The mechanisms of North Atlantic CO_2 uptake in a large Earth System Model ensemble: Revisions

We thank the reviewers for their careful consideration of our work and welcome the fact that they agree with us that this manuscript sets up clearly and addresses an important question; "*This is an important question because a decline in ocean uptake of carbon from the atmosphere, as has been hypothesized here and in other regions such as the Southern Ocean, could have important consequences for future changes in climate*." (Reviewer #1) and "*The introduction to this manuscript is well written and compelling. It is a nice review of the state of understanding with respect to mechanisms of North Atlantic carbon uptake*" (Reviewer #2). Below we detail the changes made to address the reviewer' comments in the revised manuscript.

Summary of most substantial changes made to revised manuscript:

- Mechanism validation: New figures (figs. 8 and 9) presenting **evidence from the Earth System model (ESM) alone, that the mechanisms highlighted by the box model are occurring in the ESM ensemble**. These results are described in a **new subsection** 'Evidence for these mechanisms occurring in the Earth System Model ensemble'.
- Box model validation: **New box model validation** in which the box model is tuned to a randomly selected subsample of the ESM ensemble, then used to predict those ESM ensemble members excluded from the tuning exercise. Results presented in the supplementary materials (fig. S4) and described in main text.
- Box model validation: New figure (fig. S5) and associated discussion presenting the **relationships between ESM generated and box-model predicted air-sea flux from each of the three surface boxes**.
- New figure (fig. S6) presenting the **previously un-presented input time series.**
- In response to the increased number of figures and to improve the manuscript's readability **a new supplementary materials document** has been produced to present the validation/supporting figures and associated descriptions. To further improve readability the main manuscript restructured and new subheadings added.

Full changes:

Reviewer #1:

Comment 1) I am still slightly confused about which "CO2 flux" is some times being discussed. Perhaps the authors would consider adopting the natural/anthropogenic/contemporary terminology in key places such as p14552, line 9 "we will explore subpolar North Atlantic CO2 uptake. . ." to be more specific?

Response 1) In all situations we are examining the natural *and* anthropogenic carbon fluxes, however for clarity we presented data as anomalies in the figures where we were directly comparing different simulations and this is where we suspect confusion was arising. We have modified each of the figure captions to make it clear what is being presented. We have also clarified this in the abstract and introduction:

- Page 14552 line 9 (original manuscript): ‘Here we explore subpolar North Atlantic CO_2 uptake across a large ensemble of Earth System Model simulations’ → ‘Here we explore the natural and anthropogenic subpolar North Atlantic CO_2 uptake across a large ensemble of Earth System Model simulations’. And:
- Page 14556 Line 21 (original manuscript): ‘Here we explore the mechanisms controlling ocean CO_2 uptake across a large ensemble of HadCM3 (3rd Hadley Centre Climate Model) based ESMs in which parameters have been systematically varied to efficiently sample a wide range of model behaviours (Lambert et al., 2013)’ → ‘Here we explore the mechanisms controlling natural and anthropogenic ocean CO_2 uptake across a large ensemble of HadCM3 (3rd Hadley Centre Climate Model) based ESMs in which parameters have been systematically varied to efficiently sample a wide range of model behaviours (Lambert et al., 2013)’.

Comment 2) I found the result of the absence of a role of the MOC variability in changes in carbon uptake intriguing (neither short- nor long-timescale filtering had any appreciable effect) especially as it has been implicated in recent observed subpolar North Atlantic CO2 uptake reduction (Perez et al 2013) and is central to the hypothesis for the peak and decline in subpolar North Atlantic CO2 fluxes as presented in Figure 9. Is this lack of sensitivity because the MOC in the ESM doesn’t vary that much? Or is the importance reduced because only ~30% (for case 1, Table 2) actually goes from the low latitude to the high latitude box (even less for case 2 where mixing from below appears more important)? I think perhaps plotting the other box model inputs from the ESM, as in Figure 10 would be useful for the reader.

Response 2) We thank the reviewer for highlighting the fact that our analysis suggests the CO_2 uptake impact of MOC change to be minimal despite the MOC being implicated in recent observed change by Perez et al (2013). Firstly we note that whilst the observational analysis presented in Perez et al. (2013) suggests that ‘the meridional overturning circulation was largely responsible for the [observed] reduction in carbon uptake’ they find that this occurred ’through a reduction of oceanic heat loss to the atmosphere [driven by MOC change]’. In our study this facet of MOC change will be included in the box model temperature input rather than the MOC input – the prescribed MOC simply being used to move DIC between boxes – and therefore would not show up when we hold the MOC constant. We do however find temperature to be an important control on the CO_2 uptake over both timescales (figs 5+11 of the original manuscript).

To answer the reviewer’s question (‘Is this lack of sensitivity because the MOC in the ESM doesn’t vary that much?’) we now present all of the previously un-presented input datasets (including MOC strength time-series) in a new figure, figure S6. Figure S6 shows that the MOC undergoes a significant decline in all ensemble members by 2100, however we note that this range does not span the larger of the MOC reductions simulated within the CMIP5 ensemble (Cheng et al., 2013). It would therefore appear that the relative insensitivity of air-sea CO_2 flux change to MOC reduction relates to both of the reviewers’ points, but primarily the second point - that not all of the water moving from the low to high northern latitudes flows through the surface boxes. This message has been inserted into the last paragraph in the ‘Drivers of multidecadal/centennial mode of variability’ subsection.

Comment 3) on p14557 about numerous idealized simulations suggests running the emulator with different CO2 power-law curves, but these experiments are not presented. Why not instead describe forcing the model with filtered input, which you actually do present?

Response 3) We appreciate that the ‘power-law curves’ statement could cause confusion because we don’t present any of this analysis. Here we wanted to make the point that using the box model you can play about with the system to develop a deep understanding of how it works. Whilst we did this extensively to help us develop our own understudying, we do not present any of this analysis. Consequently we have removed the mention of specific idealised runs (i.e. power-law curves) from the revised manuscript. The bullet point in the manuscript above the one mentioned here describes the sort of analysis that we do present. We have modified this point to also mention filtered input, as suggested by the reviewer: ‘…or filtering input data to remove and isolate the variability of interest’.

Comment 4) Technical corrections: 1.) Typo: p14552, line 17 “Revelle” not “Ravelle” 2.) Figure 1 is never actually referred to. 3.) The abbreviation ESPPE is used on p14577, line 7 but not actually defined until p14558, line 4. 4.) Typo: p14560, line 24 “a time series vary around zero”. 5.) Parameter names in Table 1 do not match those in Table 2. For example there is no “flux_north” in Table 2 but there is a “piston(Sp)”, which I suppose is “subpolar”, but this is not clarified. Similarly “a” and “b” in Table 1 are defined as 7/14 and 2/14 respectively, but then “alpha” and “beta” in Table 2 are given different values. 5.) I found the scatter plots (Figures 5b and 11b (now figures 4b and 7b)) with pastel coloring and transparent dots with dark outlines to be very difficult to read, largely because the color on the legend is not reflected on the plot where all the points are overlying. I appreciate that there is a lot of data on here, but the transparency really doesn’t help the reader in this respect. These plots also are missing units.

Answer 4) Thank you for highlighting these minor points. They have all been addressed in the revised manuscript, with updated tables, figures and text.

Reviewer #2:

Reviewer #2's major comments are stated and then expanded upon below.

Overarching comment) The conclusion that chemical change is to be the dominant mechanism of future CO2 uptake change in the North Atlantic appears to be a re-presentation of the results from Volker et al. (2002), but without any additional evidence that the mechanism is occurring in ESMs or in nature. The possibility that this may be just the behavior of this box model needs to be addressed carefully by the authors.

Answer to overarching comment) We feel that the work we present here moves significantly beyond Volker et al. (2002). Specifically, by applying the box model to a state-of-the-art ensemble of Earth System Model simulations we have (1) identified two distinct timescales and mechanisms of variability in subpolar N. Atlantic CO_2 uptake, (2) identified 'peak and decline' behaviour in ESMs (previously described in only a highly theoretical context by Volker et al. (2002)), (3) applied the methodology presented in Volker et al. (2002) to an ESM ensemble and by doing so explained the drivers of differences between the Earth System Model ensemble members on both short and long timescales. By doing this, we show (for example) that by using a single set of box model parameters we are able to explain the vast majority of the

ESM's ocean carbon flux variability, including inter-model differences, based on the ESM's alkalinity, salinity, AMOC and temperature variability alone. We then take this further, identifying the relative importance of these factors in driving the ensemble's behaviour. Whilst an important and useful study, Volker et al. (2002) only examines the longer-timescale mechanism, does not (and does not attempt to) identify these changes within Earth System Models (let alone a state-of-the-art perturbed parameter ensemble of such models), does not (and given their approach can not) touch on the uncertainty in the timing of this CO_2 uptake 'peak and decline', and does not explore the role of time-varying alkalinity, temperature or salinity under climate variability and/or climate change (i.e. they only explore changing atmospheric CO_2 and AMOC strength).

The reviewer's specific comments largely relate to one theme, summarised by this quote: 'The possibility that this may be just the behavior of this box model needs to be addressed carefully by the authors'. We thank the reviewer for pushing us on this and respond through the comments and answers below.

The major suggestions for change by reviewer #2 are:

Comment 1) Describe the box model in greater detail.

Answer 1) The reviewer expands upon this with the comment "Pg 14559, Box model equations need to be presented. It is not possible to understand Table 1 or to begin to understand the parameter-setting process otherwise". This is a very useful point. Whilst most of the relationships are presented in figure 3, the relationship between the piston velocity and air-sea flux is not presented. We have therefore added the text 'The gas exchange is calculated by multiplying the piston velocity by the surface area of the box and the difference between the seawater CO_2 concentration and the seawater CO_2 value that would exist at equilibrium with atmospheric CO_2' to page 14559 line 1 (of the original manuscript). We feel that with this addition, the description already in the manuscript, and references to the paper that initially presented the box model, enough detail about the box-model will be given, however please let us know if you feel that more can be given without unnecessarily repeating the information in Volker et al. (2002).

Comment 2) "prov[e] more convincingly that their method of emulation with this model is effective to capture the mechanisms.

Answer 2) This comment is built on in comment 3 (below), so our response to comment 3 extends significantly what is written here in response to comment 2.

Firstly we want to emphasise that our primary reason for considering the box model to be useful is that using only a *single set of parameters*, the box-model is able to explain the vast majority of the model differences across the ESM ensemble (which exhibits diverse carbon flux responses), driven only with change in the ESM's bulk properties. It is hard to imagine how this would occur unless the box model captured the mechanisms at play in the ESM simulations. However, to further demonstrate the skill of the box model we have included results from a new analysis in which we randomly selected half of the ESPPE members, and tuned the box model to best emulate these members, then used that tuning to predict the behaviour of the other half of the ESPPE. These results are presented in figure S4 and discussed in the first paragraph of the Results and Discussion section.

Comment 3) “They need also to more carefully describe and justify their analysis via comparison back to the mechanisms occurring in the ESMs (not just the CO2 fluxes).”

Answer 3) We would initially like to emphasise that the box model was used precisely because it is extremely difficult to robustly identify the operation of detailed mechanisms within complex ESMs. This issue and our solution is appreciated by reviewer #1 "To investigate the causes of this behavior, due to the complex nature of the coupled system and the emergent character of the potential drivers of this CO2 flux variability, Halloran et al use a box model to “emulate” the larger system. Use of simplifying frameworks such as this is becoming increasingly necessary as the ESMs become more intricate, allowing specific processes of interest (in this case temperature, salinity, alkalinity, Atlantic overturning strength and atmospheric CO2) to be considered in isolation". Reviewer #2 however helpfully provides some specific suggestions about what could be done:

Comment 3a) "They might use the latitudinal distribution of CO2 flux as compared to the ESMs, as opposed to just the integrated flux, which can vary widely in space"

Answer 3a) Our reason for using the box model is that it simplifies the system to a level that we can understand and thoroughly interrogate. The box model therefore does not represent the spatial nature of the CO_2 flux, other than that going into the separate surface boxes. The "integrated flux" we use is integrated precisely to allow it to be comparable with the area represented by each box model. No smaller-scale information is available from the box model for comparison. However, in the original manuscript, we only presented data from the box we were trying to explain (the northern box), and as the reviewer implies we can gain further information from interrogating the fluxes simulated across the other two surface boxes. We present this information in the new figure S5, and discuss it in the first paragraph of the Results and Discussion section.

Comment 3b)- "Physical comparisons indicating that the model is reasonable would also be of use"

Answer 3b) The box model only represents physical circulation as prescribed from the Earth System Model simulations, so a physical comparison will unfortunately not tells us anything about the operation of the box model. We apologise if we have misunderstood this point, and are very happy to consider further comparison if suggested.

Comment 3c) "They need also to more carefully describe and justify their analysis via comparison back to the mechanisms occurring in the ESMs (not just the CO2 fluxes)"

Answer 3c) These points relate to a number of more detailed comments made by the reviewer. We present these detailed comments below before responding:

Comment 3ci) *“By using a single box model that replicates the behaviour of a wide range of Earth System Model formulations using only a single set of parameters (i.e. not retuning the simple model to emulate each different version of the more comprehensive model), one can be confident that the box model contains (and therefore that one has identified) the key processes important to the change of interest within those Earth System Model formulations.”* COMMENT: This statement is not adequately justified. It is not clear WHY a single box model emulator leads to confidence that all key processes are captured. If the CO2 flux is an emergent behavior of a complex ESM, how can one be so sure that a box model will capture it? Similarly, points #2 and #3 here need justification. The rest of the analysis hinges on these statements being carefully justified.

Comment 3cii) “*This gives us confidence that the box model represents all of the 1st order processes involved in the ESM simulation of North*

Atlantic CO2 uptake, and provides us with a diagnostic tool to identify what drives CO2 uptake variability in the ESPPE". The reviewer makes the same point about this statement as about the statement above.

Answer 3ci-ii) These points can be summarised by saying that the reviewer is not convinced that the same mechanisms are occurring in the box model as are occurring in the Earth System Model simulations, and consequently that if they are not, we are simply re-presenting the findings from the original box modelling work. Putting aside the fact that this paper does a lot more than look just at the mechanism driving 'peak and decline' behaviour examined theoretically in Volker et al. (2002), as discussed above, we do appreciate where the reviewer is coming from - it would be fantastic to be able to definitively demonstrate that this mechanism was operating in the ESMs by looking just at the ESMs. However, because the surface ocean CO_2 concentration is a response to the movement of DIC within the ocean AND to the air-sea CO_2 flux, and both the air-sea flux and DIC concentration influence each other, we need to think of clever ways to disentangle these. This is precisely why we use the box model. However, we thank the reviewer for pushing us on this, because we think we can do more – see below.

Our prime reason for believing that the mechanisms operating in the box model are the same as those occurring in the Earth System Model is that we have 27 ESM simulations with a wide range of behaviours, but, given the inputs (AMOC strength, T, S, total alkalinity and atmospheric CO_2) from the ESM, and without retuning for different simulations, the box model does a good job at replicating the ESMs air-sea flux. We have now strengthened this with new analysis tuning the box model to half the ensemble and predicting the other half of the ensemble, as discussed above. To further strengthen our argument we present additional 'evidence that this is not just a behavior of this very coarse model', demonstrating that the ESM behaviour is consistent with the box model behaviour:

- We have added a new subsection and two new figures (figures 8 and 9), which provide compelling evidence that the mechanisms we identify through the box model analysis are occurring in the ESM ensemble. **Figure 8 presents** low-pass filtered ESM Subpolar Gyre (SPG) pCO_2 plotted against low-pass filtered ESM SPG pCO_2 calculated offline with DIC, total alkalinity, temperature and salinity sequentially held constant. The box model suggests that the low-frequency behaviour of the box model is primarily driven by changing atmospheric CO_2 (and therefore changing DIC), with secondary controls from alkalinity and temperature and no significant salinity control (figure 4). This is consistent with the behaviour of the ESM. Recalculating pCO_2 whilst holding DIC constant causes a large deviation from the ESM's interactively calculated pCO_2 (i.e. a large deviation from the one-to-one line in figure 8d), recalculating pCO2 with alkalinity or SST held constant results in small deviations from the ESM's interactively calculated pCO_2 (figure 8a and b), and recalculating pCO_2 whilst holding salinity constant results in a very small deviation from the ESM's interactively calculated pCO_2 (figure 8c). Note however that this analysis is simply indicative of what is occurring, because we cannot separate out the different contributors to DIC change.

Figure 9 presents high-pass filtered ESM Subpolar Gyre (SPG) pCO_2 plotted against high-pass filtered SPG pCO_2 calculated from ESM variables but with DIC, total alkalinity, temperature and salinity sequentially held constant. Our box model analysis implies that the high-frequency behaviour of the box model is primarily driven by changing SST and alkalinity (figure 11 (now figure 7)). This is consistent with the behaviour seen in the ESM (figure 9). The relationship with pCO_2 calculated interactively in the ESM, and that calculated offline using EMS temperature, salinity, DIC and alkalinity still holds if salinity is held constant (figure 9c), is less strong if DIC is held constant (figure 9d), and is weak where alkalinity or SST are held constant (figure 9a and b). These results are presented in a new subsection ‘Evidence for these mechanisms occurring in the Earth System Model ensemble’.

Comment 4) *“The “peak and decline” behaviour seen in the low-frequency air–sea CO2 flux signal is unlike the globally averaged signal (Fig. 2), which under a CO2 emission scenario like RCP8.5 (in which atmospheric concentrations are increasing throughout the 21st Century) would be expected to (and indeed does: Fig. 2) continue increasing, but at a progressively reduced rate. As long as the atmospheric CO2 concentration is increasing, assuming no dramatic changes in ocean circulation or biology, there will always be an air to sea CO2 concentration gradient, and therefore air-to-sea CO2 flux. The decrease in this flux through time reflects the changing speciation of carbon in seawater in response to the increase in carbonic acid concentrations – which partitions carbon progressively in the direction of CO2, elevating surface ocean CO2 concentrations, and reducing the air–sea CO2 concentration gradient (Zeebe and Wolf- Gladrow, 2001; Revelle and Suess, 1957).”* COMMENT: Is there evidence that this is happening in the ESMs? Otherwise, this is simply re-presenting the work of Volker et al. (2002) from the same box model. Is there any evidence that this is not just a behavior of this very coarse model?

Answer 4) Here we are simply describing the well-understood chemical buffering of anthropogenic CO_2 as quantified by the Revelle Factor. Our description presents neither the findings of our work or those of Volker et al. (2002). Regarding the comment ‘Is there evidence that this is happening in the ESMs’, we have not presented any such evidence because this is the bread and butter of an Earth System Model. To help clarify that this is what we are discussing, we have modified the paragraph to include the text ‘The globally averaged response is consistent with our basic understanding of seawater carbon chemistry (e.g. Zeebe and Wolf-Gladrow, 2001; Revelle and Suess, 1957), and results from other ESMs (e.g. Friedlingstein et al. 2006).’

Reviewer #2's minor comments:

Minor Comment 1) This box model is very simple. For example, the tropics extend 30S-48N. The authors should not be discussing “subtropical processes” in results. They should use the term “Tropical” to be consistent with their model setup and to clarify the very simple nature of their box model system to the reader.

Answer 1) We apologise for this. As the reviewer highlights, we have described the box model’s 30S-48N box at the subtropical box, which is clearly misleading. Given the latitudinal range of this box, we feel that ‘low latitude’ would be most appropriate term, and have used this throughout the text.

Minor comment 2) Pg 14554, line 1 “Here we attempt to develop our understanding of the possible mechanisms controlling future subpolar North Atlantic CO2 uptake within Earth System Models.” This sentence is overly caveated, remove “attempt to” and “possible”.

Answer 2) Thank you, we have made this change.

Minor comment 3) Pg 14557, line 7 “ESPPE” Acronym has not been defined yet .

Answer 3) Thank you, this has been changed.

Minor comment 4) Methods section Pg 14559 Why don’t the parameters listed in Table 1 correspond to the parameter values listed in Table 2?

Answer 4) We are sorry about this mistake, we have modified the names so that they match between the tables.

Minor comment 5) Pg 14559 “Indeed the ability of the box model is relatively insensitive to the box model parameters (Fig. 4 and Table 1), suggesting that conclusions drawn on the drivers of the box model CO2 flux are unlikely to be strongly dependent on the exact choice of box model parameters.” It is not clear how this conclusion is to be reached when given Table 1 that lists parameter names and whether they vary or not; and Figure 4 which does not offer any indication of the parameter values in the box model for each timeseries of flux. If the authors mean Table 2, they need to indicate how “Ranking” relates to the panels in Figure 4 more clearly. Are there only 6 in this ranking, or is it 1000 as indicated in the text? If 1000, Table 2 and Figure 4 do actually not correspond.

Answer 5) Thank you for pointing this out, we should have referred to Table 2 not Table 1, we have changed this, explained in the caption of figure 4 (now figure S1) how the displayed time series relate to the parameter sets presented in Table 2.

Minor comments 6) Pg 14560, line 18 and on – This methodological discussion belongs in Methods. Pg 14560, line 21 “we high-pass” FILTER
Pg 14560. Line 24 “in a time-series THAT varIES”

Answer 6) Thank you for highlighting these points. These changes have been made.

References (where not included in the manuscript):

Wei Cheng, John C. H. Chiang, and Dongxiao Zhang, 2013: Atlantic Meridional Overturning Circulation (AMOC) in CMIP5 Models: RCP and Historical Simulations. *J. Climate*, **26**, 7187–7197

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The mechanisms of North Atlantic CO_2 uptake in a large Earth System Model ensemble

P. R. Halloran[1], B.B.B. Booth[2], C.D. Jones[2], F.H. Lambert[3], D.J. McNeall[2], I.J. Totterdell[2], and C. Völker[4]

[1]Geography, College of Life and Environmental Sciences, University of Exeter, Amory Building, Rennes Drive, Exeter, EX4 4RJ, United Kingdom
[2]Met Office Hadley Centre, FitzRoy Road, Exeter, Devon, EX1 3PB, United Kingdom
[3]Exeter Climate Systems, College of Engineering, Mathematics and Physical Sciences, University of Exeter, Harrison Building, North Park Road, Exeter, EX4 4QF, United Kingdom
[3]Alfred Wegener Institute Helmholz Center for Polar and Marine Research, Am Handelshafen 12, D-27570 Bremerhaven, Germany

Correspondence to: P.R. Halloran
(p.halloran@exeter.ac.uk)

Abstract

The oceans currently take up around a quarter of the carbon dioxide (CO_2) emitted by human activity. While stored in the ocean, this CO_2 is not influencing Earth's radiation budget; the ocean CO_2 sink therefore plays an important role in mitigating global warming. CO_2 uptake by the oceans is heterogeneous, with the subpolar North Atlantic being the strongest CO_2 sink region. Observations over the last two decades have indicated that CO_2 uptake by the subpolar North Atlantic sink can vary rapidly. Given the importance of this sink and its apparent variability, it is critical that we understand the mechanisms behind its operation. Here we explore the combined natural and anthropogenic subpolar North Atlantic CO_2 uptake across a large ensemble of Earth System Model simulations, and find that models show a peak in sink strength around the middle of the century after which CO_2 uptake begins to decline. We identify different drivers of change on interannual and multidecadal timescales. Short-term variability appears to be driven by fluctuations in regional seawater temperature and alkalinity, whereas the longer-term evolution throughout the coming century is largely occurring through a counterintuitive response to rising atmospheric CO_2 concentrations. At high atmospheric CO_2 concentrations the contrasting Revelle factors between the low latitude water and the subpolar gyre, combined with the transport of surface waters from the low latitudes to the subpolar gyre, means that the subpolar CO_2 uptake capacity is largely satisfied from its southern boundary rather than through air-sea CO_2 flux. Our findings indicate that: (i) we can explain the mechanisms of subpolar North Atlantic CO_2 uptake variability across a broad range of Earth System Models; (ii) a focus on understanding the mechanisms behind contemporary variability may not directly tell us about how the sink will change in the future; (iii) to identify long-term change in the North Atlantic CO_2 sink we should focus observational resources on monitoring lower latitude as well as the subpolar seawater CO_2; (iv) recent observations of a weakening subpolar North Atlantic CO_2 sink may suggest that the sink strength has peaked and is in long-term decline.

1 Introduction

Our limited understanding of how the CO_2 emission to atmospheric CO_2 (CO_2^{atm}) concentration ratio will evolve through time constitutes one of the largest components of uncertainty in future climate projections (Booth et al., 2012). To constrain how this airborne fraction of CO_2 might change, and thereby link physical climate understanding to the development of CO_2 emission policy, we need to understand the behaviour of the major terrestrial and marine CO_2 sources and sinks (Friedlingstein et al., 2006).

Earth System Models (ESMs) are the most advanced tools we have available to calculate the link between CO_2 emissions and CO_2 concentrations. At a globally-averaged scale, the current generation of Earth System Models, those developed and run for CMIP5 (Taylor et al., 2012), the 5th Climate Model Intercomparison Project, show good agreement on 21^{st} Century global ocean CO_2 uptake. With the exception of INM-CM4.0 (Volodin et al., 2010) the CMIP5 inter-model globally averaged ocean CO_2 uptake differences are smaller than the inter-scenario differences (Jones et al., 2013). At a regional level however, models do not agree. Furthermore, regional CO_2 uptake can behave very differently from that of the global mean (figure **??**).

We need to understand the mechanisms behind differences in regional uptake to help us (i) validate models, and (ii) identify where and how to focus observations.

Whilst the carbon-cycle community is developing an increasingly comprehensive understanding of the mechanisms behind recent ocean CO_2 uptake variability in the North Atlantic (e.g. McKinley et al., 2004; Thomas et al., 2008; Ullman et al., 2009; Metzl et al., 2010; McKinley et al., 2011; Pèrez et al., 2013; Schuster and Watson, 2007), the Southern Ocean (e.g. Lenton and Matear, 2007; Le Quèrè et al., 2007; Lovenduski et al., 2013; Sallee et al., 2012; Ito et al., 2010; Lenton et al., 2009; Verdy et al., 2007), and potential broad-scale future ocean CO_2 uptake changes (e.g. Marinov et al., 2008; Murnane et al., 1999; Roy et al., 2011; Sarmiento and Le Quèrè, 1996), our understanding of the specific future mechanisms of change projected within comprehensive ESMs in these regions is much more limited (Sèfèrian et al., 2012; Russell et al., 2006; Halloran, 2012). Here we ~~attempt to~~ develop our understanding of the ~~possible~~

mechanisms controlling future subpolar North Atlantic CO_2 uptake within Earth System Models.

To understand why the North Atlantic CO_2 sink may be vulnerable to change, it is useful to review the factors that make the region such an intense CO_2 sink (figure 2) (McKinley et al., 2011; Watson et al., 2009; Schuster et al., 2013). Present-day high CO_2 uptake in the subpolar North Atlantic occurs because water that moves northwards as part of the Atlantic Meridional Overturning Circulation (AMOC) experiences steep thermal and chemical gradients and high biological activity (Rayner et al., 2003; Key et al., 2004; Carr et al., 2006). Biological activity exports carbon to depth in the form of sinking biological material, reducing surface carbon concentrations and increasing the air-sea CO_2 gradient. The cooling of water increases the solubility of CO_2 and speciates carbon into forms other than CO_2 (e.g. Zeebe and Wolf-Gladrow, 2001), further increasing the air-sea CO_2 gradient. Deep convection then removes water from contact with the atmosphere, potentially before it has had time to come into air-sea CO_2 equilibrium, maintaining a continuous strong air-sea CO_2 gradient - and therefore flux (Takahashi et al., 2009). A further complicating factor in the North Atlantic is that limited mixing between the subtropical and subpolar gyres allows the development of a strong biogeochemical gradient between waters with a high alkalinity to dissolved-carbon ratio (the warm and saline low-latitude waters), and waters with a low alkalinity to dissolved-carbon ratio (the cool and relatively fresh high-latitude waters) (Key et al., 2004). This biogeochemical gradient results in a high CO_2 buffering capacity of ~~subtropical~~ low latitude water, permitting high anthropogenic CO_2 uptake, and a low buffering capacity at higher latitudes, limiting local future CO_2 uptake (Sabine et al., 2004). Combined with the advection of water from the subtropical to subpolar gyre, this latitudinal buffering gradient will likely impact the response of the sink to rising CO_2^{atm} (Völker et al., 2002).

Presently there is no agreement on the relative importance of the different factors described above in controlling past or future subpolar North Atlantic CO_2 uptake change. The hypothesised mechanisms for past decadal to multidecadal timescale changes in subpolar North Atlantic CO_2 uptake fall into four groups:

1. **Biological drawdown**. Evidence that CO_2 uptake variability may arise from the biological transport of carbon out of the surface ocean comes from the relative timing of observed surface ocean pCO_2 and chlorophyll change (Lefevre et al., 2004). The magnitude of this effect has however been questioned (Bennington et al., 2009).

2. **Temperature**. Both observational and model studies indicate that the temperature dependence of inorganic carbon speciation and CO_2 saturation is likely to have been an important player in air-sea CO_2 flux change on various timescales (Le Quèrè et al., 2000; Lefevre et al., 2004; McKinley et al., 2011; Omar and Olsen, 2006; Pèrez et al., 2013).

3. **Vertical mixing**. Changes in vertical mixing (through deep convection or stratification) has been proposed from both models and observations to be a dominant mechanism for changing the surface total ~~DIC~~ Dissolved Inorganic Carbon (DIC) concentration and DIC-alkalinity ratio, and therefore changing the surface pCO_2 saturation (McKinley et al., 2004; Metzl et al., 2010; Schuster and Watson, 2007; Ullman et al., 2009), although this effect is likely to be damped by the associated changing vertical flux of nutrients and therefore biological CO_2 drawdown (McKinley et al., 2004).

4. **Horizontal advection**. Changes in surface ocean pCO_2 saturation driven by horizontal advection (rather than vertical transport) have been proposed from both modelling and observational studies (Omar and Olsen, 2006; Thomas et al., 2008). Debate however exists about the degree of long term DIC and alkalinity change, which brings in to question mechanisms implicating vertical and/or horizontal DIC and/or alkalinity transport (Corbi‘ere et al., 2007).

The diversity of proposed explanations for the observed subpolar North Atlantic CO_2 uptake variability could reflect different mechanisms dominating at different times and influencing uptake over different timescales. Many of the studies to-date have however examined approximately the same time-periods. The range of proposed mechanisms therefore more-likely reflects the difficulty of identifying causal drivers of change in a system, which despite huge effort, is still far from completely observed. Similar problems apply to model-based studies.

Proving causality in a model is straight forward when considering drivers external to the system (e.g. rising anthropogenic CO_2 emissions), because those drivers can be switched on and off, but when potentially important components of the mechanism are emergent properties of the model (e.g. the Meridional Overturning Circulation (MOC)), these components can not simply be switched on and off, and even where they can be stopped (e.g. in the case of the AMOC by flooding the high-latitude North Atlantic/Arctic with freshwater), their role in the mechanism can not be isolated, because many other factors will change. To understand the mechanisms operating within ESMs, it can therefore often be useful to produce an even simpler model of the system (e.g. Good et al., 2011; Hooss et al., 2001; Meinshausen et al., 2011), one that emulates the complex model's behaviour, but also allows one to separately isolate the different components of the mechanisms. This is particularly valuable when attempting to understand common (or divergent) behaviours across a large suite of models.

Here we explore the mechanisms controlling ocean CO_2 uptake across a large ensemble of HadCM3 (3^{rd} Hadley Centre Climate Model) based ESMs in which parameters have been systematically varied to efficiently sample a wide range of model behaviours (Lambert et al., 2013). We refer to this ensemble as the Earth System Perturbed Parameter Ensemble, or ESPPE. We make use of the Atlantic carbon-cycle box model presented by Völker et al., (2002) to emulate the more complex ESM and simplify this large suite of simulations. The value of simplifying our large suite of ESM simulations in this way is that:

1. By using a single box model that replicates the behaviour of a wide range of Earth System Model formulations using only a single set of parameters (i.e. not retuning the simple model to emulate each different version of the more comprehensive model), one can be confident that the box model contains (and therefore that one has identified) the key processes important to the change of interest within those Earth System Model formulations. ~~I.e. by fitting a small number of parameters within a single box model to a large number of ESM results, one in effect has a highly-constrained set of simultaneous equations describing the system. Almost all of the ESPPE uncertainty is therefore contained within the inputs to the box model rather than the parameters within the box model. The different~~

~~processes of North Atlantic Subpolar CO_2 uptake simulated by ESPPE ensemble members are therefore captured within these box-model inputs.~~

2. Within a box model one can isolate and quantify the importance of each of these drivers of change by ~~sequentially~~ separately holding the inputs representing that driver constant and re-running the ensemble, or filtering input data to remove and isolate the component of variability of interest. As discussed, this cannot be done in an Earth System Model where properties like overturning circulation emerge from the physics and are therefore impossible to prescribe.

3. Using a box model shown to replicate (without retuning) the behaviour of multiple Earth System Model formulations, one can undertake numerous idealised simulations~~(e.g. $CO_2^{atm.}$ increasing following various power-law curves)~~, and by doing so develop a thorough understanding of the mechanisms at play. To do this with a full ESM would be extremely time consuming and expensive.

2 Methods

We attempt to isolate the mechanisms controlling North Atlantic CO_2 uptake in a 27 member ESM ensemble based on a carbon cycle version of the 3rd Hadley centre Climate Model HadCM3C (an updated version of Cox et al. (2000), with increased horizontal resolution and improved aerosol representation (Lambert et al., 2013), and using the Hadley centre Ocean Carbon Cycle (HadOCC) sub-model (Palmer and Totterdell, 2001)), in which the atmosphere and ocean physics, the atmospheric sulphur cycle and terrestrial biogeochemistry parameters have been systematically varied to optimally sample parameter space (Lambert et al., 2013). The HadCM3C perturbed parameter ensemble is referred to herein as ESPPE (Earth System Perturbed Parameter Ensemble). The original ESPPE ensemble contains 57 members, but data corruption meant that only 27 of these members could be used in the analysis presented here. The ESPPE ensemble follows the CMIP5 RCP8.5 pathway (Riahi et al., 2007), and has a fully interactive carbon cycle: CO_2 emissions are prescribed, and atmospheric CO_2 concentrations calculated.

The box model we use to simplify the behaviour of the ESPPE represents the major features of the Atlantic basin and Atlantic sector of the Southern Ocean, and is made up of 6 boxes, three surface and three deep. The surface boxes represent the top 300m of the ocean south of 30S, the top 150m of the tropical ocean between 30S and 48N, and the upper 300m of the subpolar region north of 48N (~~Figure~~ figure 3). The three subsurface boxes represent the deep high-latitude ocean north of 48N, the intermediate depth ocean between 150 and 1000m in the tropical region (30S-48N), and the remaining deep Atlantic ocean. The volume fluxes between the 6 boxes, and the temperature, salinity and alkalinity of those boxes are prescribed, as is the atmospheric CO_2 concentration. The position and volume of the boxes, the mixing between the boxes, and the way advection is divided between boxes is based on observations and remains unchanged from that described in Völker et al. (2002). The model advects dissolved inorganic carbon (DIC) between boxes in quantities proportional to the prescribed overturning circulation strength, and mixes DIC between vertically adjacent boxes, as described in Völker et al. (2002). The box model does not include any representation of biological carbon fluxes, which were (and are commonly) considered to be of limited importance to anthropogenic carbon uptake (e.g. Völker et al., 2002; Pèrez et al., 2013). In each of the three surface boxes, the CO_2 concentration is calculated from the DIC, temperature, salinity and alkalinity. Any disequilibrium between partial pressures of CO_2 in the ocean and atmosphere then drives a flux which is rate limited by a prescribed piston velocity. The ~~calculated~~ gas exchange is calculated by multiplying the piston velocity by the surface area of the box and the difference between the seawater CO_2 concentration and the seawater CO_2 value that would exist at equilibrium with atmospheric CO_2. The calculated air-sea CO_2 flux then modifies the concentration of DIC in each box. The formulation of the box model remains exactly as described in Völker et al. (2002) other than the tuning of the box model's parameters (table 1) to allow the box model to replicate results from the perturbed parameter ensemble. Note that by prescribing changes in alkalinity and allowing the DIC to adjust through air-sea flux, we are implicitly assuming that there is no significant freshwater-driven dilution/concentration of DIC and alkalinity.

To allow the box model to emulate the ESPPE, a single set of box model parameters was obtained by first running a 1000 member box model ensemble in which each of the box model parameters were varied within the ranges listed in table 1. Parameter space was sampled using a latin hypercube. The fitness of each of the 1000 parameter sets was then judged by calculating the average coefficient of determination (R^2) across the 27 ESPPE members between the ESPPE subpolar ~~gyre~~ North Atlantic air-sea flux, and the box model's northern box air-sea flux. The ability of the box model to reproduce the ESM carbon flux is more dependent on the driving time-series ($CO_2^{atm.}$, temperature, salinity, alkalinity and overturning circulation strength) than it is dependent on the exact box model parameters. Indeed the ability of the box model is relatively insensitive to the box model parameters (~~Figure ?? and Table 1~~table 2 and figure S1) suggesting that conclusions drawn on the drivers of the box model ~~CO2~~ CO_2 flux are unlikely to be strongly dependent on the exact choice box model parameters. The ~~three~~ six parameter sets that gave the highest R^2 when compared with ESPPE output are presented in table 2.

Variability on different timescales is separated using high and low-pass filtering. Filtering is achieved by applying a 5th order Butterworth fast Fourier transform filter. The mechanisms driving the modes of variability isolated using the high and low-pass filters are identified by manipulating the input time-series (temperature, salinity, alkalinity, atm. CO_2 and AMOC strength) used to force the box-model. These input time-series are either filtered, held at a constant value, or left unchanged when supplied to the box-model. Initially only one input time-series is is manipulated at a time. In subsequent analysis, multiple input time-series have been manipulated to examine their additive effect on the air-sea CO_2 flux.

3 ~~Results and Discussion~~

~~Using only a single set of parameters, the box model captures much of the variability in subpolar North Atlantic air-sea CO_2 flux simulated within and across the diverse ESPPE members (see examples in figures ?? and full dataset in 4a). This gives us confidence that the box model represents all of the 1^{st} order processes involved in the ESM simulation of North Atlantic CO_2~~

~~uptake, and provides us with a diagnostic tool to identify what drives CO_2 uptake variability in the ESPPE.~~

~~To explore the mechanisms behind the ESM's variability we initially broke-down the subpolar North Atlantic air-sea flux behaviour simulated within the Earth System Model ensemble by applying high and low pass filters to the data (figure 5). This allows us to identify discreet time-scales of variability common across all ensemble members. We find that filtering the ESM results at <5 years and >30 years allows us to capture almost all of the ESM's variability whilst cleanly separating the variability in to two components (figure 5). We will explore the mechanisms behind these two timescales of variability independently.~~

To pick apart the contribution of different processes to the high and low frequency air-sea CO_2 flux simulated by the ESPPE, we sequentially control the inputs to the box model, isolating the role of that input in producing the overall change. Firstly, to understand the mechanism behind the high-frequency variability, we high-pass filter all of the inputs to the box model (temperature, salinity, alkalinity, atmospheric CO_2 concentrations and overturning circulation strength), adding to this the mean value from the original time-series (since the high-pass filtering results in a time-series varying around zero). This process removes any low-frequency variability. The high-pass filtered time-series are used to drive the box model, and results compared to high-pass filtered results from the ESPPE (figure ~~6~~S2). The input variables for the North Atlantic are then sequentially held at their mean value (i.e. removing any variability) and the box model re-run (figure ~~6). To~~ S2). Secondly, to understand the mechanisms driving the low-frequency variability the box model input time-series are sequentially low-pass filtered (all other time-series remain unchanged) and the box model run (figure ~~6)~~S3), as described for the high-pass filter analysis.

3 Results and discussion

3.1 Box model validation

Using only a single set of parameters, the box model captures much of the variability in subpolar North Atlantic air-sea CO_2 flux simulated within and across the diverse ESPPE members (see the full dataset in figure 4a and time-series examples from that dataset, figure S1). To test the predictive skill of the box model as an emulator for the ESPPE, we tuned the box model to emulate 13 randomly selected ESPPE members, as described in the methods section, then ran the box model with inputs from the remaining ESPPE members, i.e. those ensemble members excluded from the tuning ensemble. Comparison of predicted and actual ESPPE subpolar North Atlantic air-sea CO_2 flux yields a coefficient of determination of 0.66 (figure S4). Comparison of the box model's low latitude and southern box air-sea CO_2 flux with the ESPPE air-sea CO_2 flux shows that much of the variability outside of the subpolar region is also explained by the box model. This result holds independent of whether the box model is tuned to replicate the northern, low-latitude or southern box air-sea CO_2 flux (figure S5). The validation presented here gives us confident that the box model represents the 1^{st} order processes involved in the ESM simulation of North Atlantic CO_2 uptake, and provides us with a diagnostic tool to identify what drives CO_2 uptake variability in the ESPPE. Our findings imply that almost all of the ESPPE uncertainty is contained within the inputs to the box model rather than the parameters within the box model. The different processes of North Atlantic Subpolar CO_2 uptake simulated by ESPPE ensemble members are therefore also captured within these box-model inputs.

3.2 Modes of variability

To explore the mechanisms behind the ESM's variability we initially broke-down the subpolar North Atlantic air-sea flux behaviour simulated within the Earth System Model ensemble by applying high and low pass filters to the data (figure 5). This allows us to identify discreet time-scales of variability common across all ensemble members. We find that filtering the ESM results at < 5 years and > 30 years allows us to capture almost all of the ESM's variability whilst cleanly separating the variability in to two components (figure 5). We will explore the mechanisms behind these two timescales of variability independently.

Splitting the ESPPE North Atlantic subpolar air-sea CO_2 flux into a high and low frequency component a number of things become clear. Firstly, the majority of the total signal can be described by these two separate components (figure 5). Secondly, we see that the high frequency component occurs with little coherent structure across all ensemble members, but it does show an increase in variability towards 2100 (figure 5). Thirdly, we see that the ~~low-period~~ low-frequency signal tends to increase from its pre-industrial value through the 20^{th} Century, then in most cases peaks during the 21^{st} Century, then begins to decline (figure 5).

The ‘peak and decline’ behaviour seen in the low-frequency air-sea CO_2 flux signal is unlike the globally averaged signal (figure **??**), which under a CO_2 emission scenario like RCP8.5 (in which atmospheric concentrations are increasing throughout the 21^{st} Century) would be expected to (and indeed does - figure **??**) continue increasing, but at a progressively reduced rate. The globally averaged response is consistent with our basic understanding of seawater carbon chemistry (Zeebe and Wolf-Gladrow, 2001; Revelle and Suess, 1957), and results from other ESMs (e.g. Friedlingstein et al., 2006). As long as the atmospheric CO_2 concentration is increasing, assuming no dramatic changes in ocean circulation or biology, there will always be an air to sea CO_2 concentration gradient, and therefore air-to-sea CO_2 flux. The decrease in this flux through time reflects the changing speciation of carbon in seawater in response to the increase in carbonic acid concentrations - which partitions carbon progressively in the direction of CO_2, elevating surface ocean CO_2 concentrations, and reducing the air-sea CO_2 concentration gradient (Zeebe and Wolf-Gladrow, 2001; Revelle and Suess, 1957).

The difference in behaviour between the subpolar North Atlantic and the well understood chemical response of the steady-state ocean (Revelle and Suess, 1957) (as largely seen here in the global average: figure **??**) indicates that CO_2 emission (and potentially associated climate change) forced physical, biological or chemical changes in the North Atlantic are modifying the capacity of this sink to take up atmospheric CO_2. ’Peak and decline’ North Atlantic CO_2 uptake has previously been identified in an idealised study by Völker et al. (2002) (using the box-model applied in this study)~~, who demonstrated~~. Völker et al. (2002) demonstrate theoretically that the ~~region~~ high latitude North Atlantic could take up less atmospheric CO_2 in the future than it did in the preindustrial, without invoking any change in ocean circulation or biology. The ’peak and decline’ demonstrated by Völker et al. (2002) occurred in response to proportionally more CO_2 being taken up under higher atmospheric CO_2 conditions in the ~~subtropical than~~ low latitude

Atlantic than in the subpolar North Atlantic - in response to the higher alkalinity (and therefore lower Revelle Factor (Revelle and Suess, 1957) and higher buffering of surface ocean pCO_2) in the ~~subtropical~~ low latitude waters, and that excess carbon being transported north into the subpolar gyre by the overturning circulation (explained further in figure 6 and the associated caption).

3.3 Drivers of multidecadal/~~centenial behaviour~~centennial mode of variability

To assess the drivers of multidecadal/centenial variability, we first plot each annual-average value from the ESM simulations against the equivalent value generated ~~using~~ within the box model (figure 4a). We then sequentially apply a low-pass filter to each input variable (and sets of input variables) to remove the low-frequency ($>$ 30 year) variability from that/those input variable/variables, and using those input values run the box model. We then examine how the removal of low-frequency variability from the different input variables changes the output of the box model (figure 4b).

We find that the most important driver of the low-frequency ('peak and decline') variability in the subpolar North Atlantic air-sea CO_2 flux comes from the progressive increase in atmospheric CO_2 concentrations (figure 4), which drives much of both the increase and decrease (figure ~~6~~S3) in CO_2 flux, as described under idealised conditions by Völker et al. (2002). ~~It is clear however that without~~ Without a low-frequency signal in the atmospheric CO_2 concentrations fed into the box model however, a 21^{st} Century decline in air-sea CO_2 flux is still present (figure S5). This decline is driven by a slow reduction in subpolar alkalinity and to a lesser degree warming ~~typically occurring in the ESPPE simulations~~ (figures 4 and ~~7~~S6). This finding confirms the applicability to our ESM ensemble of the idea proposed by Völker et al. (2002), and described in the proceeding paragraph~~, to our ESM ensemble (figure 4)~~.

The similarity between the box model behaviour with no low-pass filtered inputs (i.e. optimally emulating the ESPPE), and with input salinity and AMOC low-pass filtered (figure 4), tells us that these two factors are not having an important impact on the low-frequency subpolar North Atlantic 'peak and decline' air-sea flux time evolution (figure 4). The minimal impact of AMOC change on subpolar North Atlantic air-sea CO_2 flux likely reflects the facts that the

AMOC decline across the ESPPE is relatively modest (figure S6), and that only a fraction of the water moved by the AMOC has an opportunity to exchange CO$_2$ with the atmosphere. Removing the low-frequency signal from the temperature time-series used by the box model has a minor effect (figure 4), causing the box model to over-predict the air-sea CO$_2$ flux at times of high flux, which translates in time-series analysis to slightly underestimating the decline ~~often~~ after peak air-sea CO$_2$ flux has been reached (figure ~~6~~S3). Similarly removing the low-frequency signal from the alkalinity time-series input to the box model causes a slightly greater over-prediction of air-sea CO$_2$ flux values during the decline phase (figures 4 and ~~6~~S5).

3.4 Drivers of annual/~~interannual behaviour~~inter-annual mode of variability

~~Moving now to~~ Considering the high-frequency variability simulated within the ESPPE (figure ~~6~~**??** and figure S2), we compare box model simulations run with all input time-series high-pass filtered, with high-pass filtered ESPPE subpolar North Atlantic air-sea CO$_2$ flux data. We then sequentially (and then together) hold the input time-series constant at their average values (figure **??**), and re-run the box model to isolate the contribution of variability in each of the input time-series to ~~ESPPE result~~the ESPPE results. We find that the box model captures the temporal variability but tends to underestimate the magnitude of variability (figure **??**a). Holding temperature and alkalinity (yellow dots) constant we find near-complete breakdown of the box model's ability to capture the ESM's CO$_2$ flux variability (figure **??**b). Independently holding temperature and alkalinity constant we find that these factors separately account for much of the correlation between the box model and ESPPE high-frequency variability. Holding salinity, meridional overturning circulation strength and atmospheric CO$_2$ concentrations constant (in turn) we find little impact on the correlation between the box model and the ESSPE results (figure **??**b). It is therefore clear that the high-frequency variability simulated by the ESM within the ESPPE is almost completely driven by variability in temperature and alkalinity, and is largely insensitive to the model's variability in salinity, AMOC and atmospheric CO$_2$ on these timescales.

3.5 Evidence for these mechanisms occurring in the Earth System Model ensemble

By emulating the ESPPE using the box model we have simplified the system to a level at which we can explore the mechanisms at play in detail. Using the box model we have identified what appears to be the dominant mechanisms controlling high-latitude Atlantic CO_2 uptake on short ($<$5yr) and long ($>$30yr) timescales. We finally ask whether these mechanisms are consistent with evidence derived purely from the Earth System Model simulations.

Earth System Model pCO_2 is calculated interactively from DIC and alkalinity concentrations, temperature and salinity. It is possible to repeat this calculation offline (e.g. Halloran, 2012), and by doing so assess the relative importance of these different variables in determining the model's pCO_2, and by inference air-sea CO_2 flux, on different timescales. We perform this analysis on the low and high-pass filtered time series to ask whether the same variables are controlling air-sea CO_2 flux in the ESM ensemble members as have been identified in the box model analysis.

Our box model analysis suggests that the low-frequency behaviour of the box model is primarily driven by changing atmospheric CO_2 (and therefore changing DIC), with secondary controls from alkalinity and temperature and no significant salinity control (figure 4). We find this to be consistent with the behaviour of the ESM ensemble members. Recalculating pCO_2 whilst holding DIC constant causes a large deviation from the ESM ensemble's interactively calculated pCO_2 (i.e. a large deviation from the one-to-one line in figure 8d), recalculating pCO_2 with alkalinity or SST held constant results in small deviations from the ESM's interactively calculated pCO_2 (figure 8a and b), and recalculating pCO_2 whilst holding salinity constant results in a very small deviation from the ESM's interactively calculated pCO_2 (figure 8c). Note however that this analysis is simply indicative of what is occurring, because we cannot separate out the different contributors to DIC change.

Our box model analysis indicates that the high-frequency behaviour of the box model is primarily driven by changing SST and alkalinity (figure **??**). We find this to be consistent with the behaviour seen in the ESM ensemble. The relationship with pCO_2 calculated interactively in the ESM, and that calculated offline using EMS temperature, salinity, DIC and alkalinity still holds if salinity is held constant (figure 9c), is less strong if DIC is held constant (figure 9d), and is weak where alkalinity or SST are held constant (figure 9a or b).

4 Conclusions

~~We find very different~~ We find that different mechanisms are controlling the interannual and centennial subpolar North Atlantic CO_2 variability in our large ensemble of perturbed parameter ESM simulations. The interannual variability ~~is~~ appears to be controlled by rapid changes in the local seawater temperature and alkalinity fields, whereas the centennial variability is largely controlled by the ~~time-evolution of~~ anthropogenically driven increase in atmospheric CO_2 concentrations interacting with the background chemical gradient (high to low alkalinity)~~and carbon transportmoving northwards up the Atlantic, with secondary and tertiary effects from alkalinity and temperature change respectively. Here we see both how increasing atmospheric CO_2 concentrations can have an unintuitive and complex impact on the subpolar North Atlanticsink, and how alkalinity can modify this behaviour.~~

, and DIC transport, in the North Atlantic. Our findings suggest that while it is important to understand the mechanisms behind recent interannual variability in the subpolar North Atlantic CO_2 flux, that understanding might not directly inform us about how the sink is likely to change in the future. ~~The fact that the future strength of the~~

CO_2 uptake change can be driven by the basic chemical response of seawater to rising atmospheric CO_2, change in the ocean's physical circulation or state, or change in biological activity. We have greatest confidence in predicting future change based on the former and least confidence in change based on the latter. This is because the chemistry is well understood and largely independent of the climate system response, whilst the physical change is subject to uncertainty in the climate system dynamics and the biological change adds structural and parameter uncertainty to the already uncertainly physical response. The fact that the 21^{st} century subpolar North Atlantic CO_2 ~~sink~~ uptake change appears to be largely controlled by the basic chemical response of seawater to rising atmospheric CO_2 concentrations ~~(rather than physical or biological components of the model - in which we would have less confidence) gives us~~

~~reason to consider it likely that this behaviour could be shared by~~ therefore implies that similar behaviour can be expected in the real-world~~future ocean~~. This raises the question, if the real-world North Atlantic CO_2 sink is to follow ~~this~~ a peak and decline trajectory, where on this trajectory do we presently sit? Perhaps the the suggestion that the strength of the subpolar North Atlantic CO_2 sink has been decreasing (e.g. McKinley et al., 2011; Schuster and Watson, 2007) indicates that the real-world system is already in long-term decline.

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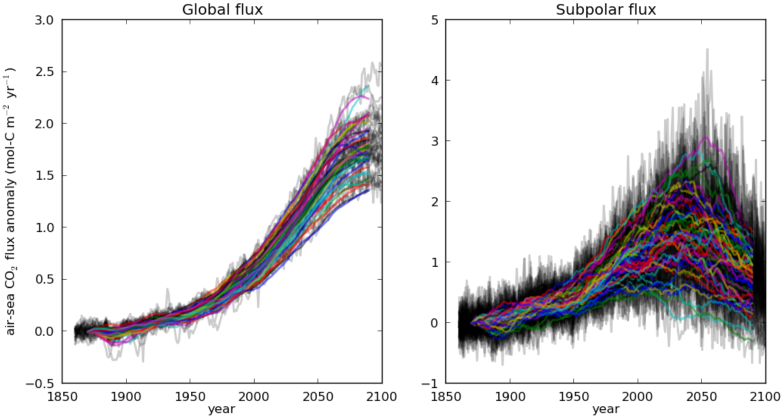


Fig. 1. **Left:** globally averaged air-sea CO$_2$ flux, and **right:** North Atlantic subpolar region averaged air-sea CO$_2$ flux. Black lines represent annually-averaged time series from all ESPPE members, and coloured lines represent those time series after application of a 20 year running mean.

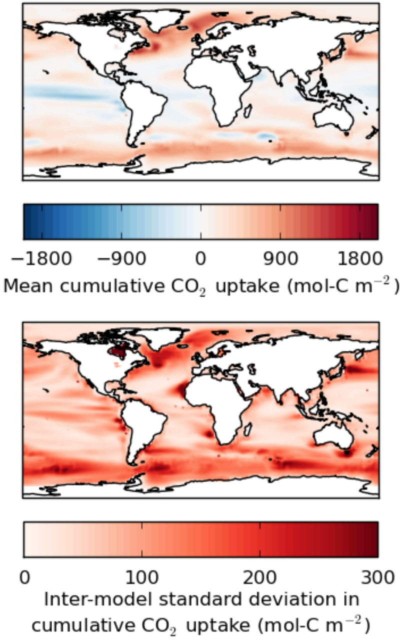


Fig. 2. Cumulative sum of air-sea CO_2 flux between the years 1860 and 2100 (RCP8.5). **a** Mean and **b** inter-model standard deviation across ESPPE.

~~Globally averaged air-sea CO_2 flux across all ESPPE members.~~

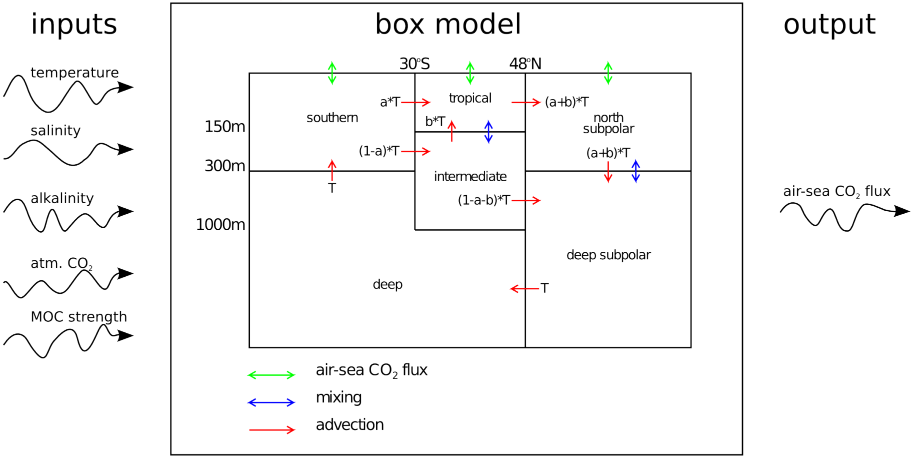


Fig. 3. Schematic description of the box model.

~~**Top:** Histogram showing the distribution of R^2 values describing the relationship between box-model and ESM simulations for each of the 27 ensemble members (using parameter set 1). **Lower plots:** Subpolar North Atlantic air-sea flux simulated within the ESPPE (grey), and emulation of that flux within the box model using the three parameter sets resulting in the lowest mean R^2 value (table 2) displayed in green, red and blue in order of decreasing mean R^2. The three ensemble members displaying the highest R^2 between ESSPE and box model with parameter set 1, and the three ensemble members displaying the lowest R^2 between ESSPE and box model with parameter set 1 are displayed on the right and left with the best (worst) fit at the top. We highlight the difference in goodness of fit between best and worst situations to demonstrate that it is small compared to common behaviour - i.e. the behaviour that we are trying to understand.~~

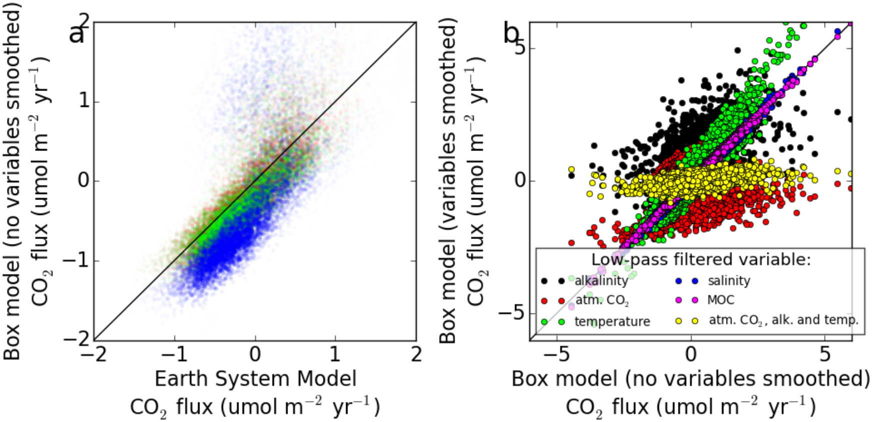


Fig. 4. **a** ESPPE subpolar North Atlantic air-sea CO_2 flux anomaly plotted against box model estimates of that same flux using the top three box model parameter sets (table 2) in red, blue and green respectively. **b** results from box model driven with low-frequency variability in all input variables, plotted against: box model results when low-frequency alkalinity signal is removed (black), low-frequency atm. CO_2 signal removed (red), low-frequency temperature signal removed (green), low-frequency salinity signal removed (blue), low-frequency meridional overturning circulation (MOC) signal removed (purple), and low-frequency atmospheric CO_2 concentration, alkalinity and temperature signals all removed. The straight line represents the one-to-one line upon which results would fall if removal of the low-frequency variability in that variable did not influence CO_2 uptake.

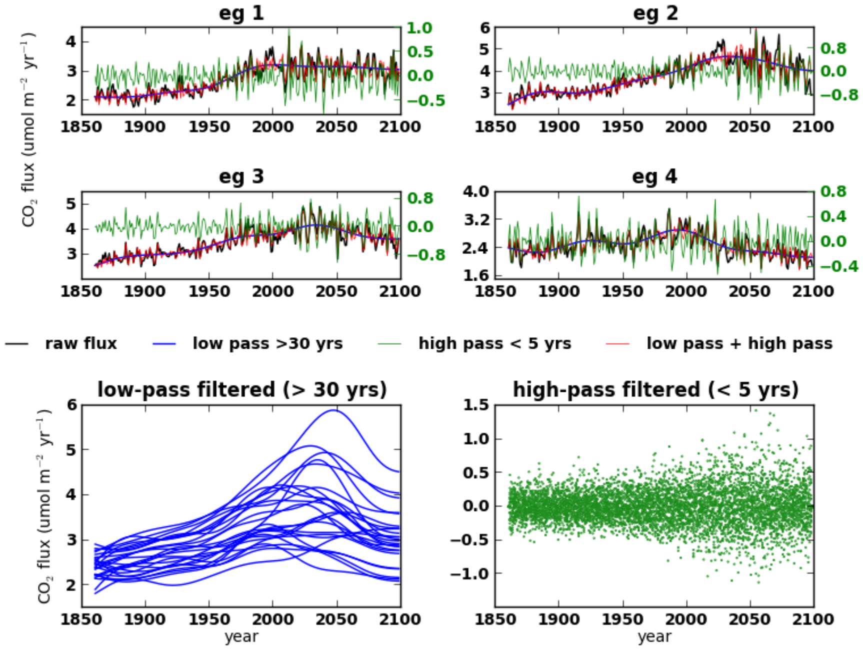
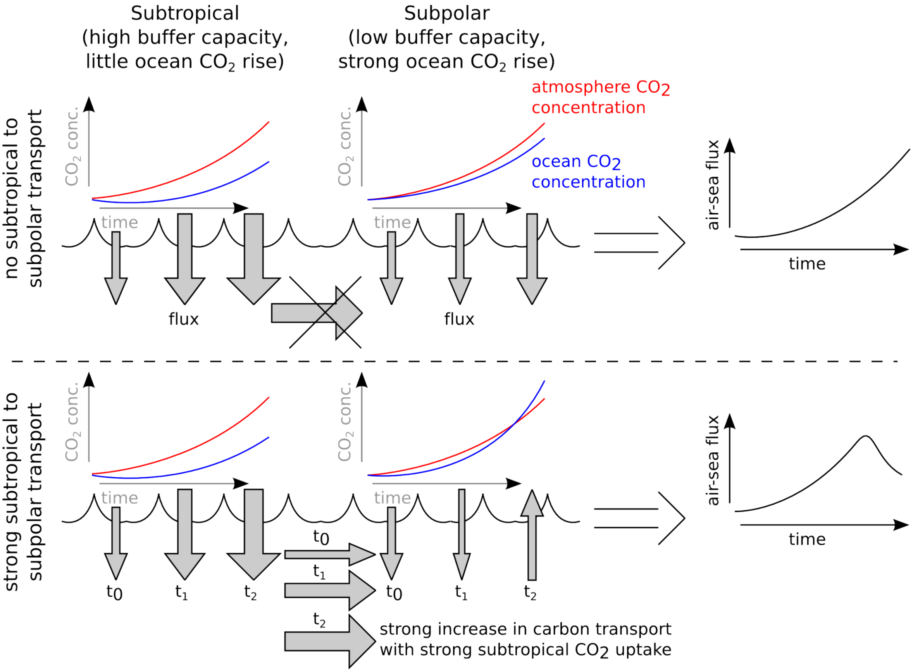


Fig. 5. High and low pass filters are applied to the ESPPE subpolar North Atlantic air-sea CO$_2$ flux simulations to identify the separate time-scales of variability. Top panel: Four random ensemble members' CO$_2$ flux is presented (black) alongside the low-pass (blue) and high-pass (green) processed fluxes. In red, the low and high pass filtered data are recombined to demonstrate that these timescales of variability together explain almost all of the original variability. Lower panel: The low-pass (blue, left) and high-pass (green, right) filtered results across all ensemble members are presented, demonstrating, in the case of the low-pass filters results, great diversity in model evolution.


Subtropical
(high buffer capacity,
little ocean CO_2 rise)
Subpolar
(low buffer capacity,
strong ocean CO_2 rise)
atmosphere CO_2 concentration
ocean CO_2 concentration
no subtropical to subpolar transport
CO_2 conc.
time
flux
air-sea flux
time
strong subtropical to subpolar transport
t_0
t_1
t_2
strong increase in carbon transport
with strong subtropical CO_2 uptake

~~Illustration of the fit (three best and worst case ensemble members as assessed by R^2) between low-pass filtered ESPPE North Atlantic subpolar Gyre air-sea CO_2 flux and the box model results when driven with individual input time-series which have been high-pass filtered. Similarity between the dark blue and green lines highlights the box model's ability to replicate the ESM ensemble's behaviour. Further colours illustrate the dependance of that fit on the time-evolution of the various input variables.~~

Diagrammatic explanation of the mechanism proposed in Völker at al. (2002) by which subpolar North Atlantic CO_2 concentration may peak then decline in response to continuously rising atmospheric CO_2 concentrations. The top half of the diagram explains what would happen if the ~~subtropical~~ low latitude and subpolar Atlantic were not connected by the circulation of the ocean (AMOC). Here, the higher alkalinity to dissolved-carbon ratio (the warm and saline low-latitude waters) of the subtropics means that these waters can strongly take up anthropogenic CO_2 without a big rise in surface ocean CO_2 concentrations. Similarly the higher latitude subpolar waters (with low alkalinity to dissolved-carbon ratios) continuously take up CO_2, but the (relatively) small buffering capacity of these waters means that the surface ocean CO_2 concentration rises (relatively) quickly. A smaller air-sea CO_2 gradient is therefore maintained, and the air-sea CO_2 flux is (relatively) small. The bottom half of the diagram represents the situation in the real ocean, and the simulations considered in this study. Here the ~~subtropical~~ low latitude and subpolar Atlantic are linked by the near-surface limb of the Atlantic Meridional Overturning Circulation. In this situation, in response to rising atmospheric CO_2, the ~~subtropical~~ low latitude CO_2 uptake continues (in our idealised example) as in the top half of the diagram, but some of that that extra carbon is being moved into the subpolar Atlantic, where the buffering capacity is lower, and the water does not have the capacity to hold as much extra carbon as CO_2. This could ultimately result in the subpolar Atlantic becoming a source for anthropogenic CO_2 rather than a sink, as it may not have the capacity to hold the extra CO_2 being passed to it from the south.

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Fig. 6. ~~Illustration of the fit (best and worst case as assessed by R^2) between high-pass filtered ESPPE North Atlantic subpolar gyre air-sea CO_2 flux and the box model results when driven with high-pass filtered input time-series. Similarity between the dark blue (ESPPE subpolar North Atlantic air-sea CO_2 flux) and green lines illustrates the box model's ability to capture high frequency variability in the ESM ensemble. Red, light blue and purple lines show how the box-model's fit to the ESPPE's high-frequency subpolar North Atlantic air-sea CO_2 flux variability is dependant on temperature and alkalinity. Factors other than temperature and alkalinity do not play an important role in variability on this timescale (figure ??) so have been excluded from this figure for clarity.~~
~~Illustration of the fit (three best and worst case ensemble members as assessed by R^2) between low-pass filtered ESPPE North Atlantic subpolar Gyre air-sea CO_2 flux and the box model results when driven with individual input time-series which have been high-pass filtered. Similarity between the dark blue and green lines highlights the box model's ability to replicate the ESM ensemble's behaviour. Further colours illustrate the dependance of that fit on the time-evolution of the various input variables.~~
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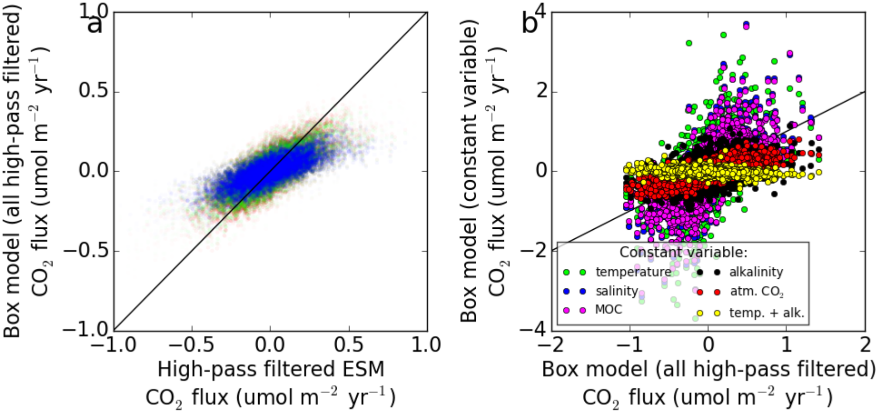


Fig. 7. ~~Subpolar North~~ **a** High-pass filtered ESPPE subpolar Atlantic ~~surface ocean temperature~~ air-sea CO$_2$ flux plotted against box model estimates of that same flux using the top three box model parameter sets (table 2) in red, green and ~~alkalinity~~ blue respectively, but forced with high-pass filtered input time-series. **b** All box model inputs high-pass filtered plotted ~~from~~ against all box model inputs high-pass filtered but one variable held constant. The constant variable in each case is named within the ~~ESPPE simulations~~ legend. All results are presented as anomalies from ~~their respective first 20 year~~ the mean.

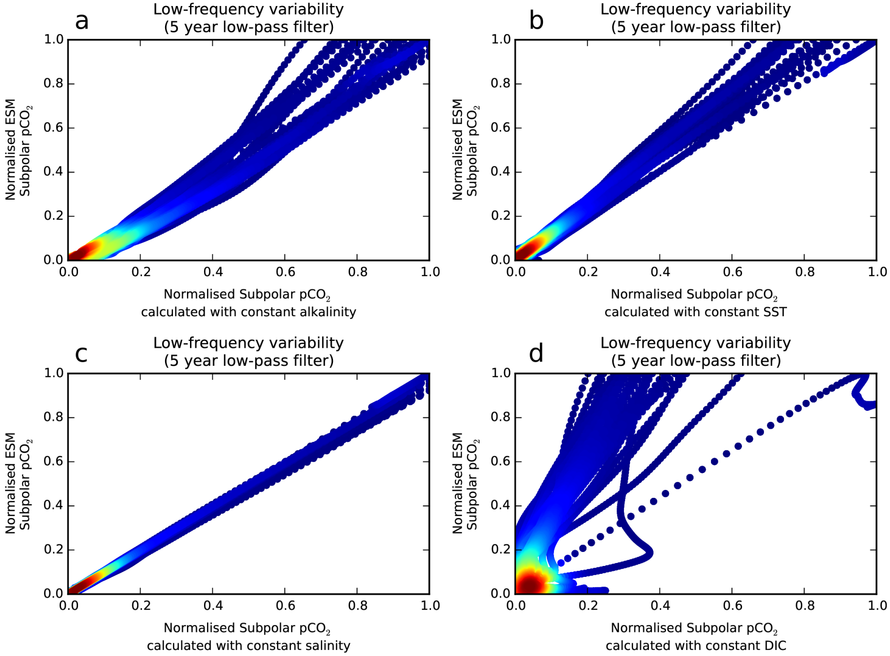
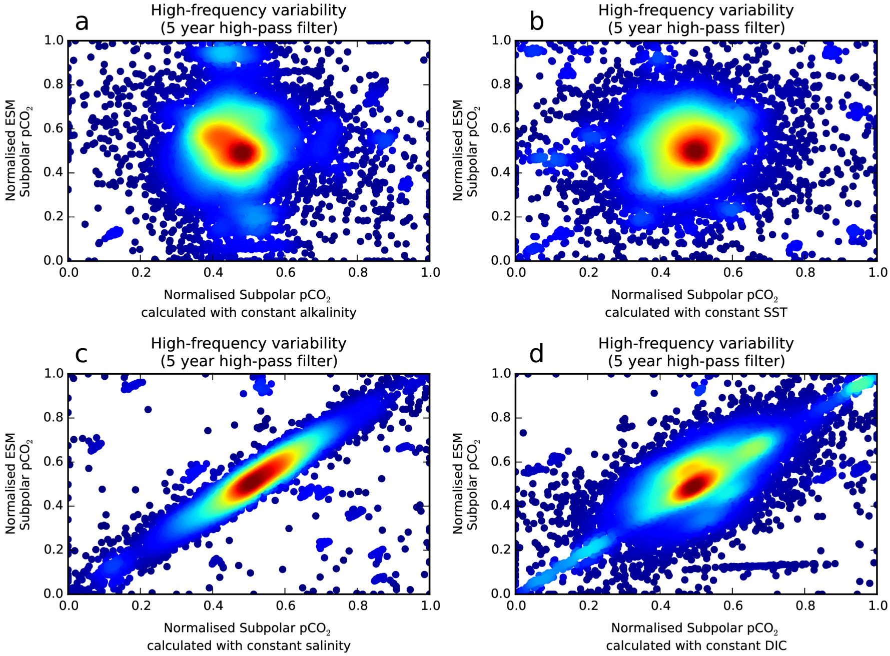


Fig. 8. Low-pass filtered ESM Subpolar Gyre (SPG) pCO_2 plotted against low-pass filtered ESM SPG pCO_2 calculated with alkalinity only (a), SST only (b), salinity only (c) and DIC only (d) held constant through time. Points represent annually averaged values. Colours from blue to red represent an increasing density of points.



~~**a** High-pass filtered ESPPE subpolar Atlantic air-sea CO_2 flux plotted against box model estimates of that same flux using the top three box model parameter sets (table 2) in red, green and blue respectively, but forced with high-pass filtered input time-series. **b** All box model inputs high-pass filtered plotted against all box model inputs high-pass filtered but one variable held constant. The constant variable in each case is named within the legend.~~

Fig. 9. High-pass filtered ESM Subpolar Gyre (SPG) pCO_2 plotted against high-pass filtered ESM SPG pCO_2 calculated with alkalinity only (a), SST only (b), salinity only (c) and DIC only (d) held constant through time. Points represent annually averaged values. Colours from blue to red represent an increasing density of points.

t

Table 1. Parameters used in box model

Parameter Name	Parameter ~~Value~~ description	Parameter ~~description~~ Range (for tunin
T	~~variable~~ overturning circulation strength (Sv)	n/a: as prescribed from ESM
a	~~7/14~~ fraction of overturning circulation strength	0-1
b	~~2/14~~ fraction of overturning circulation strength	1-a
mix_{eq}	~~5~~ vertical mixing (Sv)	0-20
mix_{north}	~~7~~ vertical mixing (Sv)	0-20
flux_{south}	~~0.0037~~ southern box piston velocity (m/hour)	0-0.4
flux_{eq}	~~0.0063~~ equatorial box piston velocity (m/hour)	0-0.4
flux_{north}	~~0.12~~ northern box piston velocity (m/hour)	0-0.4

Table 2. Box model parameter values

Ranking	Parameter						
	piston (>48°N)	piston (<30°S)	piston (30°S to 48°N)	mixing	mixing2	a	b
1st	0.177	0.0854	0.142	1.02	2.09	0.286	0.0103
2nd	0.168	0.138	0.211	19.2	12.7	5.37e-03	8.72e-02
3rd	2.82e-02	0.321	0.129	13.1	17.1	1.16e-02	0.727
4th	0.130	0.399	1.56e-03	6.76	8.65	0.423	2.42e-02
5th	8.56e-02	0.199	0.104	8.33	1.09	2.22e-03	8.60e-02
6th	0.159	0.0632	0.0136	10.9	13.1	0.608	0.288