

**The non-steady-state  
oceanic CO<sub>2</sub> signal**

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# The non-steady-state oceanic CO<sub>2</sub> signal: its importance, magnitude and a novel way to detect it

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## Abstract

The ocean's role has been pivotal in modulating rising atmospheric CO<sub>2</sub> levels since the industrial revolution, sequestering over a quarter of all fossil-fuel derived CO<sub>2</sub> emissions. Net oceanic uptake of CO<sub>2</sub> has roughly doubled between the 1960's (~ 1 PgCyr<sup>-1</sup>) and 2000's (~ 2 PgCyr<sup>-1</sup>), with expectations it will continue to absorb even more CO<sub>2</sub> with rising future atmospheric CO<sub>2</sub> levels. However, recent CO<sub>2</sub> observational analyses along with numerous model predictions suggest the rate of oceanic CO<sub>2</sub> uptake is already slowing, largely as a result of a natural decadal-scale outgassing signal. This recent and unexpected CO<sub>2</sub> outgassing signal represents a paradigm-shift in our understanding of the oceans role in modulating atmospheric CO<sub>2</sub>. Current tracer-based estimates for the ocean storage of anthropogenic CO<sub>2</sub> assume the ocean circulation and biology is in steady state, thereby missing the new and potentially important "non-steady-state" CO<sub>2</sub> outgassing signal. By combining data-based techniques that assume the ocean is in steady-state, with techniques that constrain the net oceanic CO<sub>2</sub> uptake signal, we show how to extract the non-steady-state CO<sub>2</sub> signal from observations. Over the entire industrial era, the non-steady-state CO<sub>2</sub> outgassing signal (~ 13 ± 10 PgC) is estimated to represent about 9% of the total net CO<sub>2</sub> inventory change (~ 142 PgC). However between 1989 and 2007, the non-steady-state CO<sub>2</sub> outgassing signal (~ 6.3 PgC) has likely increased to be ~ 18% of net oceanic CO<sub>2</sub> storage over that period (~ 36 PgC), a level which cannot be ignored. The present uncertainty of our data-based techniques for oceanic CO<sub>2</sub> uptake limit our capacity to quantify the non-steady-state CO<sub>2</sub> signal, however with more data and better certainty estimates across a range of diverse methods, this important and growing CO<sub>2</sub> signal could be better constrained in the future.

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# 1 Introduction

## 1.1 The evolution of our understanding of the oceanic CO<sub>2</sub> sink

### 1.1.1 Our traditional steady-state view of the oceanic CO<sub>2</sub> cycle

For thousands of years before the onset of the industrial revolution (~1800 AD), carbon cycling between atmospheric, land and oceanic biospheres was in relative steady state. Although large gross exchanges of CO<sub>2</sub> were occurring annually between land, atmosphere and oceans, atmospheric CO<sub>2</sub> remained relatively constant at about  $280 \pm 5 \mu\text{atm}$  implying a steady-state carbon cycle (Etheridge et al., 1998). Humans, via the burning of fossil fuel carbon, have emitted about  $530 \text{ Pg}^1 \text{ C}$  into the atmosphere, perturbing atmospheric CO<sub>2</sub>.

Quantifying the flows, exchanges and storage of this anthropogenic CO<sub>2</sub> in the earth system has been a primary objective for the biogeochemical research community. Due to the heterogeneity in both space and time within the land carbon system, partitioning the global carbon storage between land and ocean has largely relied on our more certain understanding of the oceans storage of anthropogenic CO<sub>2</sub>. Fortunately, a number of different independent methods have allowed researchers to quantify the oceanic anthropogenic CO<sub>2</sub> sink (Quay et al., 1992; Gruber et al., 1996; Keeling et al., 1996; Gruber and Keeling, 2001; McNeil et al., 2003; Sabine et al., 2004; Waugh et al., 2006; Khatiwala et al., 2009). Fundamental to these estimates is the assumption that large-scale natural cycling of carbon through biological and circulation pathways have remained in steady state throughout the 20th century, with the anthropogenic perturbation acting passively on top of that large natural but unchanging “background” carbon cycle. This steady-state assumption was valid during most of the 20th century whereby any climate-related alterations to the oceanic anthropogenic CO<sub>2</sub> sink have been small in comparison to the large emissions signal itself (Sarmiento et al., 1998; Mearar and

<sup>1</sup>Peta-gram =  $10^{15}$  g.

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Hirst, 1999). Although the steady-state assumption may have been adequate for the 20th century due to the small impact on net oceanic CO<sub>2</sub> uptake, as discussed below, this is no longer the case for the 21st century.

### 1.1.2 The emerging and unexpected non-steady-state oceanic CO<sub>2</sub> signal

5 Over recent decades, oceanographers have observed large-scale decadal and longer time-scale trends in the ocean associated with biological changes, circulation changes and temperature-related solubility changes. The first observational research documenting large-scale decadal changes in the oceans circulation pathways were shown in the 1990s by temperature and salinity alterations in major ventilation pathways of the  
10 ocean (Wong et al., 1999). Since then the number of hydrographic measurements have increased, leading to a detailed understanding of the large-scale ocean warming trend (Levitus et al., 2000); as well as salinity changes associated with the amplification of the hydrological cycle (Durack and Wijffels, 2010; Helm et al., 2010). In the case of biogeochemistry, although nutrients were hypothesized to be changing (Pahlow and Riebesell, 2000), it was a suite of studies showing declining oxygen concentrations in various parts of the ocean (Helm et al., 2011; Matear et al., 2000; Emerson et al.,  
15 2001), which confirmed that large-scale circulation and/or biological changes were impacting biogeochemical cycles.

A new suite of climate models (Wetzel et al., 2005; Le Quéré et al., 2007; Lenton and  
20 Matear, 2007; Lovenduski et al., 2007; Matear and Lenton, 2008; Rodgers et al., 2008) driven with observed wind, heat and freshwater fluxes showed that beginning in the late 1980's, net CO<sub>2</sub> uptake started to level off. Oceanic CO<sub>2</sub> uptake was not increasing in the way it should be if the uptake was only a function of increasing atmospheric CO<sub>2</sub> levels and was quite aptly described by Sarmiento et al. (2010) as “somewhat of a surprise”.  
25

The ground-breaking study by Le Quéré et al. (2007) combined this modeling result with atmospheric CO<sub>2</sub> measurements over the Southern Ocean to postulate that the net oceanic CO<sub>2</sub> sink was leveling in response to an observed intensification of winds

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over the Southern Ocean that caused higher outgassing of naturally CO<sub>2</sub>-rich deep waters, partially offsetting a large anthropogenic CO<sub>2</sub> uptake signal. Le Quéré et al. (2007) and a follow-up model study by Lovenduski et al. (2008) showed that the unexpected CO<sub>2</sub> outgassing in the Southern Ocean to be up to 35 % of the anthropogenic CO<sub>2</sub> flux.

These non-steady-state ocean carbon changes create systematic biases in many tracer-based techniques that attempt to quantify the anthropogenic CO<sub>2</sub> storage in the ocean and these biases will continue to grow as the non-steady-state ocean evolves through the 21st century. This new development is critically important from an atmospheric CO<sub>2</sub> perspective, since non-steady-state changes in the ocean (at least the present outgassing) are a positive CO<sub>2</sub> feedback on atmospheric CO<sub>2</sub> levels. Recent evidence appears to show that the airborne fraction of CO<sub>2</sub> maybe increasing (Raupach et al., 2007; Le Quéré et al., 2009; Gloor et al., 2010; Knorr, 2012), and climate-driven oceanic CO<sub>2</sub> outgassing may be playing a role in those atmospheric CO<sub>2</sub> trends. The non-steady-state CO<sub>2</sub> signal is what we seek to detect, since without it, the ability to monitor and predict future atmospheric CO<sub>2</sub> levels will be impeded.

In this manuscript we seek to do two things. First, we present a decomposition of total oceanic CO<sub>2</sub> changes over time into natural and anthropogenic, steady and non-steady-state components. By doing this we seek to show the important difference between what often is referred to as anthropogenic CO<sub>2</sub> change in the ocean and the very different total net change in CO<sub>2</sub> in the ocean, since they are sometimes incorrectly used interchangeably. But foremost, this decomposition clarifies the differing steady and non-steady components in the oceanic CO<sub>2</sub> signal for budgetary purposes. Second, we investigate and present a simple data-based method to partition the time-evolving CO<sub>2</sub> sink into a steady-state and non-steady-state signal.

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## 2 Decomposing the time-evolution of CO<sub>2</sub> in the ocean

The net oceanic Dissolved Inorganic Carbon change ( $\Delta\text{DIC}_{\text{net}}$ ) between a period of time ( $t_1$  to  $t_2$ ) reflects changes in both natural and anthropogenic carbon dynamics:

$$[\Delta\text{DIC}_{\text{Net}}] = [\Delta\text{DIC}_{\text{Nat}}] + [\Delta\text{ACO}_2] \quad (1)$$

5 Natural changes in DIC ( $\Delta\text{DIC}_{\text{Nat}}$ ) occur through temperature, biological and ocean circulation changes via climate/ocean variability and change. On top of these natural DIC changes are also changes in DIC due to the oceanic uptake of anthropogenic CO<sub>2</sub> ( $\Delta\text{ACO}_2$ ).

10 If there were no net changes to the natural DIC concentrations in the ocean over given time period (i.e. steady state), then the time-evolving net change would be simply equal to the anthropogenic CO<sub>2</sub> uptake by the ocean (i.e.  $\Delta\text{DIC}_{\text{Net}} = \Delta\text{ACO}_2$ ).

15 In this steady-state world, anthropogenic CO<sub>2</sub> can be treated as a passive solubility tracer such as chlorofluorocarbons, whereby its uptake is driven solely by the atmosphere-ocean gradient, gas exchange and mixing without the need to account for biology or circulation changes. This definition of anthropogenic CO<sub>2</sub> makes tracer-based approaches very attractive (Gruber et al., 1996; McNeil et al., 2003; Waugh et al., 2006; Khatiwala et al., 2009). However as models have shown (Sarmiento et al., 1992; Matear and Hirst, 1999; Plattner et al., 2001; Friedlingstein et al., 2006) and observed CO<sub>2</sub> trends show (Le Quéré et al., 2007; Lenton et al., 2012), the ocean CO<sub>2</sub> system is not in steady-state, consistent with physical oceanic properties (temperature, salinity, oxygen) that have showed temporal change.

20 To account for this time-evolving behavior in CO<sub>2</sub> it is therefore necessary to decompose the time-evolution of anthropogenic CO<sub>2</sub> ( $\text{ACO}_2$ ) into its steady and non-steady state components as follows:

$$25 \quad [\Delta\text{ACO}_2] = \underbrace{[\Delta\text{ACO}_2]}_{\text{Steady-State}} + \underbrace{[\Delta\text{ACO}'_2]}_{\text{Non-Steady-State}} \quad (2)$$

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$\overline{\Delta\text{ACO}_2}$  is the traditional steady-state definition of anthropogenic  $\text{CO}_2$  used in the literature, whereby circulation and biological carbon changes remain constant with rising  $\text{CO}_2$  in the atmosphere.  $\Delta\text{ACO}'_2$  is the non-steady-state term identified by climate-change models whereby anthropogenic  $\text{CO}_2$  is modified by changes in circulation and/or biology from global warming (like stratification or warming's effect on  $\text{CO}_2$  solubility). This non-steady-state term has been explored in ocean biogeochemical models and shows a relatively small but growing non-steady state anthropogenic  $\text{CO}_2$  uptake signal by the end of this century (10–20%) (Sarmiento et al., 1992; Matear and Hirst, 1999; Plattner et al., 2001; Friedlingstein et al., 2006; Matear and Lenton, 2008).

Natural decadal variability and change alter ocean circulation and biology, therefore impacting the time-evolving DIC signal, requiring a separate set of equations:

$$[\Delta\text{DIC}_{\text{Nat}}] = \underbrace{[\overline{\Delta\text{DIC}_{\text{Nat}}}]_{\text{Steady-State}}}_{\text{Steady-State}} + \underbrace{[\Delta\text{DIC}'_{\text{Nat}}]}_{\text{Non-Steady-State}} \quad (3)$$

By definition,  $\overline{\Delta\text{DIC}_{\text{Nat}}}$  over time is 0, therefore the total changes in the natural carbon cycle ( $\Delta\text{DIC}_{\text{Nat}}$ ) is equal to the non-steady-state changes to the natural carbon cycle ( $\Delta\text{DIC}'_{\text{Nat}}$ ). For example,  $\Delta\text{DIC}'_{\text{Nat}}$  could be those carbon changes resulting from natural variability in the climate system like perhaps those driven from decadal-scale intensification of Southern Ocean winds, El-Nino/Southern Oscillation events, trends in remineralization stoichiometry or even simply time-evolving natural movements in circulation pathways over two-different periods of time.

What is important to remember here is that correcting for the natural DIC signal in the ocean from back-calculation techniques like  $\Delta\text{C}^*$  does not account for either the natural non-steady state signal ( $\Delta\text{DIC}'_{\text{Nat}}$ ) or the anthropogenic non-steady state signal ( $\Delta\text{ACO}'_2$ ).

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In summary, the time-evolving net DIC signal is the sum of three terms:

$$[\Delta\text{DIC}_{\text{Net}}] = \underbrace{[\overline{\Delta\text{ACO}_2}]}_{\text{Steady-State}} + \underbrace{[\Delta\text{ACO}'_2]}_{\text{Non-Steady-State}} + \underbrace{[\Delta\text{DIC}'_{\text{Nat}}]}_{\text{Non-Steady-State}} \quad (4)$$

Steady-State
Non-Steady-State
Non-Steady-State

Steady-State
Non-Steady-State
Non-Steady-State

Combined Non-Steady-State Signal

To simplify Eq. (4), we combine them into steady and non-steady state signals:

1.  $\overline{\Delta\text{ACO}_2}$ ; the steady-state change in ocean  $\text{CO}_2$  over time due to rising atmospheric  $\text{CO}_2$  in a unchanging ocean.
2.  $(\Delta\text{ACO}'_2 + \Delta\text{DIC}'_{\text{Nat}})$ ; the combined non-steady-state signal that incorporates how a changing ocean alters DIC in the ocean.

The most important term in Eq. (4) for atmospheric  $\text{CO}_2$  modulation is the combined net  $\text{CO}_2$  sink ( $\Delta\text{DIC}_{\text{Net}}$ ), therefore it is important to investigate and constrain the non-steady-state  $\text{CO}_2$  signal.

## 2.1 The anthropogenic non-steady-state signal

When introducing the  $\Delta\text{C}^*$  method, Gruber et al. (1996) were clear that their technique required a steady-state assumption. The global application of the  $\Delta\text{C}^*$  method was performed by Sabine et al. (2004) and it is important to understand that they estimated the steady-state oceanic anthropogenic  $\text{CO}_2$  inventory (i.e.  $\overline{\Delta\text{ACO}_2}$  in Eq. 4) and assumed it was equivalent to the total net change in oceanic  $\text{CO}_2$  (i.e.  $\Delta\text{DIC}_{\text{Net}}$ ). Between 1880 and 1994, Sabine et al. (2004) estimated an anthropogenic  $\text{CO}_2$  storage in the ocean of  $118 \pm 19 \text{ PgC}$ , which was recently increased to  $\sim 155 \text{ PgC}$  by 2010 based on a different steady-state tracer technique (Khatiwala et al., 2009).

In a commentary to Science, Ralph Keeling highlighted the important but missing non-steady-state anthropogenic signal (i.e.  $\Delta\text{ACO}'_2$ ) within the Sabine et al. estimate (Keeling, 2005). He noted that “Anthropogenic  $\text{CO}_2$ ” as it has been used traditionally

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is “an incomplete measure of the change in the ocean carbon content”. He further discussed the necessary steady-state assumption that oceanic tracer-based techniques require and made a first attempt to calculate the non-steady state anthropogenic  $\text{CO}_2$  change in association with recent climate change (i.e.  $\Delta\text{ACO}'_2$ ).

Using a box-diffusion model, Keeling (2005) estimated a 13 PgC outgassing of  $\text{CO}_2$  due to the thermodynamic effect of increasing  $\text{CO}_2$  in warmer oceanic surface waters up until 1994. With recent climate change until that point, upper ocean density stratification would also have already occurred. Although this stratification likely limits the subduction of water masses into the interior, Keeling (2005) postulated a stable upper ocean would also result in a more efficient (at least initially) biological drawdown of  $\text{CO}_2$  from the surface to the interior. Based on a suite of ocean model simulations, he estimated a net carbon drawdown of +6 PgC up until 1994. Different non-steady-state anthropogenic processes (ocean warming and stratification/biological export) partially offset each other, resulting in a final combined estimate of  $7 \pm 10$  PgC for  $\Delta\text{ACO}'_2$  from Keeling (2005).

This non-steady state term for oceanic anthropogenic  $\text{CO}_2$  is still less than 10% of the final anthropogenic  $\text{CO}_2$  inventory estimate and well within the total uncertainty of the technique ( $\pm 19$  PgC), as noted by Sabine et al., (2005). So although  $\Delta\text{ACO}'_2$  has been known to bias the data-based estimates of anthropogenic  $\text{CO}_2$  storage in the ocean, the bias is relatively small ( $\sim 10\%$ ).

However the most important term, at least over the past twenty years, in Eq. (4) is not  $\Delta\text{ACO}'_2$ , but  $\Delta\text{DIC}'_{\text{Nat}}$  as recent discoveries have shown us.

## 2.2 The natural non-steady-state signal

Le Quéré et al. (2007) showed that wind-speed intensification in the Southern Ocean was not only causing a large natural outgassing of  $\text{CO}_2$  as other models showed (Lovenduski et al., 2007; Lenton et al., 2009), but this trend was detected from atmospheric  $\text{CO}_2$  observations, although the atmospheric trend is still the subject of debate (Law et al., 2008). Arguably this Southern Ocean outgassing could be the fingerprint

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of climate change itself, but it could also be simply a natural decadal response and therefore would be mostly identified as a natural non-steady-state signal ( $\Delta\text{DIC}'_{\text{Nat}}$ ). In any case, partitioning this signal into  $\Delta\text{ACO}'_2$ , or  $\Delta\text{DIC}'_{\text{Nat}}$  is not important, since we combine both the natural and anthropogenic non-steady-state signals for simplicity.

5 Although research has progressed in understanding the potentially large natural non-steady-state  $\text{CO}_2$  signal in the Southern Ocean (Le Quéré et al., 2007; Lovenduski et al., 2008), how important is it on a global scale?

### 3 The total non-steady-state $\text{CO}_2$ signal and how we can detect it?

Sarmiento et al. (2010) synthesize a suite of different models from earlier studies (Wetzel et al., 2005; Mikaloff Fletcher et al., 2006; Le Quéré et al., 2007; Lovenduski et al., 2008; Rodgers et al., 2008) to estimate the global magnitude of the combined non-steady state oceanic  $\text{CO}_2$  signal between 1989 and 2007. For completeness, here we add the CSIRO biogeochemistry model (Matear and Lenton, 2008) to that suite of model results.

15 To illustrate the divergence of the time-evolving net oceanic  $\text{CO}_2$  uptake in recent decades, we combine the five different model predictions that use time-evolving NCEP atmospheric forcings and compare it to the expected steady-state uptake just from atmospheric  $\text{CO}_2$  increases alone (Fig. 1).

On average, as reported by Sarmiento et al. (2010), time-varying ocean models take up  $0.35 \text{ PgCyr}^{-1}$  less  $\text{CO}_2$  between 1989 and 2007 than they would have if ocean circulation and biogeochemistry had remained in steady-state. This would equate to a combined non-steady state  $\text{CO}_2$  inventory reduction of about  $6.3 \text{ PgC}$  between 1989 and 2007 in comparison to the total net  $\text{CO}_2$  inventory ( $\Delta\text{DIC}_{\text{Net}}$ ) of about  $36 \text{ PgC}$  from these models (Fig. 1). Each model differs in magnitude, but on average the magnitude of the combined non-steady state  $\text{CO}_2$  signal between 1989 and 2007 is about 18 % of the total carbon stored in the ocean over this time period.

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By combining the Keeling (2005) and Sarmiento et al. (2010) estimate, the total non-steady-state CO<sub>2</sub> outgassing since the industrial revolution would be about 13 ± 10 PgC, which is about ~ 10% of the steady-state anthropogenic CO<sub>2</sub> inventory (~ 155 PgC). Despite the relatively small estimated non-steady-state CO<sub>2</sub> signal over the entire industrial era, this non-steady-state signal has grown to be about 18% of net oceanic CO<sub>2</sub> storage between 1989–2007. Given the large magnitude of the non-steady-state CO<sub>2</sub> signal simulated in the models since 1989, can we use data-based methods to constrain it?

### 3.1 The power of combining more accurate, but diverse methods

In recent years there has been a proliferation of new tracer-based techniques to quantify decadal changes in oceanic CO<sub>2</sub> uptake (Hall et al., 2002, 2004; Gloor et al., 2003; Waugh et al., 2006; Khatiwala et al., 2009) that follow on from the C\* method (Gruber et al., 1996) and earlier attempts (Chen, 1982). Other techniques using atmospheric observations (i.e. CO<sub>2</sub> inversions or atmospheric O<sub>2</sub>/N<sub>2</sub> methods (Ciais et al., 1995; Keeling et al., 1996; Bousquet et al., 2000; Keeling and Garcia, 2002; Patra et al., 2005; Manning and Keeling, 2006) or a combination (Jacobson et al., 2007) are typically lumped together with those ocean-tracer methods with the assumption they are quantifying the same time-evolving oceanic CO<sub>2</sub> signal, which is not true.

Different data-based techniques constrain different oceanic CO<sub>2</sub> signals. To clarify what the variety of differing techniques are actually constraining, we list these independent data-based techniques and the signals they quantify in Table 1.

Most of the ocean-based tracer techniques quantify the steady-state anthropogenic CO<sub>2</sub> signal alone ( $\overline{\Delta\text{ACO}_2}$ ), while the atmospheric techniques quantify the total time-evolving net CO<sub>2</sub> oceanic signal ( $\Delta\text{DIC}_{\text{Net}}$ ). Although each method has inherent uncertainties and biases, there is powerful information in treating them as independent, whereby their difference theoretically should constrain the combined non-steady-state response.

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For an ocean that is changing with climate change and decadal variability, combining steady-state methods with total net methods could provide a powerful way to quantify how the oceanic CO<sub>2</sub> sink is actually evolving. For example, the total net CO<sub>2</sub> sink (i.e.  $\Delta\text{DIC}_{\text{Net}}$  in Eq. 4) is best captured from two different techniques: the O<sub>2</sub>/N<sub>2</sub> atmospheric technique and surface ocean pCO<sub>2</sub> climatologies. On the other hand, CFC-based and ocean inversion methods are the probably best techniques to capture the steady-state anthropogenic CO<sub>2</sub> inventory between two different periods of time. In theory, by comparing the results of these different techniques should produce a testable signal equivalent to the non-steady-state oceanic CO<sub>2</sub> change.

The expected steady-state anthropogenic CO<sub>2</sub> uptake in the 1990's from oceanic inversions and CFC's is 2–2.2 PgCyr<sup>-1</sup> (see Table 2). The total net CO<sub>2</sub> uptake for the 1990s from oceanic pCO<sub>2</sub> climatology and atmospheric O<sub>2</sub>/N<sub>2</sub> methods is 1.7–1.9 PgCyr<sup>-1</sup>. Solving equation 4 implies that the difference between these constraints is the non-steady-state CO<sub>2</sub> signal, implying an outgassing of 0.1 to 0.5 PgCyr<sup>-1</sup> (Table 2).

The uncertainty estimates (approximately  $\pm 0.4$ – $0.6$  PgCyr<sup>-1</sup>) for each of these techniques at present makes the non-steady-state signal statistically insignificant. This example using multiple methodologies however, illustrates the power of the approach and challenges us to obtain better observations to reduce these uncertainties amongst the suite of different oceanic CO<sub>2</sub> uptake data-based techniques. It is interesting however, that models are suggesting a non-steady-state CO<sub>2</sub> outgassing ( $\sim +0.4$  PgCyr<sup>-1</sup>), which is nearing the uncertainty limits for these observational constraints, implying that into the future, with greater certainty, such a non-steady-state CO<sub>2</sub> signal could be quantified.

We formulate a revised oceanic carbon budget for the 1989–2007 period (Fig. 2) that takes into account the  $\sim +0.4$  PgCyr<sup>-1</sup> non-steady-state CO<sub>2</sub> outgassing predicted by both the models and somewhat tentatively by the multi-methodological constraint illustrated here.

## 4 Challenges to reducing uncertainty

The key limitation to this multi-methodological approach today is the current large uncertainty of the different data-based techniques to quantify anthropogenic CO<sub>2</sub> uptake in the ocean. It is not necessarily important which combination of technique is used, but rather we have certainty over its assumptions and application.

Many of the techniques suffer from a lack of measurements, which can be rectified in the future. For example, the *p*CO<sub>2</sub> database, although good coverage exists in the Northern Hemisphere, complex regimes like the equatorial Pacific and Southern Oceans have large gaps in coverage. However with autonomous CO<sub>2</sub> measurements increasing, this will change.

One complication with *p*CO<sub>2</sub> climatologies is associated with the natural outgassing of carbon that enters the ocean via rivers and estuaries. This is an uncertain but necessary constraint for the oceanic carbon budget when using *p*CO<sub>2</sub> climatologies or oceanic inversions (Jacobson et al., 2007; Gruber et al., 2009). From an atmospheric perspective, this riverine CO<sub>2</sub> outgassing is a steady-state signal since the coinciding uptake of CO<sub>2</sub> occurs on land through biomass production. However, from the “raw” *p*CO<sub>2</sub> climatologies, the riverine CO<sub>2</sub> outgassing needs to be added to the final global ocean estimate. This riverine CO<sub>2</sub> outgassing is estimated to be +0.45 PgCyr<sup>-1</sup> (Jacobson et al., 2007) with a 50 % uncertainty. The magnitude of this riverine CO<sub>2</sub> outgassing would dampen this techniques ability to detect the non-steady-state CO<sub>2</sub> signal, unless that is constrained to a much higher accuracy.

Other techniques like the modern application of CFC-tracers (Waugh et al., 2006; Khatiwala et al., 2009) are not as data-limited and provide the most accurate way of constraining the steady-state anthropogenic CO<sub>2</sub> signal over decadal time-scales. With temporal CFC or tracer measurements, this technique could go one-step further to constrain the non-steady-state circulation based changes to anthropogenic CO<sub>2</sub>, although any changes to the biological pump or natural carbon would be missed.

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In defining the oceanic non-steady-state CO<sub>2</sub> signal, the coastal ocean cannot go without some discussion of its importance. The variability of CO<sub>2</sub> within this region, which covers 8 % of the ocean, is orders of magnitude greater than that in most of the open-ocean (Borges et al., 2005). Given the coastal ocean has a direct interface with anthropogenic changes from land with respect to both nutrients and carbon (Gypens et al., 2009), coupled with an immense natural CO<sub>2</sub> variability, we would speculate that a global non-steady-state CO<sub>2</sub> signal from the coastal ocean may be significant into the future.

## 5 Conclusions

Here we decompose the time-evolution of net CO<sub>2</sub> changes in the ocean to clarify the notion of “anthropogenic” CO<sub>2</sub> uptake. Traditionally, oceanic tracer-based techniques have constrained net oceanic storage of CO<sub>2</sub> associated with elevated CO<sub>2</sub> concentrations in the atmosphere under the assumption of a steady-state ocean carbon cycle. Both climate-change and decadal changes to the oceanic CO<sub>2</sub> cycle shown recently have clearly marked a new era of non-steady-state conditions for CO<sub>2</sub> that was already known from other biogeochemical parameters like oxygen. This global non-steady-state CO<sub>2</sub> signal is estimated to have outgassed about 6.3 PgC of CO<sub>2</sub> (or ~ 3 ppm to the atmosphere) between 1989 and 2007, which is ~ 18 % of the net oceanic CO<sub>2</sub> uptake rate estimated from models.

After illustrating the different components of the time-evolving oceanic CO<sub>2</sub> sink, we present a simple concept to estimate the non-steady-state oceanic CO<sub>2</sub> signal and determine the net change in carbon stored in the ocean. With a multi-methodological budget approach, we estimate a 0.1–0.5 PgCyr<sup>-1</sup> outgassing over the last two decades, however, the uncertainty across the suite of different data-based techniques is too large at present (~ 0.4–0.6 PgCyr<sup>-1</sup>) to provide a significant non-zero estimate of the non-steady-state signal.

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Although the non-steady-state CO<sub>2</sub> signal currently provides a positive feedback to atmospheric CO<sub>2</sub> levels, the future direction and magnitude of the signal is not clear, since some models suggest that recent Southern Ocean outgassing will eventually reverse in the future and absorb greater CO<sub>2</sub> than expected from steady-state conditions (Zickfeld et al., 2008).

In the end, atmospheric CO<sub>2</sub> levels only change via the total time-evolving CO<sub>2</sub> changes in the ocean, which based on recent evidence is now entering a new non-steady-state mode. Given this paradigm shift, the challenge for the observational community is to reduce the uncertainty across a suite of independent data-based techniques to enable the clear separation between the net storage of CO<sub>2</sub> in the ocean and the steady-state anthropogenic CO<sub>2</sub> signal on decadal timeframes. By embracing more accurate and diverse techniques we can better help detect how a changing ocean is modifying rising atmospheric CO<sub>2</sub> levels.

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**Table 1.** Data-based techniques to quantify “anthropogenic” CO<sub>2</sub> storage in the ocean.

	Component constrained in Eq. (4)
Ocean data-based techniques	
Ocean DIC MLR on repeat cruises (Slansky et al., 1996; McNeil et al., 2001; Matear and McNeil, 2003; Bates et al., 2006)	? (Not easily identified)
Direct DIC difference from repeat cruises	$\Delta\text{DIC}_{\text{Net}}$
CFC's, TTDs, C* (Gruber et al., 1996; McNeil et al., 2003; Waugh et al., 2006; Khatiwala et al., 2009)	$\Delta\text{ACO}_2$
Ocean-based CO <sub>2</sub> Inversions (Gloor et al., 2003; Mikaloff Fletcher et al., 2007; Gruber et al., 2009)	$\Delta\text{ACO}_2$
Oceanic $\Delta p\text{CO}_2$ climatology (Takahashi et al., 2002)	$\Delta\text{DIC}_{\text{Net}}$
Atmospheric data-based techniques	
CO <sub>2</sub> inversions (Ciais et al., 1995; Francey et al., 1995; Bousquet et al., 2000; Patra et al., 2005; Le Quéré et al., 2007)	$\Delta\text{DIC}_{\text{Net}}$
O <sub>2</sub> /N <sub>2</sub> measurements (Keeling et al., 1996; Battle et al., 2000; Keeling and Garcia, 2002; Bender et al., 2005; Manning and Keeling, 2006)	$\Delta\text{DIC}_{\text{Net}}$

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**Table 2.** Different estimates for the time-evolving oceanic CO<sub>2</sub> uptake between 1990–1999 assuming no uncertainty.

Data-based technique	Steady-state anthropogenic CO <sub>2</sub> uptake $\Delta\text{ACO}_2$	Total net CO <sub>2</sub> uptake ( $\Delta\text{DIC}_{\text{Net}}$ )	Non steady state CO <sub>2</sub> uptake (i.e. $\Delta\text{DIC}_{\text{Net}} - \Delta\text{ACO}_2$ )
Ocean inversion (Gruber et al., 2009)	$-2.2 \text{ PgCyr}^{-1}$		
CFC's (McNeil et al., 2003; Khatiwala et al., 2009)	$-2.0 \text{ PgCyr}^{-1}$		
Oceanic pCO <sub>2</sub> climatology		$-1.9 \text{ PgCyr}^{-1}$	
Atmospheric O <sub>2</sub> /N <sub>2</sub> (Keeling and Garcia, 2002; Manning and Keeling, 2006; Bender et al., 2006)		$-1.7 \text{ to } 1.9 \text{ PgCyr}^{-1}$	
Multi-technique difference as diagnosed here			$+0.1 \text{ to } +0.5 \text{ PgCyr}^{-1}$
Suite of global climate models with recent climate variability (Sarmiento et al., 2010)	$-1.97 \text{ PgCyr}^{-1}$	$-1.46 \text{ PgCyr}^{-1}$	$+0.35 \text{ PgCyr}^{-1}$

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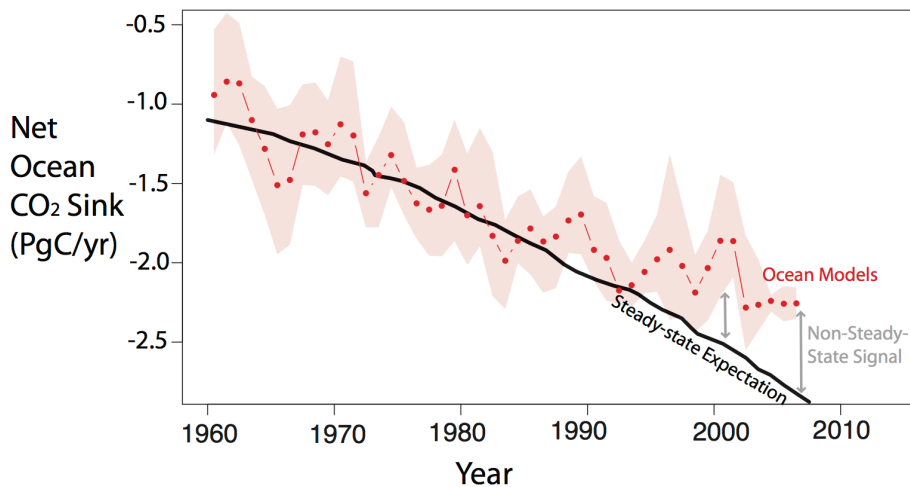


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## The non-steady-state oceanic CO<sub>2</sub> signal

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**Fig. 1.** Illustration of the time-evolving net CO<sub>2</sub> uptake from the ocean since 1970 comparing the expected steady-state uptake that assumes constant circulation and biology (black line) with the mean net uptake from five different ocean models that include time-evolving natural forcings (Wetzel et al., 2005; Le Quéré et al., 2007; Lovenduski et al., 2007; Matear and Lenton, 2008; Rodgers et al., 2008). We include the results from the hindcast CSIRO climate model from Matear and Lenton (2008) and the suite of model projections documented in Sarmiento et al. (2010). The expected steady-state CO<sub>2</sub> uptake is taken from Mikaloff-Fletcher et al. (2006) as documented by Sarmiento et al. (2010), normalized to the decade of 1970–80. The average net CO<sub>2</sub> uptake (red dots) was taken as a combined mean between the five different models with minimum and maximum bounds shaded in light red. This is an illustrative figure used to demonstrate the time-evolving changes in the various CO<sub>2</sub> signals within the ocean as described in Eq. (4). The integrated dotted red curve is the total net CO<sub>2</sub> uptake by the ocean ( $\Delta\text{DIC}_{\text{Net}}$ ), which is about 36 PgC between 1989 and 2007. The black curve is the expected steady-state CO<sub>2</sub> uptake or what has been historically called Anthropogenic CO<sub>2</sub> Uptake ( $\Delta\text{ACO}_2$ ), which is about 42 PgC between 1989 and 2007. The difference between these curves is therefore the combined non-steady-state CO<sub>2</sub> loss to the atmosphere ( $\Delta\text{ACO}_2' + \Delta\text{DIC}'_{\text{Nat}}$ ), which comes to about 6.3 PgC outgassing to the atmosphere between 1989 and 2007 (see Table 2 of Sarmiento et al., 2010).

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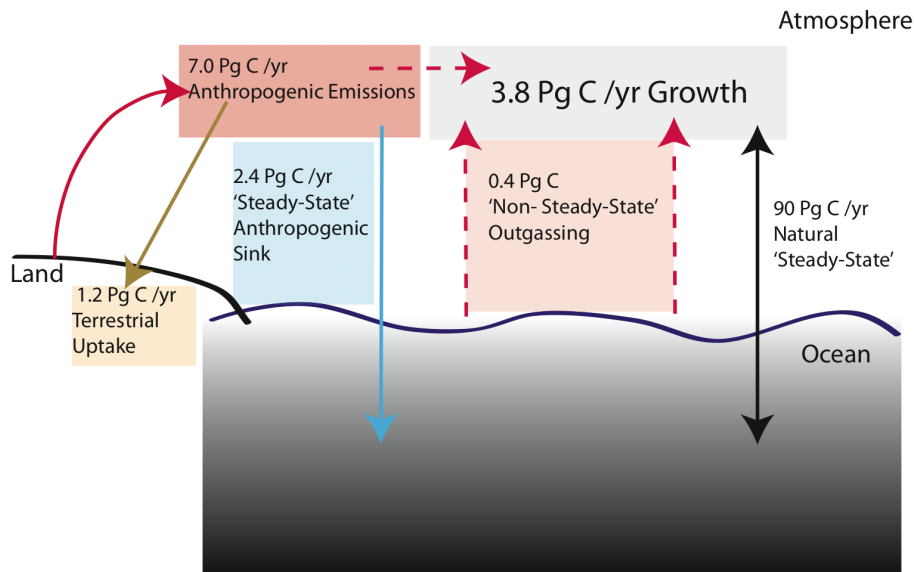
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**Fig. 2.** Estimated non-steady-state Global Carbon Budget for 1989–2007 combining oceanic steady-state observational estimates with the oceanic non-steady-state signal as diagnosed from data and models in this study.

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