO_2 sites in these high northern latitudes (Table A1)."

Interactive comment on “Variability of North Atlantic CO_2 fluxes for the 2000-2017 period” by Zhaohui Chen et al.

RESPONSE TO REVIEWER 2

Reviewer 2*: “The authors have studied the CO_2 annual fluxes in the North Atlantic during an 18-yr period with an atmospheric inverse modelling approach. They show some agreement with other estimates and present a sensitivity study with respect to the prior ocean flux constraint. The topic is obviously of great interest but the actual paper is rather deceiving, with little scientific depth. I am listing here important questions that are fully in the paper scope but that seem to be left open. How significant are the presented sensitivity tests for the inversion community? Despite a subsection and an appendix devoted to it, the description of the data assimilation system is unclear on what matters in practice. My interpretation of l. 95 is that the elementary assimilation window of the LETKF is of four weeks, a period which is too short (given mixing time scales in the atmosphere) to allow a clear distinction between the uncertainty in the prior initial state of atmospheric CO_2 and the uncertainty in the prior surface fluxes, when assimilating atmospheric measurements. The authors should therefore not separate the two. However, Incidentally, in the legend of Eq. A1 in the appendix, we understand that the uncertainty in the initial state of atmospheric CO_2 has been neglected. This rough simplification makes it hard to interpret B, officially the flux covariance matrix, in these terms.*

Response: We thank the reviewer for their comments on our manuscript. To respond to the reviewer comments above, we have added additional detail on the LETKF data assimilation system to section 2.2 and to the Appendix, to clarify our description of the system. Here we provide a summary of the model description and further information on the assimilation window in response to the reviewer’s specific questions. This information is also presented in Section 2.2 of the revised manuscript. Additional detail on the specification of prior flux uncertainties is provided in our responses to Reviewer 1 above, and in further responses to Reviewer 2 (below).

The GEOS–Chem atmospheric chemistry transport model has been used in a range of previous investigations into atmospheric CO_2 and applied in conjunction with inverse analyses to estimate surface carbon fluxes (Nassar et al., 2010, 2011; Suntharalingam et al., 2005; Liu et al., 2016). In this analysis we employ GEOSChem v11–01 at a horizontal resolution of 2° latitude by 2.5° longitude, with 47 levels in the vertical. Model transport fields are provided by GEOS–5 assimilated meteorological data from the NASA Global Modeling and Assimilation Office (GMAO, Rienecker et al., 2008). The GEOSChem configuration employed here primarily follows that of Nassar et al. (2011), but with updated representation of prior fluxes; more detail on the prior CO_2 fluxes and uncertainties implemented in this study is given in Section 2.4.
The Localized Ensemble Transform Kalman Filter (LETKF) is a data assimilation system which provides an estimate given a prior (or “background”) estimate of the current state based on past and current data (in this case, the atmospheric CO_2 mole fraction observations). The general framework of the LETKF is described in Hunt et al. (2007); it has been adapted by Miyoshi et al. (2007) to provide gridscale localized analysis of flux estimates. The LETKF system has been used to estimate CO_2 fluxes in a range of previous studies (e.g, Kang et al., 2012; Liu et al., 2016, 2019). More details on the LETKF equation system are provided in Appendix A. The LETKF provides iterative estimates of the time evolution of the system state, x, (here representing the grid-scale surface carbon fluxes). Each step involves a forecast stage (based on a physical model of the system evolution) and a state estimation stage (the ‘analysis’ step), which combines system observations, y, together with the background forecast, x^b, to derive the improved state estimate. The observation operator H provides the mapping from the state space to the observation space; in this study H is provided by the GEOS–Chem atmospheric model.
In this analysis we employ the complete GEOSChem–LETKF (GCL) data assimilation system to conduct sensitivity analyses on the ocean prior fluxes, and to provide a long term flux estimate of surface CO_2 fluxes for the North Atlantic for the period 2000–2017. We report a posteriori fluxes on monthly timescales for the 2000–2017 period; the optimized monthly fluxes are derived from four sequential weeks of assimilation cycles, as further described below. Our methods follow the implementation of the LETKF system by Liu et al. (2019), who

have extended the previous carbon data assimilation system of Kang et al. (2011, 2012). The study of Kang et al. (2011) assimilated meteorological data and atmospheric CO_2 concentrations to provide estimated atmospheric CO_2 concentrations as part of the state estimate. Kang et al. (2012) extended this method to also provide estimates of surface carbon fluxes. Both these LETKF studies assimilated meteorological data and atmospheric CO_2 concentrations and employed a short assimilation window of 6 hours in order to maintain linear behaviour of the ensemble perturbations (Kang et al., 2011, 2012). In addition, Kang et al. (2012) also tested longer assimilation windows (up to 3 weeks) for LETKF formulations that assimilated atmospheric CO_2 concentrations alone (eliminating the assimilation of the meteorological data). The LETKF system of Liu et al. (2019) extended the Kang et al. (2011, 2012) analyses by incorporating the GEOSChem atmospheric model as the forecast model, along with its representation of surface CO_2 fluxes which provide the prior flux specification for the forecast step. However, Liu et al. (2019) assimilate only atmospheric CO_2 measurements (i.e., no assimilation of meteorological measurements), and use an assimilation window of 7 days; the duration of the assimilation window was selected to maximize the correlation between observations and surface fluxes. The GEOSChem–LETKF system employed in our study follows the Liu et al. (2019) formulation; atmospheric CO_2 measurements are assimilated at 7 day timescales, with the LETKF analysis step providing updates of the surface fluxes and associated uncertainties required as initial conditions for the next weekly forecast step. We report monthly flux estimates following four assimilation cycles. Further details on the LETKF and the governing equations for flux estimation are provided in Appendix A.

Reviewer 2: *Similarly, the authors do not discuss spatial correlations in the prior errors, leaving the impression that they have neglected them as well. How credible is this hypothesis, e.g., among the prior ocean flux products tested here?*

Response: An overview of the prior ocean CO_2 fluxes and uncertainties is presented in section 2.5 of the manuscript. We have extended this section to include clarification on the spatial correlations in prior flux uncertainties, to include the following discussion:

"In this study we account for spatial correlations in the prior ocean fluxes, by inclusion of off-diagonal elements in the background error covariance matrix P^b (Equation A3). We follow the recommendations of Jones et al. (2012) on autocorrelation length scales in the surface ocean. That study derived spatial autocorrelation functions for air-sea fluxes from an analysis of the surface ocean pCO_2 database reported in Takahashi et al. (2009), combined with a gas-exchange parameterization. We currently do not account for spatial correlation in land-fluxes, but will investigate this in future analyses."

Reviewer 2: *How are the ocean flux results presented here affected by the leakage from the land fluxes noted in l. 48? The statement in l. 156 suggests there is none of significance, but without any justification.*

Response: To address this reviewer question we have added further discussion on flux leakage to Section 3.2 of the revised manuscript, as follows:

"We also note that Peylin et al. (2013) have suggested that significant inter–annual variability in atmospheric inverse estimates is a potential indicator of 'flux leakage', where significant variability of terrestrial carbon fluxes in combination with sparse atmospheric sampling can result in misattribution of carbon flux estimates between land and ocean. To assess the significance of flux leakage in our GCL analyses, we have calculated estimates of the diagnostic recommended by Peylin et al. (2013) (i.e., the correlation between the annual total land and total ocean fluxes) for the Northern Hemisphere as a whole (Equator to 90ºN), and also by latitudinal region. Estimates of this diagnostic are relatively low for our GCL analyses (values of 0.2 and 0.5 for the sub-polar and sub-tropical regions) indicating low potential for flux leakage. As a point of comparison, Peylin et al. (2013) note that six out of eleven atmospheric inverse analyses in their model inter-comparison reported correlation coefficients of greater than 0.5. "

Reviewer 2: *Additionally, a number of points of various important need clarification:*

• L. 27: the 20% value is rather artificial given the fact that the global ocean uptake is made of both sources and sinks.

Response: In our original manuscript the 20% value for North Atlantic carbon uptake was reported as it represented a percentage of global net carbon uptake by the ocean, as calculated for example, by such synthesis projects as the Global Carbon Budget (Friedlingstein et al. 2020). In our revised manuscript we have updated this section to include more recent findings, and also to include updated calculations of North Atlantic carbon uptake derived from our synthesis of the 8 pCO_2-based flux products (see comments in Response to Reviewer 1, and the updated section of Table 2 included in the Response).
Our updated version of this section in the Introduction includes the following information:

"Recent estimates of net air–sea CO_2 fluxes derived from sea surface partial pressure CO_2 measurements (pCO_2) indicate net annual uptake for the North Atlantic over the past decade (2009–2018) with a range of 0.35–0.55 PgC y^{-1} (Landschutzer et al., 2016; Rodenbeck et al., 2013; Zeng et al., 2015; Watson et al., 2020), and equivalent to about 14%–22% of the global net ocean carbon ocean sink reported for this period (Friedlingstein et al. 2020)."

Reviewer 2: *L. 57-8: bad example; the studies mentioned here are not for the same year and therefore should not use the same uncertainty budget for a frozen prior flux distribution anyway, given existing trends in the real fluxes.*
Response: Our aim in using these examples was to illustrate the relatively simple characterizations of prior flux uncertainty in previous inverse assessments of ocean carbon fluxes, due to the limited information available at the time on variability of ocean-atmosphere carbon fluxes. We have rewritten this section to clarify this message. Our revised version of this section includes the following discussion:

"Previous studies also note that estimates of carbon fluxes from the atmospheric inverse method are sensitive to the specification of the prior flux distribution and its associated uncertainty distribution (Carouge et al., 2010; Chatterjee et al., 2013; Peylin et al., 2013). While there have been recent studies evaluating the sensitivity of land–based carbon flux estimates to specification of the prior flux and its uncertainty, there has been far less examination of ocean flux estimates from inverse methods. Several global inverse model assessments of the past decade have relied on the climatological ocean–atmosphere CO_2 flux database of Takahashi et al. (2009) to specify prior ocean fluxes. In view of the limited information available on the temporal and spatial variability of ocean carbon fluxes from this climatological ocean database, these inverse analyses have adopted different approaches to the specification of prior uncertainty for ocean fluxes, ranging from uncertainties derived from a separate ocean model inversion (in the case of Nassar et al., 2011), to a specified percentage of the prior flux magnitude (Feng et al., 2016, Liu et al. 2016)."

Reviewer 2*: L. 91: this is Appendix A, not A1.*
Response: We have corrected it to Appendix A.

Reviewer 2: *L. 121: what is the rationale behind the 60% and 120% values? The authors should relate them to their knowledge of the quality of their prior fluxes, while they make it look arbitrary (except if indeed matrix B is just an ensemble of tuning factors and not an error covariance matrix; see above).*
Response: The selection of the prior uncertainty levels used in the sensitivity analyses of section 3.1 was based on the range of variability seen for the individual prior flux distributions (Takahashi et al. 2009; Landschutzer et al. 2016; and Rödenbeck et al. 2013) for the sub-regions of the North Atlantic. These ranged from average levels of ~60% for the sub-tropical North Atlantic to levels > 120% for the sub-polar North Atlantic, hence we selected a level of U1:60% to characterize the lower sensitivity case, and U2:120% for the higher case. We have also updated our discussion of Section 3.1 to include this information.

Reviewer 2: *L. 127: what is the value of K? I get the impression that only 3 flux products are used here: no standard deviation can be estimated from just three members.*
Response: In our original manuscript K = 3, as we included only three prior flux distributions. In response to suggestions from Reviewer 1, we have now included six other inter-annually varying pCO_2-based flux products in our analyses (see discussion and revised results in our Response to Reviewer 1 above). For the revised version of Figure 1 included in our response above, and in the revised manuscript, K=8 (we have omitted the climatological flux distribution of Takahashi et al. 2009, in the revised version of Figure 1).

Reviewer 2: *L. 140-1: why would the three prior ocean flux distributions have the same uncertainty statistics?*
Response: The uncertainty statistics of the prior ocean flux distributions will be dependent on the uncertainties associated with the respective inputs and methods of constructing the flux products. Ocean-atmosphere carbon flux products derived from surface ocean pCO_2 measurements are generally subject to two main sources of uncertainty: (i) in the specification of the surface CO_2 partial pressure difference across the air-sea interface, and (ii) in the specification of the gas-exchange coefficient used to derive fluxes (e.g., see discussion of Landschutzer et al. 2013; Watson et al. 2020). In the extended database of 8 pCO_2-based flux products that we present above in this Response to Reviewers, the majority of the flux products (seven of the eight) rely on the surface ocean pCO_2 data of the SOCAT database (Bakker et al. 2016, 2020). These flux products will be subject to similar uncertainties associated with data coverage in different ocean regions, although the uncertainties due to differences among surface interpolation methods may vary. We have added discussion of these sources of uncertainty to section 3.1.

Reviewer 2: *L. 170: flexibility is not the question. The question is about well modelling the prior uncertainty.*
Response: We have rewritten this section to better describe the aim of using the spread-based uncertainty-scheme. The revised version of section 2.4 includes the following discussion:

"The prior ocean flux distributions employed in atmospheric inversions are frequently derived from interpolations of the surface ocean pCO_2 database (e.g., SOCAT, Bakker et al., 2016) in combination with ocean–atmosphere gas exchange parameterizations. Uncertainties in the derived products stem from uncertainties in the input data (e.g., density of measurements), interpolation methods, and gas–transfer parameterizations (Landschutzer et al., 2013). However, some ocean regions, the North Atlantic in particular, have a higher density of pCO_2 measurements and more consistent flux estimates from pCO_2–based products (Schuster et al., 2013, Landschutzer et al., 2013). Here we exploit the recent expansion of pCO_2–based ocean flux products to outline a new specification of ocean prior flux uncertainty based on the ensemble-spread of the different flux products (the "spread–based" uncertainty scheme). Towards the development of the spread-based scheme, we have compiled a set of eight global gridded interannually varying ocean-atmosphere CO_2 flux products. These are Landschutzer et al., 2016, Rodenbeck et al., 2014, Denvil–Sommer et al., 2019, Iida et al., 2015, Zeng et al., 2015, Gregor et al., 2019, Chau et al., 2020, and Watson et al., 2020.
The spread–based prior flux uncertainty scheme uses a diagnostic derived from the variation among the set of ocean atmosphere carbon flux products (see Eq. (1)). This scheme specifies lower uncertainty levels where alternative prior flux representations are in accord (e.g., when well–constrained by availability of surface pCO_2 measurements), and higher uncertainty levels where the prior flux distributions differ significantly (typically in under–sampled regions or those of significant flux variability). This specification follows previously used methods to characterize uncertainties in ocean flux distributions (e.g., Bopp et al., 2013)."

Reviewer 2: *Table 2: if the numbers behind plus/minus signs for the mean values across studies are standard deviations, how can they have been computed on 6, 3, or even 2 members only?*
Response: As noted above our analysis has been extended to 8 inter-annually varying ocean CO2 flux products. Table 2 has also been revised in the manuscript, and as described above in our Response to Reviewer 1. In addition, the ranges shown in Tables 2,3,4 and in Figure 3 now correspond to the minimum to maximum range of the individual sets of (i) atmospheric inverse analyses; (ii) surface pCO2-based products; (iii) global ocean biogeochemistry models.

Reviewer 2:*L. 339: what is the value of L?*
Response: The parameter L represents the horizontal localization radius, and is set to 2000 km for this study, following Liu et al. (2016). The localization radius is used in the LETKF in a latitude-dependent weighting function which characterizes the spatial scale of the region within which atmospheric CO_2 observations are assimilated at each gridpoint (Miyoshi et al. 2007). We have added further clarification of this to section 2.2 and to the Appendix.

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