The non-steady state oceanic CO$_2$ signal: its importance, magnitude and a novel way to detect it

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Abstract. The role of the ocean has been pivotal in modulating rising atmospheric CO$_2$ levels since the industrial revolution, sequestering nearly half of all fossil-fuel derived CO$_2$ emissions. Net oceanic uptake of CO$_2$ has roughly doubled between the 1960s ($\sim$1 Pg C yr$^{-1}$) and 2000s ($\sim$2 Pg C yr$^{-1}$), with expectations that it will continue to absorb even more CO$_2$ with rising future atmospheric CO$_2$ levels. However, recent CO$_2$ observational analyses along with numerous model predictions suggest the rate of oceanic CO$_2$ uptake is already slowing, largely as a result of a natural decadal-scale outgassing signal. This recent CO$_2$ outgassing signal represents a significant shift in our understanding of the oceans role in modulating atmospheric CO$_2$. Current tracer-based estimates for the ocean storage of anthropogenic CO$_2$ assume the ocean circulation and biology is in steady state, thereby missing the new and potentially important “non-steady state” CO$_2$ outgassing signal. By combining data-based techniques that assume the ocean is in a steady state, with techniques that constrain the net oceanic CO$_2$ uptake signal, we show how to extract the non-steady state CO$_2$ signal from observations. Over the entire industrial era, the non-steady state CO$_2$ outgassing signal ($\sim$13 ± 10Pg C) is estimated to represent about 9 % of the total net CO$_2$ inventory change ($\sim$142 Pg C). However, between 1989 and 2007, the non-steady state CO$_2$ outgassing signal ($\sim$6.3Pg C) has likely increased to be $\sim$18 % of net oceanic CO$_2$ storage over that period ($\sim$36 Pg C). The present uncertainty of our database techniques for oceanic CO$_2$ uptake limit our capacity to quantify the non-steady state CO$_2$ signal, however with more data and better certainty estimates across a range of diverse methods, this important and growing CO$_2$ signal could be better constrained in the future.

1 Introduction

1.1 The evolution of our understanding of the oceanic CO$_2$ sink

1.1.1 Our traditional steady state view of the oceanic CO$_2$ cycle

For thousands of years before the onset of the industrial revolution ($\sim$1800 AD), carbon cycling between atmospheric, land and oceanic biospheres was in relative steady state. Although large gross exchanges of CO$_2$ were occurring annually between land, atmosphere and oceans, atmospheric CO$_2$ remained relatively constant at about 280 ± 5µatm, implying a steady state carbon cycle (Etheridge et al., 1998). Humans, via the burning of fossil fuel carbon, have emitted about 530 Pg C into the atmosphere, perturbing atmospheric CO$_2$.

Quantifying the flows, exchanges and storage of this anthropogenic CO$_2$ 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$_2$. Fortunately, a number of different independent methods have allowed researchers to quantify the oceanic anthropogenic CO$_2$ 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\textsubscript{2} sink have been small in comparison to the large emissions signal itself (Sarmiento et al., 1998; Matear and Hirst, 1999). Although the steady state assumption may have been adequate for the 20th century due to the small impact on net oceanic CO\textsubscript{2} uptake, as discussed below, this is no longer the case for the 21st century.

### 1.1.2 The emerging non-steady state oceanic CO\textsubscript{2} signal

Over recent decades, oceanographers have observed large-scale decadal and longer timescale 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 with temperature and salinity alterations in major ventilation pathways of the 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 and/or changing oxygen concentrations in various parts of the ocean (Emerson et al., 2001; Matear et al., 2000; Whitney et al., 2007; Stendardo and Gruber, 2012), 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; 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 1980s, net CO\textsubscript{2} uptake started to level off. Oceanic CO\textsubscript{2} uptake was not increasing in the way it should be if the uptake was only a function of increasing atmospheric CO\textsubscript{2} levels and was quite aptly described by Sarmiento et al. (2010) as “somewhat of a surprise”.

Le Quéré et al. (2007) combined this modeling result with atmospheric CO\textsubscript{2} measurements over the Southern Ocean to postulate that the net oceanic CO\textsubscript{2} sink was leveling in response to an observed intensification of winds over the Southern Ocean that caused higher outgassing of naturally CO\textsubscript{2}-rich deep waters, partially offsetting a large anthropogenic CO\textsubscript{2} uptake signal. Le Quéré et al. (2007) and a follow-up model study by Lovenduski et al. (2008) showed that the CO\textsubscript{2} outgassing in the Southern Ocean to be up to 35\% of the anthropogenic CO\textsubscript{2} flux.

These non-steady state ocean carbon changes create systematic biases in many tracer-based techniques that attempt to quantify the anthropogenic CO\textsubscript{2} 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\textsubscript{2} perspective, since non-steady state changes in the ocean (at least the present outgassing) are a positive CO\textsubscript{2} feedback on atmospheric CO\textsubscript{2} levels. Recent evidence appears to show that the airborne fraction of CO\textsubscript{2} (the fraction of anthropogenic emissions which remain in the atmosphere) maybe increasing (Raupach et al., 2007; Le Quéré et al., 2009; Gloor et al., 2010; Knorr, 2012), and climate-driven oceanic CO\textsubscript{2} outgassing may be playing a role in those atmospheric CO\textsubscript{2} trends. The non-steady state CO\textsubscript{2} signal is what we seek to detect, since without it, the ability to monitor and predict future atmospheric CO\textsubscript{2} levels will be impeded.

In this manuscript we seek to do two things. First, we present a decomposition of total oceanic CO\textsubscript{2} 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\textsubscript{2} change in the ocean and the very different total net change in CO\textsubscript{2} in the ocean, since they are sometimes incorrectly used interchangeability. But foremost, this decomposition clarifies the differing steady and non-steady components in the oceanic CO\textsubscript{2} signal for budgetary purposes. Second, we investigate and present a simple data-based method to partition the time-evolving CO\textsubscript{2} sink into a steady state and non-steady state signal.

### 2 Decomposing the time evolution of CO\textsubscript{2} in the Ocean

The net oceanic dissolved inorganic carbon change (\(\Delta\text{DIC}_{\text{Net}}\)) between a period of time (\(t1\) to \(t2\)) reflects changes in both natural and anthropogenic carbon dynamics:

\[
\Delta\text{DIC}_{\text{Net}} = [\Delta\text{DIC}_{\text{Nat}}] + [\Delta\text{ACO}_2].
\]

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\textsubscript{2} (\(\Delta\text{ACO}_2\)).

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

In this steady state world, anthropogenic CO\textsubscript{2} can be treated as a passive solubility tracer such as chlorofluorocarbons (CFCs), whereby its uptake is driven solely by the
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atmosphere–ocean gradient, gas exchange and mixing without the need to account for biology or circulation changes. This definition of anthropogenic CO$_2$ makes tracer-based approaches very attractive. However, as models have shown (Joos et al., 1999; Friedlingstein et al., 2006; Matear and Hirst, 1999; Sarmiento et al., 1998) and observed CO$_2$ trends show (Le Quéré et al., 2007; Lenton et al., 2012), the ocean CO$_2$ system is not in steady state, consistent with physical oceanic properties (temperature, salinity, oxygen) that have showed temporal change.

To account for this time-evolving behavior in CO$_2$ it is therefore necessary to decompose the time evolution of anthropogenic CO$_2$ (ACO$_2$) into its steady and non-steady state components as follows:

$$\Delta ACO_2 = \Delta ACO_2^\text{Steady State} + \Delta ACO_2^\text{Non-Steady State}$$  \hspace{1cm} (2)

where $\Delta ACO_2$ is the traditional steady state definition of anthropogenic CO$_2$ used in the literature, whereby circulation and biological carbon changes remain constant with rising CO$_2$ in the atmosphere. $\Delta ACO_2^\text{Steady State}$ is the non-steady state term identified by climate change models whereby anthropogenic CO$_2$ is modified by changes in circulation and/or biology from global warming (like stratification or warming’s effect on 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 CO$_2$ uptake signal by the end of this century (10–20%) (Sarmiento et al., 1998; 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:

$$\Delta DIC_{Nat} = \Delta DIC_{Nat}^\text{Steady State} + \Delta DIC_{Nat}^\text{Non-Steady State}$$  \hspace{1cm} (3)

By definition, globally $\Delta DIC_{Nat}$ over time is 0, therefore the total changes in the natural carbon cycle ($\Delta DIC_{Nat}$) are equal to the non-steady state changes to the natural carbon cycle ($\Delta DIC_{Nat}^\text{Non-Steady State}$). For example, $\Delta DIC_{Nat}$ could be those carbon changes resulting from natural variability in the climate system, such as perhaps those driven from decadal-scale intensification of Southern Ocean winds, El Niño–Southern Oscillation events, trends in remineralization stoichiometry or even regional time-evolving 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, such as $\Delta C^*$, does not account for either the natural non-steady state signal ($\Delta DIC_{Nat}^\text{Non-Steady State}$) or the anthropogenic non-steady state signal ($\Delta ACO_2^\text{Non-Steady State}$).

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

$$[\Delta DIC_{Net}] = \frac{[\Delta ACO_2]}{\text{Steady State}} + \frac{[\Delta ACO_2^\text{Non-Steady State}]}{\text{Non-Steady State}} + \frac{[\Delta DIC_{Nat}^\text{Non-Steady State}]}{\text{Non-Steady State}}.$$  \hspace{1cm} (4)

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

1. $\Delta ACO_2$: the steady state change in ocean CO$_2$ over time due to rising atmospheric CO$_2$ in a unchanging ocean.

2. $(\Delta ACO_2^\text{Non-Steady State} + \Delta DIC_{Nat}^\text{Non-Steady State})$: 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 CO$_2$ modulation is the combined net CO$_2$ sink ($\Delta DIC_{Net}$); therefore, it is important to investigate and constrain the non-steady state CO$_2$ signal.

2.1 The anthropogenic non-steady state signal

When introducing the $\Delta C^*$ method, Gruber et al. (1996) were clear that their technique required a steady state assumption. The global application of the $\Delta C^*$ method was performed by Sabine et al. (2004) and it is important to understand that they estimated the steady state oceanic anthropogenic CO$_2$ inventory (i.e. $\Delta ACO_2$ in Eq. 4) and assumed it was equivalent to the total net change in oceanic CO$_2$ (i.e. $\Delta DIC_{Net}$). Between 1880 and 1994, Sabine et al. (2004) estimated an anthropogenic CO$_2$ storage in the ocean of 118 ± 19 Pg C, which was recently increased to ~155 Pg C 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. $\Delta ACO_2$) within the Sabine et al estimate (Keeling, 2005). He noted that “Anthropogenic CO$_2$” as it has been used traditionally 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 CO$_2$ change in association with recent climate change (i.e. $\Delta ACO_2$).

Using a box-diffusion model, Keeling (2005) estimated a 13 Pg C outgassing of CO$_2$ due to the thermodynamic effect of increasing CO$_2$ in warmer oceanic surface waters. 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 CO$_2$ from
the surface to the interior. Based on a suite of ocean model simulations, he estimated a net carbon drawdown of +6 Pg C 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 ± 10 Pg C for \( \Delta ACO_2 \) from Keeling (2005).

This non-steady state term for oceanic anthropogenic CO\(_2\) is still less than 10% of the final anthropogenic CO\(_2\) inventory estimate and well within the total uncertainty of the technique (±19 Pg C), as noted by Sabine and Gruber (2005). So although \( \Delta ACO_2 \) has been known to bias the data-based estimates of anthropogenic CO\(_2\) storage in the ocean, the bias is relatively small (~10%).

However, the most important term, at least over the past twenty years, in Eq. (4) is not \( \Delta ACO_2 \) but \( \Delta DIC_{\text{Nat}} \) 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 CO\(_2\) as other models showed (Lovenduski et al., 2007; Lenton et al., 2009), but this trend was detected from atmospheric 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 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 \( \Delta DIC_{\text{Nat}} \). In any case, partitioning this signal into \( \Delta ACO_2 \), or \( \Delta DIC_{\text{Nat}} \) is not important, since we combine both the natural and anthropogenic non-steady state signals for simplicity.

It is important to emphasize that the above decomposition is relevant for global changes in ocean carbon inventories that can then translated into global air–sea CO\(_2\) fluxes. On a regional basis however, translating carbon inventory changes to regional air–sea CO\(_2\) fluxes could be largely independent. For example a lateral change in the position of a gyre would translate into large changes in \( \Delta DIC_{\text{Nat}} \) within the ocean interior without a corresponding air–sea CO\(_2\) flux signature.

Although research has progressed in understanding the regional natural non-steady state CO\(_2\) signal (Le Quéré et al., 2007; Lovenduski et al., 2007; Levine et al., 2008; Goodkin et al., 2011), how important is it on a global scale?

### 3 Can we detect the global non-steady state CO\(_2\) signal?

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 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.

To illustrate the divergence of the time-evolving net oceanic 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 CO\(_2\) increases alone (Fig. 1).

On average, as reported by Sarmiento et al. (2010), time-varying ocean models take up 0.35 Pg C yr\(^{-1}\) less 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 CO\(_2\) inventory reduction of about 6.3 Pg C between 1989 and 2007 in comparison to the total net CO\(_2\) inventory \( \Delta DIC_{\text{Net}} \) of about 36 Pg C from these models (Fig. 1). Each model differs in magnitude, but on average the magnitude of the combined non-steady state CO\(_2\) signal between 1989 and 2007 is about 18% of the total carbon stored in the ocean over this time period.

By combining the Keeling (2005) and Sarmiento et al. (2010) estimate, the total non-steady state CO\(_2\) outgassing since the industrial revolution would be about 13 ± 10 Pg C, which is about ~10% of the steady state anthropogenic CO\(_2\) inventory (~155 Pg C). It is important to emphasize that the Keeling (2005) estimate was made using the Sabine et al. (2004) estimate for anthropogenic CO\(_2\) sink up until 1994. The Sarmiento et al. (2010) model results were made from 1990 onwards, so that by combining these estimates results in a four year overlap. However, if we take the models from Sarmiento et al. (2010) as a gauge, the largest signal in the non-steady state comes after the mid-1990s, so this overlap would probably result in a small bias (~0.5 Pg C of the estimate).

Despite the relatively small estimated non-steady state CO\(_2\) signal over the entire industrial era, this non-steady state signal has grown to be about 18% of net oceanic CO\(_2\) storage between 1989–2007. Given the large magnitude of the non-steady state CO\(_2\) signal simulated in the models since 1989, can we use data-based methods to constrain it?

### 3.1 The multi-method approach to estimate the non-steady state signal

In recent years there has been a proliferation of new tracer-based techniques to quantify decadal changes in oceanic CO\(_2\) uptake (Hall et al., 2002, 2004; Gloer 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\(_2\) inversions or atmospheric O\(_2/N_2\) 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 CO₂ signal, which is not true.

Different data-based techniques constrain different oceanic CO₂ signals. Or to put it another way, each data-based technique has a different sensitivity towards capturing the non-steady state signal in the ocean. For example, the C* method uses actual carbon measurements, that to some degree will contain some but not all of the non-steady state signal. The multi-method approach here requires the use of methods that capture either the steady-state-state signal or the net oceanic CO₂ signals with the most certainty. A technique which partially captures both the steady state and non-steady state signals is not a method that is helpful when applying the multi-method approach here. To clarify what signal differing data-based techniques are actually constraining, we list them in Table 1.

Most of the ocean-based tracer techniques quantify the steady state anthropogenic CO₂ signal alone (ΔACO₂), while the atmospheric techniques quantify the total time-evolving net CO₂ oceanic signal (ΔDIC_Net). Although each method has inherent uncertainties and biases, there is powerful information in treating them as independent, whereby there difference theoretically should constrain the combined non-steady state response.

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 CO₂ sink is actually evolving. For example, the total net CO₂ sink (i.e. ΔDIC_Net in Eq. 4) is best captured from two different techniques: the O₂/N₂ atmospheric technique and surface ocean pCO₂ climatologies. On the other hand, CFC-based and ocean inversion methods are the most accurate techniques to capture the steady state anthropogenic CO₂ 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 CO₂ change.

The expected steady state anthropogenic CO₂ uptake in the 1990s from oceanic inversions and CFCs is 2–2.2 Pg C yr⁻¹ (see Table 2). The total net CO₂ uptake for the 1990s from oceanic pCO₂ climatology and atmospheric O₂/N₂ methods is 1.7–1.9 Pg C yr⁻¹. Solving equation 4 implies that the difference between these constraints is the non-steady state CO₂ signal, implying an outgassing of 0.1 to
0.5 Pg C yr\(^{-1}\) (Table 2). If we assume uncertainties are independent between the different methods (Table 2) within a range of ±0.4–0.6 Pg C yr\(^{-1}\), a simple propagation of the uncertainties within a combination of any two of these methods implies a total uncertainty on the multi-method approach of about ±0.6–0.8 Pg C yr\(^{-1}\). This uncertainty at present makes the non-steady state signal as estimated through the multi-method approach statistically insignificant given a signal of 0.1 to 0.5 Pg C yr\(^{-1}\) (Table 2).

The application of the multi-method approach, however, illustrates the potential benefit of using such an approach but also challenges us to obtain better observations to reduce these uncertainties amongst the suite of different oceanic CO\(_2\) uptake data-based techniques. It is interesting, however, that models are suggesting a non-steady state CO\(_2\) outgassing (~+0.4 Pg C yr\(^{-1}\)) which is nearing the uncertainty limits for a multi-method approach (i.e. ±0.6–0.8 Pg C yr\(^{-1}\)), implying that into the future, with greater certainty, such a non-steady state CO\(_2\) signal could become observationally statistically significant.

We formulate a revised oceanic carbon budget for the 1989–2007 period (Fig. 2) that takes into account the ~+0.4 Pg C yr\(^{-1}\) non-steady state CO\(_2\) 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\(_2\) 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 pCO\(_2\) 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\(_2\) measurements increasing, this will change.

One complication with pCO\(_2\) 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 pCO\(_2\) climatologies or oceanic inversions (Jacobson et al., 2007; Gruber et al., 2009). From an atmospheric perspective, this riverine CO\(_2\) outgassing is a steady state signal since the coinciding uptake of CO\(_2\) occurs on land through biomass production. However, from the “raw” pCO\(_2\) climatologies, the riverine CO\(_2\) outgassing needs to be added to the final global ocean estimate. This riverine CO\(_2\) outgassing is estimated to be +0.45 Pg C yr\(^{-1}\) (Jacobson et al., 2007) with a 50 % uncertainty. The magnitude of this riverine CO\(_2\) outgassing would dampen this techniques ability to detect the non-steady state CO\(_2\) 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\(_2\) signal over decadal timescales. With temporal CFC or tracer measurements, this

<table>
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<tr>
<th>Table 1. Data-based techniques to quantify “anthropogenic” CO(_2) storage in the ocean.</th>
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<tr>
<td><strong>Ocean Data-based Techniques</strong></td>
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<tr>
<td>Ocean DIC MLR on repeat cruises (Slansky et al., 1997; McNeil et al., 2001; Matear and McNeil, 2003; Bates et al., 2006)</td>
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<tr>
<td>Direct DIC difference from repeat cruises</td>
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<tr>
<td>CFCs, TTDs, C(^*) (Gruber et al., 1996; McNeil et al., 2003; Waugh et al., 2006; Khatiwala et al., 2009)</td>
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<tr>
<td>Ocean-based CO(_2) Inversions (Gloor et al., 2003; Mikaloff-Fletcher et al., 2007; Gruber et al., 2009)</td>
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<tr>
<td>Oceanic ∆pCO(_2) climatology (Takahashi et al., 2002)</td>
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<tr>
<td><strong>Atmospheric Data-based Techniques</strong></td>
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<tr>
<td>CO(_2) Inversions (Ciais et al., 1995; Francey et al., 1995; Bousquet et al., 2000; Patra et al., 2005; Le Quéré et al., 2007)</td>
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<tr>
<td>O(_2/N(_2) Measurements (Keeling et al., 1996; Battle et al., 2000; Keeling and Garcia, 2002; Bender et al., 2005; Manning and Keeling, 2006)</td>
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Table 2. Different estimates for the time-evolving oceanic CO₂ uptake between 1990–1999 assuming no uncertainty.

<table>
<thead>
<tr>
<th>Data-based Technique</th>
<th>Steady State Anthropogenic CO₂ Uptake (ΔACO₂)</th>
<th>Total Net CO₂ Uptake (ΔDICNet)</th>
<th>Non-steady state CO₂ uptake (i.e., ΔDICNet − ΔACO₂)</th>
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<tbody>
<tr>
<td>Ocean Inversion (Gruber et al., 2009)</td>
<td>−2.2 Pg C yr⁻¹</td>
<td>−1.9 Pg C yr⁻¹</td>
<td>+0.1 to +0.5 Pg C yr⁻¹</td>
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<tr>
<td>CFCs (McNeil et al., 2003; Khatiwala et al., 2009)</td>
<td>−2.0 Pg C yr⁻¹</td>
<td>−1.7 to 1.9 Pg C yr⁻¹</td>
<td></td>
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<tr>
<td>Oceanic pCO₂ Climatology</td>
<td>−1.97 Pg C yr⁻¹</td>
<td>−1.46 Pg C yr⁻¹</td>
<td>+0.35 Pg C yr⁻¹</td>
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<tr>
<td>Atmospheric O₂/N₂ (Keeling and Garcia, 2002; Manning and Keeling, 2006; Bender et al., 2006)</td>
<td></td>
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<tr>
<td>Multi-technique Difference as diagnosed here</td>
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<tr>
<td>Suite of Global Climate Models with Recent Climate Variability (Sarmiento et al., 2010)</td>
<td>−1.97 Pg C yr⁻¹</td>
<td>−1.46 Pg C yr⁻¹</td>
<td>+0.35 Pg C yr⁻¹</td>
</tr>
</tbody>
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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.

5 Conclusions

Here we decompose the time evolution of net CO₂ changes in the ocean to clarify the notion of “anthropogenic” CO₂ uptake. Traditionally, oceanic tracer-based techniques have constrained net oceanic storage of CO₂ associated with elevated CO₂ concentrations in the atmosphere under the assumption of a steady state ocean carbon cycle. Both climate-change and decadal changes to the oceanic CO₂ cycle shown recently have clearly marked a new era of non-steady state conditions for CO₂ that was already known from other biogeochemical parameters like oxygen. This global non-steady state CO₂ signal is estimated to have outgassed about 6.3 Pg C of CO₂ (or ~3 ppm to the atmosphere) between 1989 and 2007, which is ~18% of the net oceanic CO₂ uptake rate estimated from models.

After illustrating the different components of the time-evolving oceanic CO₂ sink, we present a simple concept to estimate the non-steady state oceanic CO₂ 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 Pg C yr$^{-1}$ outgassing over the last two decades, however, the
uncertainty across the suite of different data-based tech-
niques is too large at present ($\sim$ 0.4–0.6 Pg C yr$^{-1}$) to provide
a significant non-zero estimate of the non-steady state signal.

Although the non-steady state CO$_2$ signal currently pro-
vides a positive feedback to atmospheric CO$_2$ 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$_2$
than expected from steady state conditions (Zickfeld et al.,
2008).

In the end, atmospheric CO$_2$ levels only change via the
total time-evolving CO$_2$ changes in the ocean, which based
on recent evidence is now entering a new non-steady state
mode. Given this emerging mode, the challenge for the ob-
servational 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$_2$ in the ocean
and the steady state anthropogenic CO$_2$ signal on decadal
time frames. By embracing more accurate and diverse tech-
niques, we can better help detect how a changing ocean is
modifying rising atmospheric CO$_2$ levels.

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