Response to the referees concerning BGD manuscript: The ocean carbon sink – impacts, vulnerabilities, and challenges, by C. Heinze, S. Meyer, N. Goris, L. Anderson, R. Steinfeldt, N. Chang, C. Le Quéré, and D. C. E. Bakker.

The authors would like to thank the two anonymous referees for their thorough reviews and excellent suggestions for improving the manuscript. We respond below to their comments and explain the changes carried out in the revised version of the manuscript. Line and page numbers refer to the originally submitted version of the manuscript. The *referee comments* are marked by *text in italics*.

Response to referee #1:

MAIN COMMENTS:

Referee's comment: General remarks. The paper submitted reviews the state of the knowledge of the marine carbon cycle research, in particular in respect to the uptake of anthropogenic carbon into the ocean. The paper addresses an important topic, which is suitable to be published in the Journal "Earth System Dynamics". The paper is generally well written, and I recommend it for publication after minor to moderate revision by consideration of the points listed below.

Referee's comment: I recommend to reorganize Chapter 3 and to address the variability of the oceanic carbon sink by distinguishing between the natural and anthropogenic carbon variability (perhaps in two independent sections) and then address the processes that lead to the changes. Include a discussion on how the natural and anthropogenic sources and sinks depend on seasonal variability, interannual (e.g. ENSO), and decadal variability e.g. NAO/NAM, PDO, AMO).
Authors' response: We re-organised the manuscript as described below, also taking into account the suggestions made by referee #2. We tried to reconcile the requests of both referees in the new version of the manuscript. Concerning the climate variability modes, we do not completely agree with the referee. We had in fact discussed ENSO and NAO in our first submitted version. But we added more detail about these and other climate modes in the new section 3.3 and also explicitly mention the Southern Annular Mode on page 18, line 5.

<u>---START OF DESCRIPTION OF THE RE-ORGANISATION OF THE MANUSCRIPT---</u>

WISH FOR RE-ORGANISATION BY referee #1:

Address the variability of the oceanic carbon sink (perhaps in two independent sections) by distinguishing between the
(a) natural carbon variability and
(b) anthropogenic carbon variability,
(c) and then address the processes that lead to the changes.

Further: Include a discussion on how the natural and anthropogenic sources and sinks depend on seasonal variability, interannual (e.g. ENSO), and decadal variability e.g. NAO/NAM, PDO, AMO).

WISH FOR RE-ORGANISATION BY referee #2:

Section 2.
(a) main physico-chemical processes responsible for the uptake of excess CO2,

(b) processes regulating the natural distribution of carbon within the ocean and the inventory split between ocean and atmosphere,
(c) feedbacks from climate and environmental changes on a) and b).

Further: …and by clearly separating different time scales, e.g. seasonal, interannual, decadal, century-scale, glacial-interglacial.

Section 3.
Section 3 on variability. What I miss here is a proper discussion of internal modes of variability (ENSO; SAM, NAO, NPD, etc.) and of external drivers of variability and their imprints, e.g. volcanic forcing. As well as on detection of signals (signal-to-noise, time of emergence) and the attribution of signals to underlying processes.

OUR CONCEPT IN THE ORIGINALLY SUBMITTED VERSION:

Section 2. (GENARAL CONCEPTS AND MAIN PROCESSES – NATURAL AND ANTHROPOGENIC. MAYBE TITLE SHOULD HAVE BEEN CHOSEN MORE CLEARLY.)
2 The role of the oceans for carbon cycling.
2.1 Inorganic carbon cycle processes (includes pristine and anthropogenic carbon).
2.2 Biological carbon pumps (includes pristine and anthropogenic carbon, also glacial).

Section 3. (VARIABILITY OF THE OCEANIC SINK FOR ANTHROPGENIC CARBON. MAYBE TITLE SHOULD HAVE BEEN CHOSEN MORE CLEARLY.)
3 Variability, time evolution, and kinetics of the ocean carbon sink.
3.1 Variability of the oceanic carbon sink (more a brief introduction and an analysis).
3.2 Time evolution and kinetics of the oceanic carbon sink (deep time in future, dependence on future scenario).
3.3 Observations of ocean carbon variability (can we already now see any changes in the ocean carbon sink, focus not intended on discussing variability of climate modes).

CHALLENGES IN RECONCILING THE SUGGESTIONS FOR RE-ORGANISATION:

Referee #1 agrees mostly with the structure of original section 2.
Referee #2 agrees mostly with original section 3.

Referee #1 wishes a more detailed discussion of time scales of variability in section 3 (seasonal, interannual, decadal, climate modes).
Referee #2 wishes a more detailed discussion of time scales of variability in section 2 (seasonal, interannual, decadal, century-scale, glacial-interglacial) and a discussion of internal climate modes in section 3.

Referee #1 wishes a separation between natural and anthropogenic variability in section 3.
Referee #2 wishes a separate description on main physical-chemical anthropogenic carbon uptake processes and natural atmosphere-ocean partitioning processes in section 2 (further referee #2 would like to see how anthropogenic carbon uptake gets altered under climatic and environmental change and how the natural partitioning of carbon between atmosphere and ocean gets altered under climatic and environmental change).

Conclusion: The two referees would like to see a clearer separation between natural and anthropogenic carbon cycle processes, a more detailed discussion of time scales, and an inclusion of climate modes (ENSO etc.), but sometimes in different sections.

SUGGESTION FOR ADJUSTMENT OF STRUCTURE:

In our originally submitted version, we summarise the basic processes for the general cycling of carbon and the key inorganic buffering mechanism as well as a discussion of the biological pump mainly for natural carbon cycle modifications (including glacial) in section 2. Section 3 is focusing on anthropogenic carbon uptake discussing the kinetics and variability of the ocean sink for excess carbon and how this sink can be simulated (for the future) and diagnosed from observations.

We think that this structure (section 2 – basic processes, section 3 – timing of anthropogenic marine carbon sink) is still a valid one, but agree with the referees that the section names did not clarify our intention. In our revised version, we make the separation between natural and anthropogenic carbon cycling clearer between sections 2 and 3:

2 General concepts of ocean carbon cycling.
2.1 Inorganic carbon cycle processes .
2.2 Biological carbon cycling.
2.3 Natural variability, timescales, and feedbacks *(for climate modes refer to section 3).*

3 Evolution of the ocean sink for anthropogenic carbon.
3.1 The key process for anthropogenic carbon uptake *(mainly inorganic buffering, slight modulation by biological processes)*.
3.2 Long-term ocean carbon uptake kinetics.
3.3 Detection of ongoing ocean carbon sink strength variability *(we bring here also climate modes, time of emergence, North Atlantic and Southern Ocean diagnosed sink strength variability, do modes cancel out over time etc.)*.

Section 2 focuses now only on natural carbon cycling and variability. Section 3 focuses now only on the uptake of anthropogenic carbon and changes therein. The requested discussion about climate modes is mainly included in section 3, but already briefly mentioned in section 2 (new sub-section 2.3). We added a section on time scales in section 2 (new sub-section 2.3) and move the discussion of inorganic buffering of excess CO_2 from section 2 to section 3 (new sub-section 3.1). Ocean carbon cycle feedbacks to anthropogenic physical and chemical forcing are included in the new sub-section 3.2.

<u>---END OF DESCRIPTION OF THE RE-ORGANISATION OF THE MANUSCRIPT---</u>

Referee's comment: Section 6.4 should be extended, for example on the North Atlantic and North Pacific Ocean. The North Atlantic Ocean is of interest because carbon uptake is sensitive to changes in the Atlantic meridional overturning circulation. Results from eddy-resolving ESMs with freshwater pulses due to melting glaciers by climate change (e.g. Weijer et al., GRL, 2012) indicate a significantly different tracer distribution than non-eddy resolving ESMs.
Authors' response: We aimed with this section 6.4 at highlighting those regions which need more attention than traditionally well observed oceanic domains. But we see that this could lead to misunderstandings and therefore added a short paragraph concerning the North Atlantic and North Pacific. A discussion on eddy-resolving glacier melt experiments would go beyond the scope of our article.
Authors' changes in manuscript:
We added a paragraph under 6.4 – "7. Of course, the traditionally comparatively well observed North Atlantic and North Pacific domains (see, e.g., Bakker et al., 2014) should be further kept in the focus of monitoring and modelling programmes. The North Atlantic is a critical area for anthropogenic marine carbon uptake and changes in this may occur due to a changes in meridional overturning and

deep-water production. It has still to be firmly established whether any long-term (more than two decades) changes in the trend of anthropogenic CO_2 uptake occur in these regions which are marked also by internal variability in coupling to prevailing climate variability modes such as the North Atlantic Oscillation and the Pacific Decadal Oscillation."

Referee's comment: Please change "ppm" to "ppmv" throughout the text.
Authors' response: Accepted.
Authors' changes in manuscript: We changed "ppm" to "ppmv" throughout the text.

SPECIFIC LINE BY LINE COMMENTS:

Referee's comment: Abstract L. 1 Change "important" to "abundant".
Authors' response: We disagree. We want to stress that CO_2 is the key greenhouse gas. It would be correct to also say that it is the most abundant one after water vapour for many regions (water vapour content varies strongly and can go down to almost 0%). However, greenhouse gas importance does generally not only depend on the abundance of the gas, but also on its specific greenhouse gas potential (radiative forcing per molecule of greenhouse gas).
Authors' changes in manuscript: No change.

Referee's comment: Abstract L. 2 Change "mitigation" to "adaptation and mitigation".
Authors' response: Accepted.
Authors' changes in manuscript: Change made.

Referee's comment: Abstract L. 12 Change "load" to "inventory".
Authors' response: Accepted.
Authors' changes in manuscript: Change made.

Referee's comment: Abstract L. 13 Replace "services" with "impact".
Authors' response: We disagree. Writing "affect its ecosystems" includes that ecosystems will be impacted. "Ecosystem services" is an established term, which goes beyond impacts. It also includes what consequences less well functioning ecosystems will experience in terms of food production, greenhouse gas neutralisation, etc.
Authors' changes in manuscript: No change.

Referee's comment: Page 4 L. 2 Reference Figure 1, L. 4. During the geologic past atmospheric pCO2 was variable. Specify geologic time for which the 280 ppmv value is applicable (e.g. Holocene).
Authors' response: We do not know what the referee would like to have changed at line 2 on page 1610 (would be Page 4 in the non-printer-friendly format of the pdf; in the printer-friendly pdf version always two manuscript pages show up on one pdf page). We assume that the referee wants to stress that the preindustrial value of atmospheric pCO_2 is variable. We describe this in our text on page 1610 lines 14 and following.
Authors' changes in manuscript: In order to be consistent in the caption of Figure 1 and the text on page 1610, we changed "280 ppm" to "278 ppmv" on page 1610. In the caption of Figure 1, we changed "278 ppm" to "around 278 ppmv".

Referee's comment: Page 6 Section 2 L. 2 Change "gas exchange" to "air-sea gas exchange". Include references for air-sea gas exchange (e.g. Liss and Merlivat, 1986; Wanninkhof, 1992), solubility (Weiss, 1974), and carbon dioxide dissociation (Broecker and Peng, 1982).
Authors' response: Accepted.
Authors' changes in manuscript: We changed "gas exchange" to "air-sea gas exchange" and added the references Liss and Merlivat, 1986; Wanninnkhof, 1992; and Nightingale et al., 2000. For

solubility, we added the reference of Weiss, 1974. For the CO_2 dissociation, we cite the more up-to-date work of Dickson et al., 2007, instead of Broecker and Peng, 1982.

Referee's comment: Page 7 L. 17 Quantify the solubility with an example, e.g. for an increase of 3 °C (For 2XCO_2) from 14 °C (which is about the global mean SST); references Weiss (1974).
Authors' response: Rejected. The idea of giving an example for the solubility dependence on temperature is appealing and we thank the reviewer for the suggestion. However, the antagonistic effect of the improved dissociation of CO_2 (and hence improved buffering ability of seawater with temperature increase) would need to be given in addition. But this is not so easily done as for the pure solubility effect. In order to not complicate the present manuscript and keep it easily accessible also to multi-disciplinary communities, we decide for the time being not to add this. Mentioning the solubility effect only would easily give rise to misunderstandings concerning a temperature dependent change in marine anthropogenic carbon uptake.
Authors' changes in manuscript: No change.

Referee's comment: Page 8 L. 13 Reference Volk and Hoffert (1985).
Authors' response: Accepted.
Authors' changes in manuscript: We now cite Volk and Hoffert (1985) in addition to the referee's request already after introducing the solubility pump (Page 7, line 19). Further, we change the sentence in the beginning of section 2.2 addressed by the referee to: "While purely inorganic carbon cycling leads to a slight increase of DIC with depth, biological carbon cycling - via the two biological carbon pumps (see Volk and Hoffert, 1985) - is responsible…"

Referee's comment: Page 15 L. 20 Change "oxygen" to "dissolved oxygen".
Authors' response: Accepted.
Authors' changes in manuscript: Change made.

Referee's comment: Page 16 L. 3. Perhaps the section title could be renamed to "Observed variability of the ocean carbon sink".
Authors' response: We adjusted the section title to "Detection of ongoing ocean carbon sink strength variability". Mainly observational evidence is discussed there, but models are needed as well for this task.
Authors' changes in manuscript: New section title : "Detection of ongoing ocean carbon sink strength variability".

Referee's comment: Page 17 L. 21 Reference Wetzel et al., 2005, Global Biogeochemical Cycles.
Authors' response: Accepted.
Authors' changes in manuscript: Reference was added.

Referee's comment: Page 19 L. 27 Reword sentence, include reference: Copenhagen Accord U.N. Framework Convention on Climate Change. United Nations. 18 December 2009.
Authors' response: Accepted. However, the reviewer does not say specifically why the sentence should be reworded or what is wrong with it.
Authors' changes in manuscript: We changed the sentence including a more up-to-date citation of the Copenhagen Accord to: "A future global warming limit of 2°C above the average preindustrial surface temperature has been suggested as a not yet very ambitious and thus potentially achievable political target for greenhouse gas emission strategies (see Tol, 2007; Meinshausen et al., 2009; United Nations, 2010; Schellnhuber, 2010)."

Referee's comment: Page 30 L. 13 Change "reviewed in Heinze and Gehlen, 2013" to "see e.g. Heinze and Gehlen, 2013, for review".
Authors' response: Accepted.

Authors' changes in manuscript: Change made.

Referee's comment: Page 30 L. 23. Reword "not easily done". For the reader it is rather of interest what the novel approaches or challenges are.
Authors' response: Accepted.
Authors' changes in manuscript: The sentence was changed to: "Even for still fairly coarse resolutions this is currently not easily done and quite costly in terms of super computer processing time."

Referee's comment: Page 30 L. 27 Reference see CMIP5 simulations,
Authors' response: We agree that many of the CMIP5 simulations show the effect, but there is no summarising paper on this particular aspect. Therefore, we give examples.
Authors' changes in manuscript: We inserted an "e.g." at the beginning of the references given.

Referee's comment: Page 30 Figure 2. Carbonic acid can be deleted from the equation because the concentration is less than 0.1% (see Schulz, Marine Chemistry, 2006). In the figure you can replace H_2CO_3 with dissolved CO_2.
Authors' response: Accepted.
Authors' changes in manuscript: Changes made.

Response to referee #2:

MAIN COMMENTS:

Referee's comment: This review summarizes the current view on the marine carbon cycle and its perturbation by humans. Research challenges and knowledge gaps are discussed.
The review is useful and I suggest publication.

I miss a discussion on useful carbon isotopes and non-carbon tracer variables, including ^{13}C, ^{14}C and CFCs, SF_6, Ar-39 (new interest as low water volume samples may be in reach with atomic trap technology) that provide information on the time scales of water mass movements relevant for the mixing of excess carbon to the deep ocean.
Authors' response: We accept that the manuscript can benefit from filling this gap. However, we did not include SF_6 in our discussion as this is used mainly for tracing water masses in a Lagrangian way and is not really essential for quantifications of marine carbon uptake.
Authors' changes in manuscript: A corresponding text was added in our new section 3.1. after "…be validated by age tracers" – "To demonstrate that ocean carbon cycle models work properly, the inclusion of the organic carbon cycling in these models, therefore, is an important necessary condition. On the other hand, uptake of anthropogenic excess CO2 from the atmosphere is mainly determined by the physico-chemical buffering mechanism and transport of water with high anthropogenic carbon concentrations into the ocean interior. Nevertheless, simulations of biologically mediated tracers such as O2, PO43- etc. help to constrain the oceanic velocity field of the respective model, especially because respective measurements are abundant. Further, the biologically mediated CO32- ion distribution is a powerful constraint on whether the inorganic carbon cycle is correctly described by the models. The simulation of anthropogenic marine carbon uptake in purely inorganic carbon cycle models (i.e. those which do not include ecosystem representations, no nutrient tracers, and no oxygen cycle) can to some degree be validated by age tracers which are employed also for evaluation of ocean model velocity fields in general.. Radiocarbon ^{14}C, which enters the ocean mainly from the atmosphere, is still the most used age tracers for validating oceanic transport rates as well as patterns in ocean circulation models. With its half-life of 5730 years (sometimes also the slightly smaller Libby half-life is used; see Stuiver and

Polach, 1977), radiocarbon of DIC results in substantial surface to deep gradients. The natural radiocarbon distribution is contaminated by bomb ^{14}C, which entered the ocean in large amounts due to atmospheric tests of nuclear weapons until the atmospheric test ban treaty in the mid-1960s was implemented. To some degree, bomb ^{14}C can also be used as tracer for water mass exchange in itself, but the lack of knowledge about the pristine ^{14}C distribution on already contaminated areas remains a problem in spite of attempts to reconstruct natural pre-bomb ^{14}C values in the ocean interior (Broecker et al., 1995). Nevertheless, for the large scale ocean, ^{14}C remains one of our best tracers for assessing turnover rates of water masses in the ocean (cf. Schlitzer, 2007). Another, in principle powerful age oceanic tracer is the noble gas isotope ^{39}Ar. Its shorter half-life of 269 years (Stoenner et al., 1965) would even be more suitable to resolve upper ocean gradients for validation of ocean ventilation time scales in models (Müller et al., 2006). New measurement techniques allowing for small sample size may enable building a larger ^{39}Ar data base for the ocean (Collon et al., 2004).

As supporting evidence for pathways of anthropogenic carbon from the atmosphere over the surface layer and into the ocean interior, also ^{13}C and chlorofluorocarbons are used. Fossil fuel CO_2 in the atmosphere has a low ^{13}C signature (plant material that had been the basis for crude oil formation has a deficit in the stable carbon isotope ^{13}C relative to ^{12}C, also known as the Suess effect; see Keeling, 1979). Waters with a deficit of ^{13}C in DIC relative to natural background conditions, therefore contain carbon from anthropogenic sources (Racapé et al., 2013). Unfortunately, the reconstruction of the pristine ^{13}C distribution in the ocean is not straightforward (Olsen and Ninnemann, 2010), and further the ^{13}C distribution in the ocean is strongly influenced by formation as well as degradation of biogenic matter (Kroopnick, 1985). Chlorofluorocarbons or 'CFCs' (such as $CFCl_3$ or 'F-11' and CF_2Cl_2 or 'F-12') are purely human-produced substances (also known for their negative effect on the stratospheric ozone layer) which entered the oceans from the atmosphere in small amounts following their atmospheric concentration and their respective solubilities in seawater. Though their atmospheric concentration time series and their uptake mechanisms in the ocean are different than for CO_2, they nevertheless give a constraint on where large amounts of anthropogenic carbon have entered deeper layers and what time scales are involved with this uptake (Smethie, 1993; Schlitzer, 2007; Steinfeldt et al., 2007)."

Referee's comment: Section 2. I miss a clear organization and structure of the presentation. Personally, I would have preferred if the authors would have organized this section by clearly distinguishing between
a) main physico-chemical processes responsible for the uptake of excess CO2,
(b) processes regulating the natural distribution of carbon within the ocean and the inventory split between ocean and atmosphere,
(c) feedbacks from climate and environmental changes on a) and b)
and by clearly separating different time scales, e.g. seasonal, interannual, decadal, century-scale, glacial-interglacial.
The relevant text in the carbon cycle chapter in the IPCC Third Assessment Report provides a good example.
Authors' response and respective changes in manuscript: We have rearranged section 2. However, we find the critics of the referee not completely justified. Our presentation in the submitted version discriminating between physical/chemical and biological carbon cycling is a valid one and has been used also in other presentations of the ocean carbon cycle. For our re-arrangement: Please see our discussion on the re-organisation of the manuscript as given under the response to referee #1 where we also account for the suggestions of referee #2. Further, we added the following text on page 1617, line 24 – "Among these biological changes are a potential decrease in biological $CaCO_3$ production (Heinze, 2004; Gehlen et al., 2007; Ridgwell et al., 2007) and a potential change in carbon to nitrogen ratios in oceanic organic matter under high CO_2 (Riebesell et al., 2007).".

Referee's comment: Section 3 on Variability What I miss here is a proper discussion of internal modes of variability (ENSO; SAM, NAO, NPD, etc) and of external drivers of variability and their imprints, e.g. volcanic forcing. As well as on detection of signals (signal-to-noise, time of emergence) and the attribution of signals to underlying processes.
Authors' response and authors' changes in manuscript: We do not completely agree with the referee. We have in fact discussed ENSO and NAO as the most important variability modes in section 3, but added more detail about these and other climate modes in the revised section 3.3 and also explicitly mention the Southern Annular Mode on page 1624, line 6. We now also mention the PDO.

DETAILED COMMENTS:

Referee's comment: Line 15, abstract: Would argue for the entire hierarchy of models, including EMICS, and not just so-called state-of-the-art ESMs.
Authors' response: Partially accepted. The term Earth system models includes EMICs as a sub-group already.
Authors' changes in manuscript: We deleted "state-of-the-art".

Referee's comment: Abstract: what about inverse methods/models and probabilistic approaches?
Authors' response: Rejected. Such methods would be mentioned in the text if necessary, but do not add to the essence of the article which should be given in the abstract (we write "including" and thereafter highlight the most important issues).
Authors' changes in manuscript: No change.

Referee's comment: Page 1610: the first paragraph in the introduction needs to be revised. page 1610, line 6: I am not so familiar with the history. As far as I know, Tyndall was the first to prove experimentally that CO_2 is a greenhouse gas in the 1860ies or so. Not sure that the structure of the molecule was known back then as this requires knowledge on quantum mechanics.
Authors' response: We checked that Tyndall in his papers (Tyndall, J., 1861, On the absorption and radiation of heat by gases and vapours, Philosophical Magazine, 22, 169-194; Tyndall, J., 1859, Gaseous Note on the Transmission of Heat Through Radiant Bodies, Proc. R. Soc. Lond. 1859-1860 10, 37-39) did not identify CO_2 as a greenhouse gas, but a series of other gases. He may have identified CO_2 in another study, but so far we did neither find an original reference or anything in the secondary literature. The greenhouse effect as such was already detected by Fourier in 1824 (Fourier, J.B. 1824. Mémoire sur les températures du globe terrestre et des espaces planetaires. Mem. Acad. Sci. Inst. Fr. 7, 569-604) and followed up by Pouillet, 1838 (Pouillet CSM. 1838. Mémoire sur la chaleur solaire, sur les pouvoirs rayonnants et absorbants de l'air atmosphérique, et sur la température de l'espace. Comptes rendus de l'Académie des Sciences 7: 24–65). From original sources, Arrhenius publication seems to be the first one to explicit mention CO_2. Of course, the CO_2 molecule structure was not known at that time.
Authors' changes in manuscript: The introduction lines have been changed to avoid misunderstandings – "In the atmosphere, carbon dioxide (CO_2) occurs only in a very small fraction (currently around 400 ppmv; ppmv = parts per million of volume; http://scrippsco2.ucsd.edu/graphics_gallery/mauna_loa_record.html) .Nevertheless, due to its high abundance as compared to other greenhouse gases, it is considered to be the overall most important greenhouse gas next to water vapour. Its importance in regulating the global heat budget has already been documented in the 19th century by Arrhenius (1886)."

Referee's comment: Page 1610, line 7: What about rotational modes?
Authors' response: Accepted.

Authors' changes in manuscript: We mention now also rotational motion, see our response to the referee's comment concerning page 1610, line 9-11, below.

Referee's comment: Page 1610, line 8: "discrete wavelength intervals" I find the use of 'discrete' together with 'intervall' perhaps a bit confusing in this context. Perhaps distinct intervalls?
Authors' response: Accepted.
Authors' changes in manuscript: We now use the term "bands centred at different discrete wavelengths".

Referee's comment: Page 1610, line 9-11: Again a somewhat inaccurate sentence mixing different concepts. Solar radiation is electromagnetic as well and most of the energy radiated by the sun and from Earth's surface is thermal. Would also say that the vast majority of the energy from the sun is in the short-wave range and similar that most of the radiation from Earth's surface and the atmosphere is in the long-wave range. Perhaps you wish to give relevant wavelengths.
Authors' response: Accepted.
Authors' changes in manuscript: We changed the text to – "Vibrational and rotational motions of the gaseous CO_2 gas molecules resonate with the thermal radiation leaving Earth's surface at bands centred at different discrete wavelengths intervals, thereby heating up the lower atmosphere (e.g. Barrett, 2005; Tomizuka, 2010). The main absorption band (combined vibrational and rotational resonance mode) of CO_2 is cantered at 15 µm wave length (Wang et al., 1976; Liou, 1980). The incoming solar radiation is of short wavelength (mainly between 0.5-1 µm). The thermal radiation outgoing from the Earth is of longer wave length (typically between 5 and 20 µm). Without the natural greenhouse effect, an average temperature of -19°C would dominate Earth's surface instead of the actual average value of around 15°C (Ramanathan et al., 1987)."

Referee's comment: Page 1610, line 21 ff: If you wish to be so precise with the dates you need to give the location of measurements for direct atmospheric samples. The date derived from the Law Dome ice core data should come with an uncertainty estimate. Note that individual air samples from the ice have an age distribution about the mean age. In addition individual samples have a measurement error.
Authors' response: Accepted. It is better to be more precise here (though the increased precision is not essential for our arguing).
Authors' changes in manuscript: We modified the text to – "The 300 ppmv boundary was crossed in the early 20th century according to ice core measurements from Law Dome (Etheridge et al., 2001; samples from Law Dome core D08 show values of 296.9 ppmv and 300.7 ppmv for mean air ages given in calendar years of 1910 and 1912 respectively, with an overall accuracy due to analytical errors and age determination errors of ±1.2 ppmv)."

Referee's comment: Page 1612, lines 13,14: indicate that the percentages for the different species of DIC are only approximations, in particular for dissolved CO_2.
Authors' response: Accepted.
Authors' changes in manuscript: Text changed to – "Carbon dioxide, or carbonic acid (H_2CO_3) when combined with water (H_2O), dissociates in seawater mostly into bicarbonate (HCO_3^-) and carbonate (CO_3^{2-}), while only a small amount of the CO_2 is kept in its dissolved state (as an order of magnitude estimate the partitioning of HCO_3^-: CO_3^{2-}:CO_2 is 100:10:1 but significant deviations from this can occur especially with respect to CO_2)".

*Referee's comment: Page 1612, line 18: I find 70 times more DIC than in the atmosphere a high estimate for preindustrial,e.g. 278 ppm*2.12 Pg/ppm/37500 PgC = 1/64.*
Authors' response: Accepted.
Authors' changes in manuscript: We corrected the value to 65 PgC (according to the preindustrial reservoir sizes as given in IPCC AR5, WG1, ch. 6, Fig. 6.1).

Referee's comment: Page 1616 top: I miss here a discussion on the controversy whether calcite and aragonite can dissolve above the saturation horizon or not.
Authors' response: Accepted.
Authors' changes in manuscript: We inserted (page 1616, line 4) the following text – "Nevertheless, it is likely that also partial re-dissolution of calcitic and aragonitic plankton hard parts occurs in shallower depths than the respective $CaCO_3$ saturation horizon. Potential contributors to this are, e.g., zooplankton metabolisms (dissolution of shell material in copepod guts; Jansen and Wolf-Gladrow, 2001), local undersaturation hot spots due to lateral admixture of water or in micro-environments on biogenic particles due to remineralisation of organic matter (Barrett et al., 2014), and admixture of larger amounts of Mg in the $CaCO_3$ material (high-Mg calcites; Feely et al., 2004).".

Referee's comment: Page 1616, line 13: 'biological pump' should be 'biological productivity' as strong upwelling leads often to high surface nutrient concentrations and thus a low degree of surface nutrient utilization and comparably small surface-to-deep nutrient gradients.
Authors' response: Accepted.
Authors' changes in manuscript: Word replaced accordingly.

Referee's comment: Page 1617, line 23: 'biological uptake' should be 'marine uptake'.
Authors' response: Accepted.
Authors' changes in manuscript: Of course, this was corrected.

Referee's comment: Page 1618: The discussion on the comparison with DIC misses the point. Uptake of anthropogenic carbon is by large a physico-chemical process, with the uptake capacity given by carbonate chemistry and the rate limiting step of surface-to-deep transport. A proper representation of ocean uptake of excess CO_2 does require
(i) a correct representation of carbonate chemistry, and
(ii) a correct representation of surface-to-deep transport of excess carbon.
The first process is well known from field and lab studies, the second can be gauged by comparing with, e.g., radiocarbon and CFCs. The comparison with DIC is blurred by the less well understood marine biogechemical cycle.
Authors' response: We agree with the referee on the processes leading to excess carbon in the oceans. However, initially we wanted to stress the following: Purely inorganic carbon cycle models show a distinctly different DIC structure than nature. In nature, the marine DIC distribution including DIC gradients is still dominated by biological processes and not by anthropogenic CO_2 uptake. Therefore, models must include the biological carbon cycle if one wants to compare them with DIC data from the real ocean.
Authors' changes in manuscript: In order to avoid misunderstanding, we changed the text of page 1618, l. 2-11 to – " To demonstrate that ocean carbon cycle models work properly, the inclusion of the organic carbon cycling in these models, therefore, is an important necessary condition. On the other hand, uptake of anthropogenic excess CO_2 from the atmosphere is mainly determined by the physico-chemical buffering mechanism and transport of water with high anthropogenic carbon concentrations into the ocean interior. Nevertheless, simulations of biologically mediated tracers such as O_2, PO_4^{3-} etc. help to constrain the oceanic velocity field of the respective model, especially because respective measurements are abundant. Further, the biologically mediated CO_3^{2-} ion distribution is a powerful constraint on whether the inorganic carbon cycle is correctly described by the models. The simulation of anthropogenic marine carbon uptake in purely inorganic carbon cycle models (i.e. those which do not include ecosystem representations, no nutrient tracers, and no oxygen cycle) can to some degree be validated by age tracers which are employed also for evaluation of ocean model velocity fields in general."

Referee's comment: Page 1621, line 10, next to ocean only and ESM, there are also EMICs that were used to quantify the different processes and feedbacks discussed in this manuscript.
Authors' response: Accepted.
Authors' changes in manuscript: We inserted a respectively updated text – "The oceanic bottleneck effect is obvious in several decade-long future scenarios with ocean models (Maier-Reimer and Hassel Mann, 1987; Sarmiento and Le Quéré, 1996), fully coupled Earth system models (Friedlingstein et al., 2006; Roy et al., 2011; Arora et al., 2013), as well as EMICs (Earth system models of intermediate complexity; these have a lower resolution than usual Earth system models, but demand much less computational resources; Steinacher et al., 2013; Zickfeld et al., 2013). Earth system models are complex computer programmes, which include dynamical representations…."

Referee's comment: Section: 3 I miss here a proper discussion of modes of variability, e.g. the work by Lovenduski, Gruber et al for SAM, Keller et al, Tellus, 2012 for NAO, and very recently Keller et al, GRL, 2015 for ENSO, and of signal-to-noise and time of emergence of a signal as discussed e.g. by Ilyina for Alkalinity, McKinley et al for pCO_2, or Keller et al,BG, 2014 for various variables and reference to detection and attribution (Seferian et al, GRL, 2014).
Authors' response: We are a bit surprised by the comment of the referee concerning the variability modes as we mention NAO and ENSO (and implicitly also SAM) in our original manuscript. But we comply with the suggestion of the referee in general.
Authors' changes in manuscript: We have added more detail to the discussion of internal variability modes in section 3.3 Further we added new paragraphs on external factors for variability and time of emergence – "Not only internal variability modes affect the air-sea CO_2 flux, but also external factors such as aerosol forcing from volcanic eruptions. Such volcanic forcing tends to temporarily cool the troposphere and the sea surface with respective implications for carbon cycling. Brovkin et al. (2010) could identify a temporary small decline of atmospheric pCO_2 by about 2 ppmv a few years after major eruptions over the last millennium, where decreasing respiration on land is a potential leading candidate with the ocean having only a small effect. This is corroborated by Frölicher et al. (2011) for a model study on the effect of Mt. Pinatubo type eruptions on the carbon cycle, where again the terrestrial carbon cycle dominates the atmospheric pCO_2 signal. Nevertheless, transient changes in ocean uptake of about 2 GtC are in a realistic realm as consequences to large volcanic eruptions (Frölicher et al., 2011). Further, it cannot be excluded that also the biological carbon binding is stimulated under deposition of volcanic dust to the ocean surface (Hamme et al., 2010).

In view of the internal and external factors on ocean carbon cycle variability, it is intriguing to ask, when long-term climate change signals become identifiable against the background noise. This problem is of specific concern for large impacts of ocean acidification (see detailed discussion below). Ilyina et al. (2009) identified the equatorial Pacific Ocean to be the oceanic domain where a change in marine biogenic $CaCO_3$ production due to ocean acidification may become at first visible through large-scale changes in ocean surface alkalinity. This can be explained by large background values of pelagic $CaCO_3$ production in the tropical Pacific, though the impact per unit of $CaCO_3$ produced would be highest in the high-latitude surface waters where decreasing $CaCO_3$ saturation proceeds fastest. Generally, the time of emergence of a climate change signal is an important variable: When can we see changes in oceanic state variables which clearly can be attributed to human-induced climate change, i.e. when do trends in key ocean variables emerge as robust on the background of analytical uncertainty and interannual variability? Keller et al. (2014, 2015) provided new insight into this issue. Earth system modelling suggested that sea surface pCO_2 and sea surface pH trends could rise beyond the detection threshold already after 12 years from now. DIC trends would become clear after 10-30 years and trends in the sea surface temperature after 45-90 years (Keller et al., 2014). Accordingly, an earlier detection threshold for changes in mean ENSO-induced carbon cycle variability (pCO_2, pH, biological productivity) than for ocean temperature changes during the 21st century was predicted by Keller et al. (2015). Therefore, ocean carbon cycle observations play a key role as early warning indicators when monitoring climate change. For the time interval 1960-2005, Séférian et al. (2014), however, state that the evolution of the global carbon sink can mainly be explained through rising

CO_2 in the atmosphere and oceanic carbon uptake without invoking a climatic feedback. Nevertheless, at regional scale, trends in climate change become also visible in shaping the regional sink strength pattern."

Referee's comment: Page 1623, top: I think it is not so clear that SO uptake of excess CO2 really slowed and there is a controversial discussion in the literature which should be referenced here.
Authors' response: We think that the referee refers to page 1624 (and not 1623). The evidence against a (transient) weakening of the Southern Ocean carbon sink is small and mainly based on GCM results. Simulations of Southern Ocean deep mixing and dynamics are usually a weak point in GCMs. Therefore, we rather refrain from stressing this point too much.
Authors' changes in manuscript: We added the following text on page 1624 in line 6 and moved the reference for Lenton et al. (2013) – "Due to the fairly short observational time series for the Southern Ocean, a weakening of the Southern Ocean anthropogenic carbon uptake has been controversially discussed. While atmospheric inversion approaches give results consistent with Le Quéré et al. (2007), the bulk of forward biogeochemical ocean models do not predict a decrease in Southern Ocean CO_2 uptake strength (Lovenduski et al., 2008; Lenton et al., 2013)."

Referee's comment: Page 1625, top: "Scenarios with Earth system models (advanced climate models, for a more detailed explanation see Sect. 3.2) reveal that the ocean sink may become less efficient in the future as higher cumulative CO_2 emissions counteract the general tendency for oceanic CO_2 uptake." *Why not simply state that the fraction of fossil fuel emissions absorbed by the ocean over the 21st century is projected to be lower for high emission BaU scenarios than stringent emission mitigation scenarios (Jones et al., J. Clim, 2013).*
Authors' response: Accepted.
Authors' changes in manuscript: The sentence was changed to: "Scenarios with Earth system models (advanced climate models, for a more detailed explanation see chapter 3.2) reveal that the fraction of fossil fuel emissions absorbed by the ocean over the 21st century is projected to be lower for high emission scenarios (business as usual scenarios) than stringent emission mitigation scenarios (Jones et al., 2013)."

Referee's comment: Page 1625, top: This is an incorrect statement and it should be deleted from the MS: "It, thus, remains to be explored what the ocean's ultimate uptake capacity for atmospheric CO_2 is, when it may be reached, and how until then the ocean may regulate the environmental effects of anthropogenic CO_2." *There is no ultimate uptake capacity for atm. CO_2. Perhaps you refer here to excess CO_2. A certain fraction of emission will always end up in the ocean on multi-century time scales and then excess atm. CO_2 will be further removed by $CaCO_3$ compensation and removed from the ocean-atm system by weathering-sediment (imbalances). See e.g. Archer et al, GBC, 1999.*
Authors' response: Accepted.
Authors' changes in manuscript: The sentence was deleted.

Referee's comment: Page 1626, line 11: delete: "such as Integrated Assessment Models" IAMs are not reservoir models.
Authors' response: Accepted.
Authors' changes in manuscript: The respective sentence was changed to: "Simplified climate models as, e.g., employed in Integrated Assessment Models (for simulations of economical developments under climatic change and for construction of typical future scenarios) are insufficient for this purpose as they do not account for internal feedbacks in the Earth system in a dynamical way (Jones et al., 2013)."

Referee's comment: Page 1627, line 18: delete "future".
Authors' response: Accepted.
Authors' changes in manuscript: Change made.

Referee's comment: Page 1630, line 7: replace "in designing correct future scenarios for" by "in".
Authors' response: Accepted.
Authors' changes in manuscript: Change made.

Referee's comment: Page 1631, line 23: Miss $\delta^{13}C$ here (e.g. Heimann and Maier Reimer, GBC, 199x, and refs therein, Resplandy et al., ..).
Authors' response: We do not understand the request of the referee. We did not find a corresponding reference "Heimann and Maier Reimer, GBC, 199x" and also do not know why the recent publications of L. Resplandy should possibly be taken into account here.
Authors' changes in manuscript: As we do not know what the referee means, we do not change the text.

Referee's comment: Page 1641, bottom: give a time frame, e.g. within the last 850,000 years.
Authors' response: Accepted.
Authors' changes in manuscript: We have added the 850,000 yr reference period from ice core measurements.

Referee's comment: Figure 4: Is this mid-depth pH change the result of anthropogenic invasion or of water mass changes in this area?
Authors' response: The area is a hot spot of anthropogenic carbon uptake (see, e.g., Sabine et al., 2004, and numerous numerical modelling studies). Of course, it cannot be excluded that parts of the difference can be explained by advection of older water masses, but then it would be the question where these water masses would come from in the northern North Atlantic. Another factor contributing to the lowering pH trend could be an increase in particle flux of organic matter. But this effect would mainly hold for the upper 1000 m. In view of the generally high anthropogenic CO_2 contributions to DIC in the area, we judge it likely that the effect indeed comes from the invasion of anthropogenic carbon.
Authors' changes in manuscript: No change of text.

Referee's comment: Figure 5: I am confused here and do not understand this figure. The title talks about modelled trend, but my impression is that the bars refer to the state of the system? What is the relation between the bars and the y-axis labels (atm. CO_2) and the x-axis label? What is a seasonal trend? . . . The figure caption definitely needs much more work.
Authors' response: Accepted.
Authors' changes in manuscript: The figure and figure caption have been corrected.