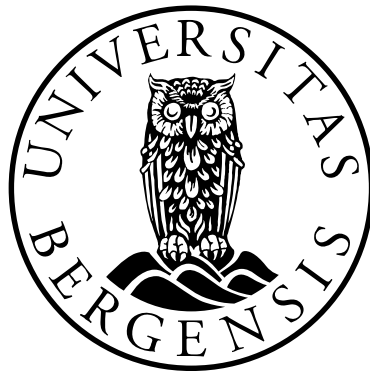# Transport of inorganic carbon through, within, and below the ocean surface

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at the University of Bergen

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Transport of inorganic carbon through, within, and below the ocean surface

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Acknowledgements

After three surprisingly short years, I am immensely proud of myself for the accomplishment this thesis is. I have had great help and great support along the way from family and friends, from my fellow PhD students, from the research groups at GFI and BCCR, and not least from my supervisors.

I have had really great supervisors: Are Olsen, Truls Johannessen, Ingunn Skjelvan, and Karen Assmann. Thank you all for believing in me and in my ideas, and for offering advice when I needed it. A special thank you to Are for all the time, dedication, advice, and inspiration you have given. I have really enjoyed the many hours of fruitful and inspiring discussion. You have been a fantastic supervisor and mentor. I really could not have done this without you.

Anna and Helene – you have both been such great friends throughout these past years, and you deserve a very special thank you for dragging me out of the office to eat, drink and be social these last few weeks and months. I will make sure to return the favor. And my office mates, Emil and Anna – it has been a great time and lots of fun. And lots of coffee breaks… I don't think I will ever forget the wonderfully misunderstood sarcastic jokes – which were so much more funny because they were misunderstood.

Solveig and Eva, thank you for all those early morning coffee breaks, and the accompanying stories and lively discussions. And last, but not least, a thank you to my family - for always believing, even when you didn't understand. I could not have done it without your support.

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Siv Kari Lauvset

Bergen, December 2010

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Abstract

Human emissions of CO_2 through the burning of fossil fuel, cement production, and land use change have increased the atmospheric CO_2 concentration by 39 % since 1750. Without the large uptake of anthropogenic CO_2 by the ocean and land the atmospheric CO_2 increase would have been much larger. The focus of this thesis is the ocean carbon sink in the North Atlantic and the Nordic Seas, and how this responds to transport of carbon.

PAPER I deals with transport of carbon through the atmosphere and ocean boundary layer. Using the eddy covariance method, fluxes of CO_2 were measured in the Greenland Sea, and this flux data set was used to test and verify a recently published correction method for the CO_2-H_2O crosstalk problems. After successful correction the average flux was in accord with calculations using parameterizations of the transfer velocity, showing that directly measuring correct CO_2 fluxes over the ocean is possible.

PAPER II presents a large-scale study of the distribution of the CO_2 fugacity (fCO_2) within the North Atlantic surface ocean, and its relationship with sea surface temperature and surface ocean circulation. The key finding is that the circulation variations driven by the North Atlantic Oscillation (NAO) impacts the large-scale distribution of fCO_2, and that in most regions of the North Atlantic this can explain the recent decreasing trend in the carbon sink. The link between NAO and fCO_2 also provides some evidence of reversibility of the recent changes in the North Atlantic carbon sink.

PAPERS III and IV focus on the transport of carbon below the surface ocean. PAPER IV presents a carbon budget for the Nordic Seas, using the mean state of volume and carbon fluxes into and out of this region. PAPER III uses the carbon tracer C* to study the processes of vertical mixing and water mass transformations in the Nordic Seas and North Atlantic Subpolar Gyre. C* is found to be a particularly good tracer for Nordic Seas' overflow water. PAPERS III and IV present results that have impact on how we understand the global ocean carbon uptake.

Combined, the four papers in this thesis increase our understanding of the recent trends in the North Atlantic carbon sink, as well as the variability of this carbon sink. In addition, these papers are a contribution to our understanding of the possible future feedbacks on the global ocean carbon sink.

List of papers

PAPER I:

Lauvset, S.K., W.R McGillis, L. Bariteau, C.W. Fairall, T. Johannessen, A. Olsen, C.J. Zappa. Direct Measurements of CO_2 flux in the Greenland Sea. *Submitted to Geophysical Research Letters*

PAPER II:

Lauvset, S.K., A. Olsen, R. Wanninkhof, T. Takahashi, A.V. Borges, I. Skjelvan, X.A. Padin, F.F. Perez, A.F. Rios, W.-J. Cai, N. Lefèvre, M. Gonzalez Davila, J.M. Santana-Casiano, T. Steinhoff, T. Johannessen, D. Pierrot, U. Schuster. North Atlantic Relationships between Surface fCO_2 and Hydrography. *Submitted to Deep Sea Research – Part I*

PAPER III:

Lauvset, S.K., E. Jeansson, K. Assmann, A. Olsen. C* as a water mass tracer: The Nordic Seas overflow waters. *Submitted to Geophysical Research Letters*

PAPER IV:

Jeansson, E., A. Olsen, T. Eldevik, I. Skjelvan, A.M. Omar, S. Lauvset, J.E.Ø. Nilsen, R.G.J. Bellerby, T. Johannessen, E. Falck. The Nordic Seas carbon budget: Sources, sinks and uncertainties. *Submitted to Global Biogeochemical Cycles*

Contents

1. Introduction and motivation

CO_2 emissions from burning of fossil fuels, cement production and land use changes have increased the atmospheric CO_2 concentration by on average ~1.5 µatm yr^{-1} since 1960 (IPCC, 2007) . Moreover, since the emissions are continuing to rise – from 6.4±0.4 GtC yr^{-1} between 1990 and 2000 to 7.7±0.5 GtC yr^{-1} between 2000 and 2008 (Le Quéré, 2010) – the rate of change in the atmospheric CO_2 concentration is also increasing and was 1.9 µatm yr^{-1} over the past decade (Global Carbon Project, 2010). The total increase since the beginning of the industrial revolution is 39 %, from ~280 ppm to 387 ppm in December 2010 (Global Carbon Project, 2010).
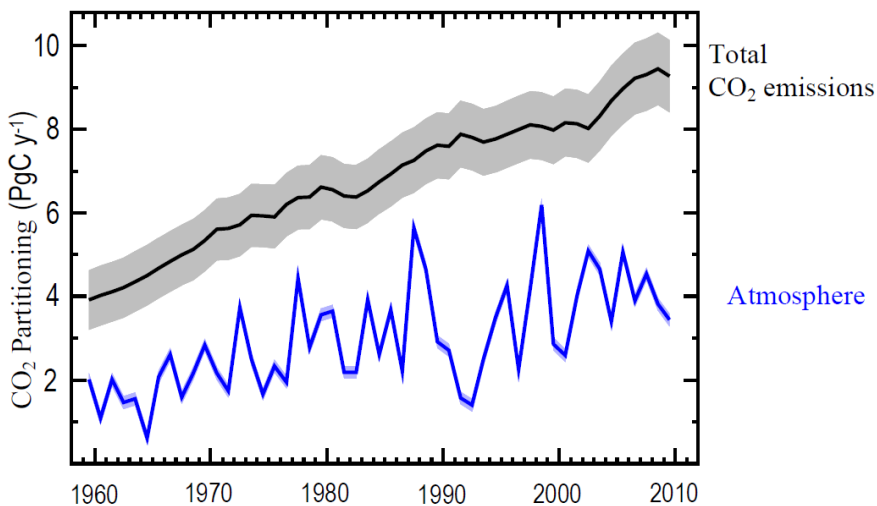

Figure 1. This schematic shows the total CO_2 emissions (black) from fossil fuel burning, cement production, and land use change after 1958, with uncertainties in grey shading. In blue is shown the atmospheric CO_2 content and the increase and variability of this since 1958. For details on data and uncertainties see the Global Carbon Project (2010).

If all of the CO_2 emitted from human activities were accumulated in the atmosphere the increase of the atmospheric CO_2 concentration would have been much larger than what is observed today (Fig. 1). This discrepancy is due to the natural carbon sinks, which act to remove carbon from the atmosphere. The two major, short-term (*i.e.* within a thousand years), natural carbon sinks are the land biosphere and the oceans (Fig. 2). Between the Industrial Revolution and 2008 the ocean accumulated 140±25

Gt anthropogenic carbon (Khatiwala *et al.*, 2009), and after 1958 the ocean and land sinks have taken up 57 % of the emissions (Global Carbon Project, 2010). Without these two sinks the current atmospheric CO_2 level would be ~500 ppm instead of the observed 387 ppm (Friedlingstein and Prentice, 2010; IPCC, 2007). The natural sinks have thus substantially reduced the effect of human CO_2 emissions on global climate.
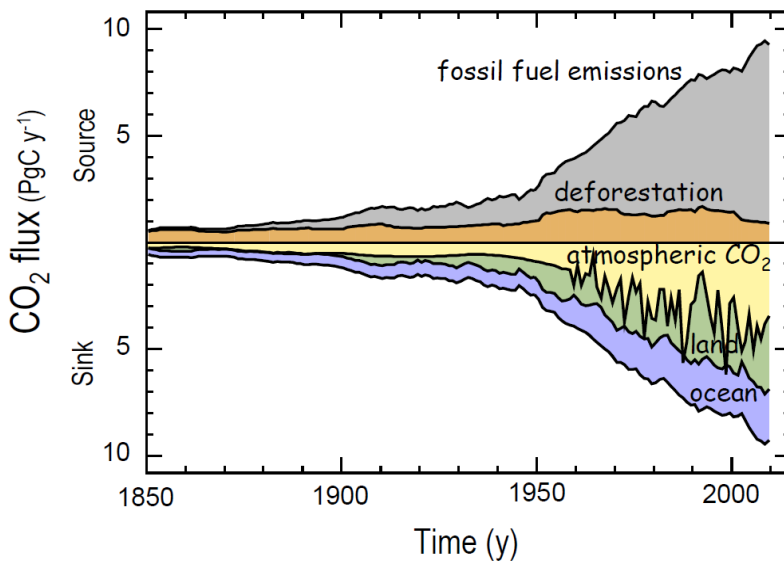

Figure 2. This schematic shows all the major sources and sinks of CO_2 and their development after 1850. The fossil fuel emissions make up the largest source, and though there is large interannual variability in the airborne fraction (atmospheric CO_2), the land and ocean sinks have remained approximately about equal in size. The land sink is here estimated as the residual from the balance of the other sources and sinks. For quantitative size estimates of the source and sinks see the Carbon Budget 2009 (Global Carbon Project, 2010).

The most recent global carbon budget from the Global Carbon Project (2010) is calculated using both observations and models, and show that the ocean and land sinks are about equal in size. In Fig. 2 the ocean sink is an average based on the results of five different models, and the results show that currently the ocean takes up about 25 %, or 2.3±0.4 GtC yr^{-1}, of the emissions (Le Quéré *et al.*, 2010). The land carbon sink can be estimated by terrestrial biogeochemical models, but the Fig. 2 estimate is the residual from total emissions minus the ocean and atmosphere sinks

(Global Carbon Project, 2010). The ocean sink has also been calculated using observational methods (Khatiwala *et al.*, 2009; Takahashi *et al.*, 2009) with very similar results as the model estimates. There are still uncertainties attached to the quantification of today's ocean carbon sink, but the largest unknowns are related to the future efficiency of the ocean carbon sink as it is responding to changes in carbon chemistry, climate change, and ocean circulation changes (Friedlingstein *et al.*, 2006; Le Quéré *et al.*, 2010). Moreover, since the equilibration of CO_2 between the surface and deep ocean takes several hundred to a thousand of years (Broecker and Peng, 1982), it will take a long time to bring the carbon system back to a new equilibrium.
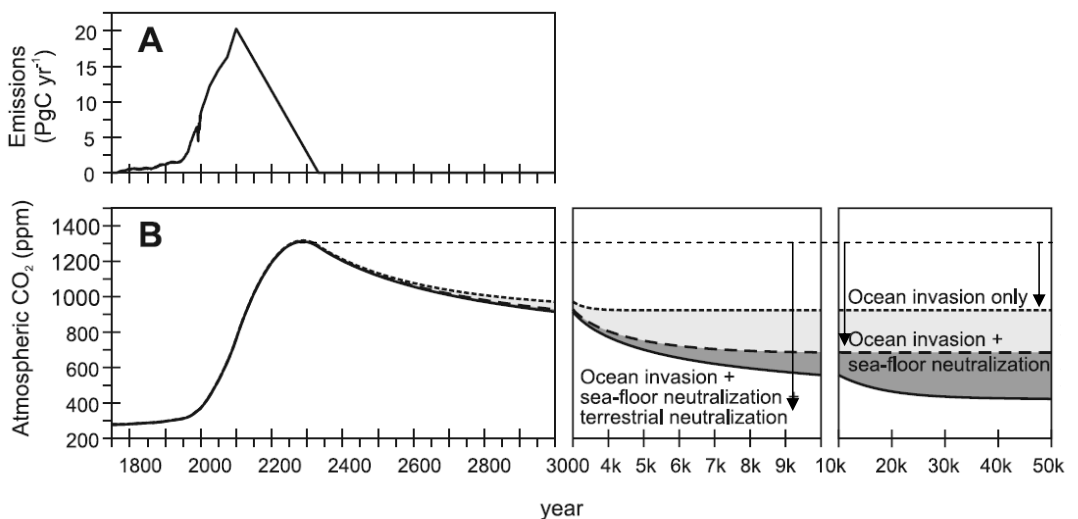

Figure 3. A) This panel shows a pulse in CO_2 emissions peaking in 2100 and decreasing linearly to a total of 4000 GtC by 2400. B) These panels show the atmospheric response to the emission scenario shown in A). The total invasion in the ocean, shown in white, is the immediate response, which is followed by dissolution of $CaCO_3$ deposits on the sea floor, shown in light grey, which begins to dominate the CO_2 removal ~1000-1500 years after the peak in emissions. At the same time the more gradual, and longer lasting, neutralization of CO_2 by terrestrial weathering vs. burial of $CaCO_3$, shown in dark grey, also remove carbon from the atmosphere. For details on the model and method see Ridgwell and Hargreaves (2007). Note that this figure shows results without a climate feedback on the ocean carbon sink.

On long time scales the ocean sink seems to dominate over the land sink (Sabine *et al.*, 2004), and eventually the ocean will remove almost all anthropogenic carbon from the atmosphere. The long-term effect of the anthropogenic CO_2 emissions will

however also involve the long-term carbon sinks: dissolution of calcium carbonate ($CaCO_3$) deposits on the ocean floor; and burial of $CaCO_3$ in the ocean versus weathering of $CaCO_3$ on land. On even longer timescales the weathering of silicate becomes an important sink (Walker *et al.*, 1981). Fig. 3 shows the results of a model study which describe a pulse of carbon emissions totaling 4000 GtC, which is the estimated total fossil fuel reserve, and the effect of this on atmospheric CO_2 (Ridgwell and Hargreaves, 2007). The first process to remove CO_2 from the atmosphere is the ocean uptake. The oceans require ~1000 years to fully renew the deep ocean water masses (Broecker and Peng, 1982), thus the ocean uptake reaches its equilibrium ~1000 years after the peak in CO2 emissions. The second process is the dissolution of $CaCO_3$ in the ocean sediments, and the third process is the neutralization of atmospheric CO_2 by the balance between terrestrial weathering and oceanic burial of $CaCO_3$ (Fig. 3) (Ridgwell and Hargreaves, 2007). This model study was run for 50,000 years, and even after this new steady state is reached the atmospheric CO_2 concentration is significantly higher than it was in the pre-industrial era (Archer *et al.*, 2009; Ridgwell and Hargreaves, 2007).

How long the peak in atmospheric CO_2 concentration lasts (*i.e.* how wide the peak becomes) is directly related to how long it will take the ocean carbon uptake to significantly reduce the atmospheric CO_2. Ocean uptake depends on the transport of carbon in the ocean in the form of fluxes through the ocean surface, advection within the ocean surface, and deep vertical transport below the ocean surface. This thesis focuses on the North Atlantic Ocean and studies all these transports and the processes that drive them.

The carbon uptake from air-sea flux through the ocean surface is controlled by turbulence in the atmosphere-ocean boundary layer (Section 1.1) and the CO_2 saturation state in the surface ocean (Section 1.2). Turbulence is caused by local forces, while the CO_2 saturation state depends on biological activity and on transport of carbon within the surface ocean. The surface transport of carbon is important for the variability in ocean carbon uptake on a regional scale. The carbon uptake from deep vertical transport below the ocean surface is controlled by the meridional

overturning circulation (Section 1.3). Without vertical transport the surface ocean would fill up with carbon within a year, so this transport into the deep ocean is necessary to maintain the carbon sink on the global and long term scale.

In sum, this thesis provides a novel and comprehensive view of both the current state of the ocean carbon cycle in the North Atlantic and its recent changes.

1.1 Transport through the ocean surface

The surface ocean continuously exchanges gas with the atmosphere, and since this is an equilibrium process gas is transferred both to and from the atmosphere. The flux of CO_2 can be directly measured using micrometeorological techniques and knowledge of boundary layer turbulence in the atmosphere, and this is commonly done over land (*e.g.* Järvi *et al.*, 2009). However, a major problem the past 30 years or more has been the difficulty in making such direct measurements over the ocean (Broecker *et al.*, 1986; Wanninkhof *et al.*, 2009). Over the ocean the measurements are often inaccurate and too high (Broecker *et al.*, 1986; Kondo and Tsukamoto, 2007; Prytherch *et al.*, 2010), and there are a number of reasons for this: the net flux is usually small; the corrections needed to account for the movement of the platform at sea are often large; there is a correlation between CO_2 and H_2O measurements due to density changes (Webb *et al.*, 1980); and there is a cross-sensitivity between CO_2 flux and fluctuations in water vapor (Prytherch *et al.*, 2010). Because of these difficulties with direct flux measurements parameterizations of the CO_2 flux are necessary. These are of the form

$$F = kS\Delta fCO_2 \qquad\qquad (1)$$

where ΔfCO_2 is the difference in fugacity of CO_2 (fCO_2) between ocean and atmosphere, S is the solubility of CO_2 in sea water, and k is the transfer velocity. In The term k is usually parameterized as a function of wind speed (U), but k is really a function of turbulence in the atmospheric and oceanic boundary layers for which wind speed is only one forcing (Wanninkhof *et al.*, 2009). Other important forcing processes for k (Fig. 4) include energy dissipation (Zappa *et al.*, 2007), bubble

entrainment (Asher and Wanninkhof, 1998), fetch (Woolf, 2005), and atmospheric stability (Erickson, 1993). Wind speed is used since it has a dominant effect on k, and it is also theoretically sound since wind speed is closely related to the friction velocity, u_* (Wanninkhof *et al.*, 2009).

Over the past decades several k-U parameterizations have been experimentally derived using a variety of methods: radiocarbon invasion (Naegler *et al.*, 2006; Sweeney *et al.*, 2007; Wanninkhof, 1992); dual tracer release experiments (Ho *et al.*, 2006; Liss and Merlivat, 1986; Nightingale *et al.*, 2000); wind-wave tank experiments (Liss and Merlivat, 1986); and micrometeorological techniques (McGillis *et al.*, 2001; Wanninkhof and McGillis, 1999). There are several limitations to these k-U parameterizations (Wanninkhof *et al.*, 2009) and recently efforts have been made to find parameterizations that do not depend on U. One of the most promising of these approaches is the mean square surface slope, which is an estimate for surface roughness and can be determined through remote sensing (Frew *et al.*, 2007).
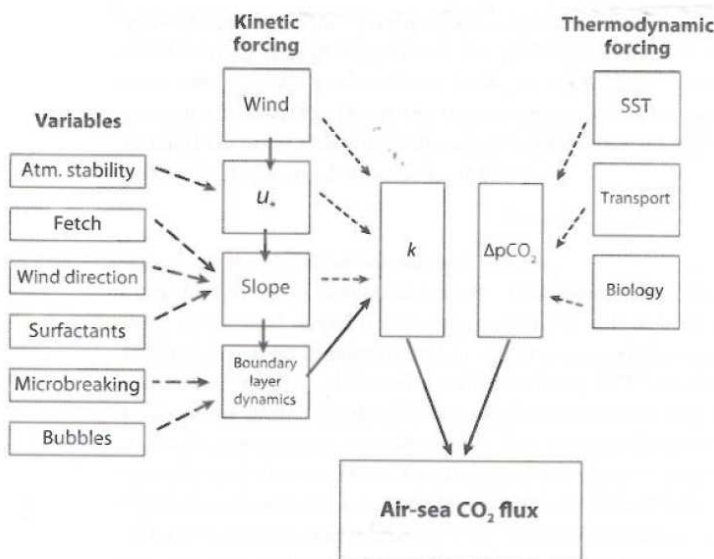

Figure 4. This schematic shows many of the different variables and forcing processes that control air-sea gas exchange of CO_2. From Wanninkhof *et al.* (2009).

As discussed above much research has gone into furthering our understanding of k, and improving our ability both to measure and to parameterize k. Regardless, when comparing even the most recent studies (Naegler *et al.*, 2006; Sweeney *et al.*, 2007) they have quite a large span in k, which leads to a significant uncertainty on the estimates of CO_2 flux. Successful direct measurements are therefore very valuable and there would be much to gain if the direct measurement techniques were perfected and could be used to reduce this uncertainty. The past couple of decades new methods have been developed to help solve the issues of platform motion and correction for this (*e.g.* Edson *et al.*, 1998; Griessbaum *et al.*, 2010), and much effort is also going into further improvements of the correction for flow distortions around moving platforms (*e.g.* Yelland *et al.*, 2009) and the CO_2-H_2O crosstalk (Prytherch *et al.*, 2010). PAPER I adds to the efforts of the latter by using a new data set from the Greenland Sea in summer 2006 in an independent test and verification of the Prytherch *et al.* (2010) method.

1.2 Transport within the ocean surface

The surface ocean CO_2 saturation varies regionally, and generally the tropics have high fCO_2 and the high-latitude regions have low fCO_2 (Fig. 5). The saturation state of CO_2 in the surface ocean is controlled by the combined forces of solubility, which is lower in warm water than in cold water, and biological activity. The effects of biology are related to photosynthesis and respiration as well as formation and dissolution of $CaCO_3$, but these processes are beyond the scope of this thesis and will not be further discussed. The surface circulation is a major force for both the heat (Joyce and Zhang, 2010) and the carbon (Anderson and Olsen, 2002) transport in the ocean, and therefore also a major force for the regional differences in the oceanic carbon uptake. In the North Atlantic there is large variability in the surface circulation, particularly linked to variability in the North Atlantic Oscillation (NAO) (Bersch, 2002; Flatau *et al.*, 2003; Hátún *et al.*, 2005). Model studies indicate that these variations also affect the North Atlantic carbon sink (Thomas *et al.*, 2008; Ullman *et al.*, 2009).
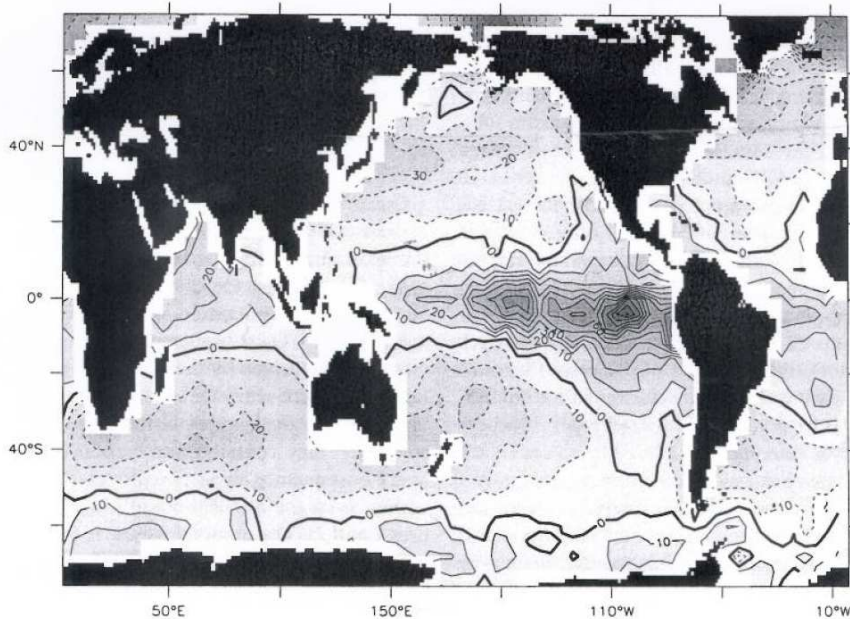
Figure 5. This map shows the ΔpCO_2 (*i.e.* difference between atmosphere and surface ocean partial pressure of CO_2) in the world oceans in 2000. Positive values mean release of CO_2 to the atmosphere while negative values mean uptake of CO_2 by the ocean. From Takahashi *et al.* (2009).

Since the North Atlantic is a large oceanic carbon sink, different aspects of this sink – *e.g.* changes, variability, and sensitivity – have been extensively studied using both observations (*e.g.* Corbière *et al.*, 2007; Schuster and Watson, 2007; Telszewski *et al.*, 2009; Watson *et al.*, 2009) and models (*e.g.* Sarmiento *et al.*, 2000; Thomas *et al.*, 2008; Ullman *et al.*, 2009). Observations of the ocean carbon system in the North Atlantic have revealed clear interannual variations in sea surface fCO_2 (Corbière *et al.*, 2007; Lefèvre *et al.*, 2004; Omar and Olsen, 2006; Schuster and Watson, 2007; Schuster *et al.*, 2009; Skjelvan *et al.*, 2008), which are different from the rise expected of a surface ocean carbon system that changes in pace with the atmospheric fCO_2 (Fig. 6). There are many different suggestions as to the forces and mechanisms behind these changes in the surface ocean, *e.g.* an increase in sea surface temperature (Corbière *et al.*, 2007), advection of water with high anthropogenic carbon content from the equatorial Atlantic (Omar and Olsen, 2006) and the associated changes in the buffer capacity (Olsen *et al.*, 2006), decreased biological activity (Lefèvre *et al.*,

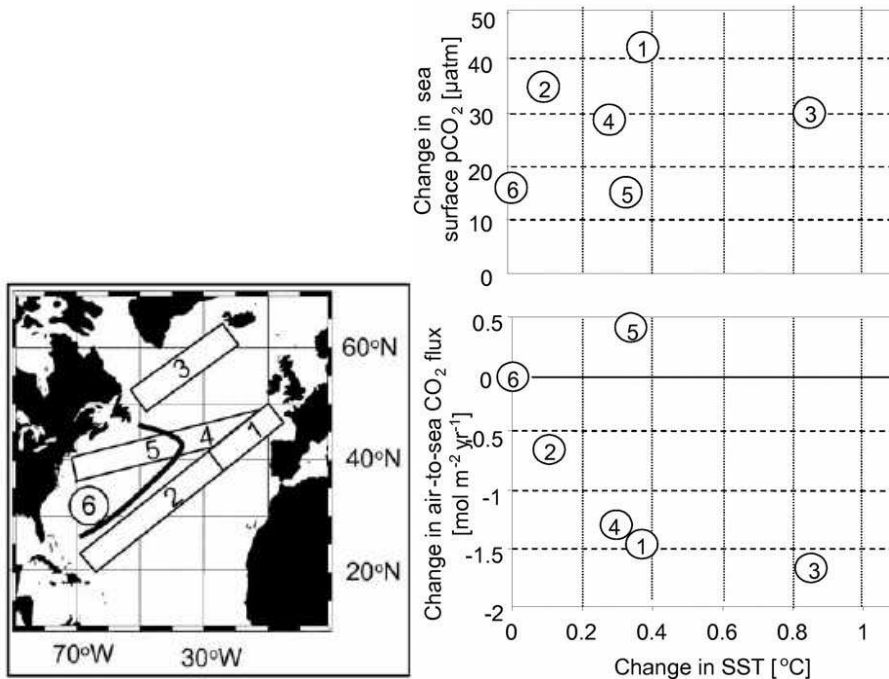2004), changes in ocean mixing and ventilation (Schuster and Watson, 2007), or a combination of the above.

Figure 6. The map on the left hand side shows the study area of six different publications reporting on changing trends in oceanic fCO$_2$. On the right hand side the results are shown, and it is seen that the carbon sink (*i.e.* the air-to-sea CO$_2$ flux) has decreased in four of the six studies. For more details see Schuster and Watson (2007).

The efficiency of the carbon sink in the North Atlantic appears to be decreasing (Corbière *et al.*, 2007; Lüger *et al.*, 2006; Schuster and Watson, 2007), but most observations are made after 1995 which gives us a trend over less than two decades. There are still great uncertainties about whether the sink efficiency will continue to decrease (Gruber, 2009), but models predict a significant decrease in the sink efficiency in the future (Friedlingstein *et al.*, 2006; Friedlingstein and Prentice, 2010; Le Quéré, 2010; Le Quéré *et al.*, 2010). In this thesis the focus is on the distribution of fCO$_2$ in the surface ocean and how this distribution is related to the different water masses in the surface ocean. Using this method the reversibility in the North Atlantic carbon sink is investigated.

One problem with all the observational studies above is the lack of a sufficiently large-scale scope, especially concerning the spatial variability of fCO$_2$. PAPER II changes this by looking specifically at the large-scale spatial distribution of fCO$_2$ in the North Atlantic, and its relationship to water masses. A significant decrease in the efficiency of the North Atlantic carbon sink has serious implications for our future climate, but PAPER II implies some reversibility to the recent changes, as transport of carbon is found to be directly linked to the variability in the North Atlantic carbon sink.

1.3 Transport below the ocean surface

While the transport within the ocean surface lead to regional variability in carbon uptake, the vertical transport below the ocean surface and the transport in the deep ocean leads not only to regional change but is also a controlling factor in the global carbon uptake (Sabine *et al.*, 2004). Without vertical transport there would be no significant ocean carbon sink and the long-term magnitude of the carbon sink (Fig. 3) is therefore dependent on how quickly carbon can be transported from the surface to the deep ocean. The reason for the large inventory of anthropogenic carbon in the North Atlantic (Sabine *et al.*, 2004) is therefore its link to the Atlantic Meridional Overturning Circulation (Fig. 7). The high ventilation rates in this region infuse the entire water column with anthropogenic carbon.

The Nordic Seas (*i.e.* the Norwegian, Greenland, and Iceland Seas) is a deep water formation region (Fig. 7), where in the Greenland Sea surface water becomes dense enough to sink to ~1000-1500 m depth (Karstensen *et al.*, 2005). In the global carbon budget calculations (*e.g.* Gruber *et al.*, 2009; Le Quéré *et al.*, 2009) the Nordic Seas is a very small region, but the dense water production here is an important part of AMOC (Hansen and Østerhus, 2000), and the gross transport of carbon associated with this Nordic Seas' dense water overflowing the Greenland-Scotland Ridge is estimated to ~3 % of the global ocean carbon uptake (Olsen *et al.*, 2010). The relatively large carbon flux, considering that the Nordic Seas make up only 0.3 % of the world ocean volume, found by Olsen *et al.* (2010) means that the Nordic Seas is

important for the global carbon budget. Still, only one dedicated carbon mass balance budget for the Nordic Seas has been published (Lundberg and Haugan, 1996). That study had a very limited data set, and PAPER IV is an updated carbon mass budget for the Nordic Seas only, which use much more comprehensive carbon data and volume flux estimates.
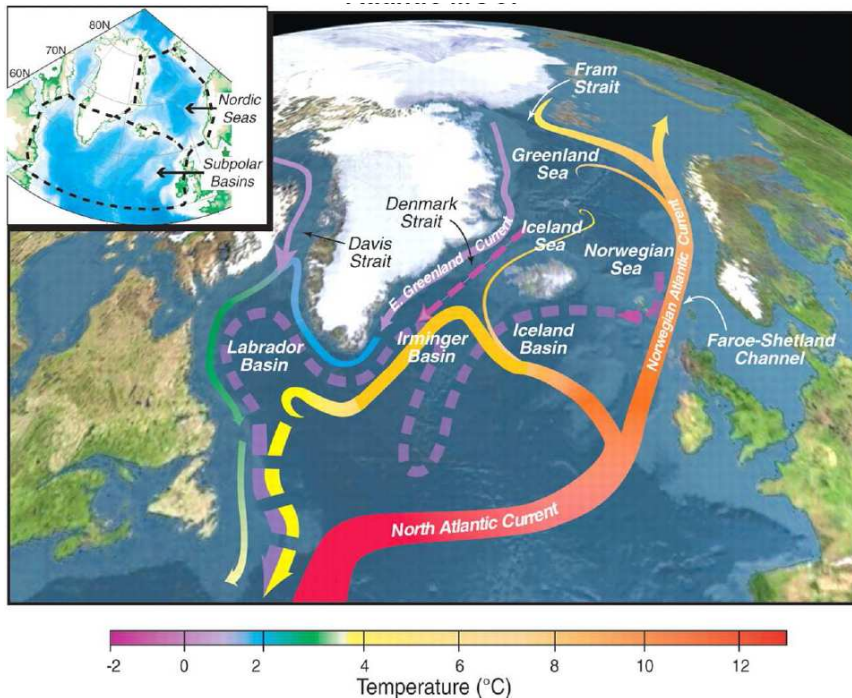
Figure 7. A map of the northern North Atlantic Ocean, with the northernmost part of the Atlantic Meridional Overturning Circulation (AMOC) schematically shown. The surface currents are shown as solid arrows while deep currents are shown as dashed arrows. From Curry and Mauritzen (2005).

Calculating the mean state of the carbon uptake in the Nordic Seas using mass balance is important, but of equal interest are the processes of mixing and water mass transformation in the interior ocean, and how these affect the long-term variability of the carbon uptake. Observations agree that the volume of dense water from the Nordic Seas that overflows the Greenland-Scotland Ridge is ~30 % of the total North Atlantic Deep Water (NADW) volume (Hansen *et al.*, 2004; Voet and Quadfasel, 2010). However, both the mechanisms of formation of the Nordic Seas' overflow

water and the entrainment of ambient water into the overflow plumes south of the Greenland-Scotland Ridge are questions of many years of research and debate (Dickson and Brown, 1994; Eldevik *et al.*, 2009; Fogelqvist *et al.*, 2003; Jeansson *et al.*, 2008; Tanhua *et al.*, 2005; Voet and Quadfasel, 2010). PAPER III is a new contribution to this research, which uses a quasi-conservative carbon tracer C* (Gruber *et al.*, 1996), to estimate both the entrainment south of the Greenland-Scotland Ridge and the mixing processes within the Nordic Seas which form the overflow. Both these processes are important for understanding the long-term distribution of CO_2 in the world's oceans.

2. Discussion and wider implications

2.1 Transport through the ocean surface

In PAPER I a new data set from the Greenland Sea in summer 2006 was successfully used in an independent verification of the PKT correction method for the CO_2–H_2O crosstalk (Prytherch *et al.*, 2010). This is an important result as improved correction methods for direct measurements of CO_2 flux will be valuable in constraining accurate values for the gas transfer velocity (k, Section 1.1). Using direct flux measurements to find k still has its limitations, not least the requirement for large ocean CO_2 undersaturation during measurements, but the PKT method and this verification of it helps resolve one of the most difficult issues, and as such presents a step in the right direction.

In addition we now have a data set of observed CO_2 flux in the Greenland Sea, and given the general lack of direct observations the importance of this data set is obvious as the Greenland Sea is considered a strong carbon sink. However, the CO_2 flux in the Greenland Sea in the summer of 2006 is small—0.72×10^{-2} mol m^{-2} day^{-1} into the ocean—despite the significant undersaturation (>100 µatm) that points to a large potential for air-sea carbon uptake. This unused potential, and its effect on estimates of carbon uptake in the Greenland Sea, should be carefully considered as changes in the CO_2 saturation state not necessarily will lead to changes in the CO_2 flux. The small flux is due to the low wind speeds in the Greenland Sea in the summertime and thus highlights a seasonal bias. It is therefore important to keep in mind that single season measurements of CO_2 flux are not enough to resolve the annual carbon uptake in this region.

The results in PAPER I point to the possibility of future measurements and quantifications of the CO_2 flux. This implies that we will have the means of quantitatively comparing the estimated Greenland Sea mass balance carbon uptake (*e.g.* PAPER IV) to direct observations of the CO_2 fluxes. This will lead the way to a

full synthesis and comparison of the different methods used to measure and calculate the carbon uptake in the Nordic Seas.

2.2 Transport within the ocean surface

The past few years there has been much research on the North Atlantic carbon sink, including its trends and variabilities (Section 1.2). PAPER II focuses on explaining the recent changes in oceanic fCO_2 (fCO_2^{sea}) in the North Atlantic by using a large-scale view to determine the mechanisms behind the changes in the carbon sink. It is shown that in the North Atlantic there are strong links between ocean physics and ocean chemistry, and the overall transport of carbon within the surface ocean is linked to the surface circulation. This has previously only been shown in model studies (*e.g.* Thomas *et al.*, 2008; Ullman *et al.*, 2009).

The changes in the North Atlantic carbon sink are driven by two main mechanisms: the variability in the circulation forced by the NAO; and the changes in supply of carbon enriched subsurface waters due to variations in vertical mixing and ventilation. In the North Atlantic Subpolar Gyre the influence and supply of carbon enriched subsurface water is most important, while in the temperate North Atlantic circulation changes dominate. The results show that many, though not all, of the observed changes in fCO_2^{sea} are linked to changes in the NAO index, and the implication is that many of the changes in the North Atlantic carbon sink observed so far are reversible, since the response of the different surface currents to NAO variability is reversible. A possible reversibility of the recent trends in the North Atlantic carbon sink is highly relevant for our understanding of the future climate – carbon cycle feedback on the global ocean carbon sink. While short-term and regional studies of fCO_2^{sea} changes are valuable, PAPER II clearly show that a more large-scale and long-term approach is necessary to resolve the as yet open questions about the development of the ocean carbon sink.

In addition to improving our understanding and interpretation of the observations, the more dynamical understanding of fCO_2^{sea} and its distribution and variability could

lead to improvements in modeling both present and future oceanic fCO_2, and thus the overall efficiency of the ocean carbon sink. Models have so far not been able to reproduce the observed trends (see Section 1.2) in the North Atlantic (Le Quéré *et al.*, 2010; Wetzel *et al.*, 2005), but they are able to reproduce the observed trends in the Southern Ocean (Le Quéré *et al.*, 2010). The results in PAPER II might help resolve this mismatch.

2.3 Transport below the ocean surface

PAPER IV presents an updated mean state mass balance carbon budget for the Nordic Seas, using data from 2002 and 2003. This data set is both more recent and significantly larger than the data set used in the only previous Nordic Seas carbon budget (Lundberg and Haugan, 1996). The Nordic Seas' overflow waters clearly have a climatic importance as they remove CO_2 from the atmosphere and surface ocean by transporting carbon into the deep North Atlantic ocean. In this new budget the transport of anthropogenic carbon into the deep North Atlantic with the overflow water amounts to 4 % of the global ocean carbon uptake, consistent with recent estimates using different methods (Olsen *et al.*, 2010). The budget also estimates the present-day CO_2 uptake to be 23 % lower than the pre-industrial uptake, giving supporting evidence for decreasing carbon sink efficiency (Section 1.2). The uncertainties associated with this decrease highlight the need for observing the transport of carbon over many years.

Carbon transport and carbon uptake in the ocean is inextricably linked to the ocean circulation. There is evidence that the variability in ocean volume fluxes has a large impact on the carbon sink. This is discussed in PAPER IV but our current knowledge of the variations in volume fluxes, and especially the variability associated with individual water masses, is not good enough to say anything definitive about the consequences this has on the carbon uptake. If ocean circulation changes lead to a greater volume transport into the Nordic Seas, then the result will be a greater volume transport out of the Nordic Seas. However, this is not necessarily true for carbon as greater carbon transport into the Nordic Seas might lead to greater accumulation of

carbon here, and thus changes in surface saturation of carbon, carbon storage within the Nordic Seas, and transport of carbon out of the Nordic Seas with the overflow water.

PAPER III links the quasi-conservative carbon tracer C* (Gruber *et al.*, 1996), to the Nordic Seas' overflow waters which have high C* because they are cold and well-ventilated. By using C* the Nordic Seas' overflow waters are tracked to the Greenland-Scotland Ridge and into the North Atlantic, and the amount of mixing with ambient water masses are calculated. These results add to our understanding of the processes of mixing and entrainment in the Nordic Seas and North Atlantic Subpolar Gyre, and also introduce a new tracer specifically for these water masses. An overall understanding of the processes that modify and form water masses in the ocean interior is important for understanding variability and change in the deep ocean carbon sink. Our current understanding of the future of the ocean carbon sink (Section 1.3) says that stratification will increase in the future ocean due to rising temperatures, and since maintaining the carbon sink requires vertical transport a stronger stratification will lower the carbon sink efficiency. In this context understanding the transport of carbon vertically in the ocean today is essential, and PAPERS III and IV provide both new knowledge, and new tools for researching these processes.

3. Future work

During my work on this thesis several questions and topics have arisen that would be worth pursuing in future research. Related to PAPER I and PAPER IV plans are being made to use the data base of fCO_2^{sea} observations in the Nordic Seas and Barents Sea to estimate the total carbon uptake. This would allow for comparison of uptake estimates from three different methods: direct air-sea flux measurements; mass budget calculations; and ΔfCO_2 based estimates. Do the results from these methods agree? If not what are the reasons for this disagreement and what can be done to converge the results?

In PAPER IV the variability in the volume fluxes is highlighted as a source of uncertainty, and there is still much we do not know about this. The consequences this has for the carbon sink should be further investigated.

Directly related to PAPER I a further analysis of the CO_2–H_2O crosstalk correction is needed. Specifically for the experiment in PAPER I the influence of that particular instrument set-up is interesting, as well as the effect of the added motion bias correction. More generally there are unresolved issues involving the crosstalk correction when the latent heat flux is very low. Also related to the direct flux measurements an expansion of the data set is desired. In 2009 direct flux measurements were made in the Nordic Seas over a period of eight weeks. This data set will be analysed, and it covers a larger region as well as a larger range in wind speeds than the data presented in PAPER I. Parts of the 2009 data set cover the same region as the 2006 data, so I want to do a comparison analysis of the two years.

In both 2006 and 2009 full carbon chemistry measurements were made on the same cruises that measured CO_2 flux. The 2009 cruise is particularly interesting since it covered all the gateways to the Nordic Seas. Related to the work in PAPERS I and IV It would be interesting to see if using the 2009 carbon chemistry and the 2009 flux measurements would improve our carbon budgets for the Nordic Seas.

It want to expand the work done with C* in PAPER III. It would be particularly interesting to use C* in a full water mass analysis to constrain all the components making up Denmark Strait Overflow Water (DSOW) in the Nordic Seas. C* can also be used to do a more detailed analysis of the modification of DSOW in the Irminger Sea. Formation of DSOW is a highly dynamic process and there is still no consensus on the formation process, so I want to add C* as a defining parameters and see if this can increase our understanding.

C* is such a good tracer for the Nordic Seas' overflow water that I want to see how far we can track the DSOW/ISOW part of the North Atlantic Deep Water. The signal might become too diluted to track very quickly, but this is yet an unknown and would be interesting to look at in more detail.

Particularly related to PAPER II there are still open questions regarding determination of where the high fCO_2 in the Subpolar Gyre comes from, and what mechanisms are behind the recent trends in this carbon sink. For this a quantification of the effects of air-sea exchange vs. deep mixing and biology will be necessary. There are obviously still one or more unknown factors in this region, and maybe in all regions of the North Atlantic, and before we know what these are we cannot fully understand how the region will behave in the future. In addition there are some interesting implications of the subsurface "nutrient stream" (Williams *et al.*, 2006) which is observed beneath the Gulf Stream. How is this related to the carbon changes seen both in the eastern temperate North Atlantic and in the Subpolar Gyre? Have there over the past decades been changes in the "nutrient stream" that might affect the surface carbon system?

In PAPER II I have only looked at wintertime data. Including all seasons means that biological activity needs to be considered, but it would be interesting to find out what effect the large-scale seasonal variabilities have on the North Atlantic carbon sink, and what part circulation plays in this.

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