Carbon Exchange Dynamics in Coastal and Marine Ecosystems

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Abstract

This project's goal is to understand how climate change will affect the air-sea exchange of climatically relevant gases (carbon dioxide-CO_2, dimethylsulfide-DMS, and nitrous oxide-N_2O). Oceans exert considerable influence on climate through their role on the global cycling of climatic active gases. For example, the world's oceans are nature's largest sink for CO_2 and they globally account for a significant proportion of the natural emissions of the greenhouse gas nitrous oxide (N_2O), and the vast majority of dimethyl sulfide (DMS) production. DMS is the largest source of sulfate in the marine environment, and once in the atmosphere the compound can trigger the formation of aerosols that serve as cloud condensation nuclei. While greenhouse gases (CO_2 and N_2O) act to warm the atmosphere, the increased production of DMS may have a cooling effect on climate by increasing back-scattered solar radiation. Our understanding of the effect that a changing Arctic climate and sea ice regime will have on the air-sea (and sea ice) exchange of these trace gases is currently somewhere between partially understood and mostly unknown, and hence important feedbacks that involve sea ice and the cycles of climate-active gases are currently not represented in general circulation models.

A requirement of this project is to parameterize those processes affecting the distribution of dissolved CO_2, N_2O, and DMS in surface waters of the Arctic, and their exchange with the atmosphere. Newly developed parameterizations are being implemented into a coupled atmosphere-sea ice-ocean biogeochemistry models to learn how the ocean's response (physical, biogeochemical and biological) to climate change and variability will affect the atmosphere-ocean cycling of these climate active gases within the Arctic, and in turn how these regional processes may affect the global budgets of these gases.

Key Messages

- The Arctic Ocean is more vulnerable to ocean acidification than its Antarctic counterpart, because the Arctic experiences larger seasonal signals in temperature and hydroprahic properties.
- Based on a recovered data set spanning from 1974 to 2009, pCO_2 is increasing and $CaCO_3$ saturation state is decreasing in the halocline of the Beaufort Sea. This is primarily a result of increasing DIC and decreasing alkalinity.
- The mineral, ikaite, is prevalent in seasonal sea ice, and its distribution is inversely related to temperature.
- The precipitation of ikaite between crystal boundaries enriches brine network with dissolved CO_2.
- Analysis of sea-ice carbonate system data from CFL also found that physical processes, $CaCO_3$ precipitation/dissolution and gas exchange, dominated carbonate system cycling in the upper ice, even in spring. In contrast, the carbonate system in the bottom ice was strongly influced by bacterial respriation and exchange with the underlying water in deep winter, and primary production did not become important until May.
- A five week time series of discrete carbonate system (DIC, Alk, pCO_2) and isotopic tracer ($del^{13}C$-DIC and $del^{18}O$-H_2O) measurements from sea ice, sack hole brine, and the underlying water column during the 2010 winter-spring transition in Resolute Passage, Nunavut confirmed that while biological processes dominated the biogeochemistry of the bottom ice, physical processes still dominated in the upper parts of the ice, even as CO_2 exchanges with the atmosphere switched from outgassing to uptake.
- DMS measurements from the three activities showed very high DMS concentrations in the different Arctic environments investigated: bottom sea ice (2,000 nmol l^{-1}), under sea ice (11 nmol l^{-1}), and open water (13 nmol l^{-1}).
- Elevated concentrations of DMS (up to 14 nmol l^{-1}) were measured in the melt ponds, confirming the importance of this new source of gases for the Arctic atmosphere in spring. This is an important finding considering that the extent of melt ponds is predicted to increase with global warming.

Objectives

Our overarching objective is to understand the effects of climate change on the air-sea exchange and associated exchange budgets of climatically relevant gases (carbon dioxide-CO_2 , dimethylsulfide-DMS, and nitrous oxide-N_2O) in the Canadian coastal Arctic environment. Sub-objectives follow.

Research, particularly that of our group, has shown that sea ice in both the Arctic and Antarctic actively exchanges CO_2 with the atmosphere. Subsequent work is required to better understand the myriad of physical, biogeochemical and biological processes that underpin the release and uptake of CO_2. The latter would benefit from directed field measurements over sea ice, and controlled cold tank experiments (at SERF at CEOS and at IOS) to focus on the response of specific carbonate parameters, including carbonate minerals, to various effects (e.g, temperature, sea water properties). The results from these experiments, in conjunction with our fieldwork will provide valuable parameterizations for our group's sea ice modelling specialists.

Sub-Objective 1: Development of robust relationships between sea ice properties and surface CO_2 exchange. The work will produce modelling algorithms widely applicable within the ArcticNet domain.

The bulk micrometeorological technique for air-sea CO_2 exchange has minimal data requirements, and therefore is commonly used in carbon balance studies and remote sensing inversion algorithms (e.g., Else et al., 2007). Central to the model is the transfer velocity and their parameterizations have been developed for open sea conditions and having infinite wind fetch. In addition, previous results from our group indicate that a surface micro layer (SML) is widely prevalent in the ArcticNet domain and of a different composition than observed in temperate Pacific waters. The effect of the SML is an addition resistance to the air-water exchange gas, and is implicitly represented by a transfer velocity parameterization. Additional sampling of the SML and in presence of direct flux measurements is required both to characterize the nature of the SML and its affect on transfer velocity in Arctic waters. Existing parameterizations for the transfer velocity have little predictive value in Arctic ice-dominated seas.

Both the seawater pCO_2 and resulting air-sea flux show tremendous variability on short space and time scales within the ArcticNet domain. Ship-based measurements blend both spatial and temporal variability. Satellite-based remote sensing offers a means of regionally monitoring the drivers of the air-sea exchange at reasonable temporal frequency. Attempts to apply remote sensing to estimating air-sea CO_2 fluxes and budgets in Arctic seas have been limited by sea ice and the 'universality' of empirically-derived sea water pCO_2 relationships involving sea water temperature and water colour. The development of robust 'Arctic' parameterizations requires the improved understanding on the myriad of physical, biogeochemical and biological processes that underpin the observed variability.

Sub-Objective 2: To improve our understanding of the physical, chemical and biological drivers of the carbonate system variability.

Sub-Objective 3: Development of robust relationships between detectable water properties and surface sea water pCO_2. Collectively (Arctic-optimized transfer velocity and pCO_2 relationships) the work will produce remote sensing inversion algorithms widely applicable within the ArcticNet domain.

Research, particularly that of our group, has shown that sea ice in both the Arctic and Antarctic actively produces DMS for release to the atmosphere. Subsequent work is required to better understand the myriad of physical, biogeochemical and biological processes that underpin the release DMS.

Peaks in DMS emissions and cloud condensation nucleation events have been observed by our group and immediate collaborators.

Sub-Objective 4: To address key uncertainties regarding the production and emission of DMS in the ArcticNet domain, their impact on climate, and to understand how these emissions may be affected by the decline of the summer extent of the sea ice pack.

Introduction

Observations show that most regions of the Arctic Ocean have warmed (Steele et al., 2008). Its sea ice cover is thinner, younger, and absent altogether for a longer portion of the annual cycle (Nghiem et al., 2007). The intensity of cyclones has increased, particularly in the fall (Simmonds and Keay, 2009), and associated with changes to the climate is a continued freshening of the surface waters (Manabe et al., 2004; Lenaerts et al., 2013). A consequence of the ongoing and anticipated changes in the Arctic Ocean, its watershed and overlying atmosphere, are feedbacks to the climate system through changes in the air-sea exchange of CO_2 and other climate-active gases (McGuire et al., 2010).

A result of the recent retreat of summer Arctic sea ice is the increased potential for air-sea gas exchange, particularly over the extensive Arctic shelves which are ice-free over larger temporal and spatial scales. A hypothesis is that because sea ice is thought to impede gas exchange and radiative transfer, and because waters within the shelf system tend to be undersaturated in CO_2 relative to atmospheric levels, the retreating and thinning sea ice should enhance the ocean's uptake of CO_2 (Bates and Mathis, 2009), and possibly the air-sea fluxes of other biogenically-produced climate-active gases, including nitrous oxide (N_2O) and dimethyle sulphide (DMS).

Related to the greenhouse gas CO_2, a number of alternative scenarios may unfold depending on competing responses of the ocean's carbonate system to climate change. Increased freshwater loading of the Arctic Ocean will support water-column stratification, which should limit the CO_2 draw-down potential through: (i) access to nutrients that sustain primary production, and (ii) warming of the surface layer that raises the seawater partial pressure of CO_2 (pCO_2) (Cai et al., 2010; Else et al., 2013). The prevalence of open water and mobile sea ice on the other hand may encourage episodes of the vertical mixing. While strong upwelling will replenish nutrients in the surface layer, thereby supporting primary production (e.g., Tremblay et al., 2011), the concomitant introduction of inorganic carbon from the halocline will tend to supersaturate CO_2 in the surface layer relative to atmospheric levels, transforming a region which may otherwise exist as CO_2 sink into a source of atmospheric CO_2 (Else et al., 2012a,b, Lansard et al., 2012; Mucci et al., 2010). An additional consequence of the 'new' ice regime is greater ice production during the cold season, which may enhance a sea ice carbon pump (Rysgaard et al., 2009; 2011), ultimately pushing the system toward stronger CO_2 uptake (Else et al. 2011).

Nitrous oxide (N_2O) is a powerful greenhouse gas, and dimethylsulfide (DMS) is the most important source of material for secondary aerosol formation (and cloud condensation nuclei) in the marine boundary layer (Lovelock et al., 1972). The enhanced production and emission of DMS from a warmer ocean forms the basis for a negative feedback through an increase in backscattered solar radiation that is hypothesized to accompany the heightened loading of sulphate aerosols (Charlson et al., 1987). DMS is mainly produced by the enzymatic conversion of dimethylsulphoniopropionate (DMSP). A third organosulfur compound, dimethylsulfoxide (DMSO), acts both as a source and sink for DMS through photochemical and bacterial processes. While a proposed climate regulation mechanism involving a feedback loop between climate warming and DMS production has been questioned (Quinn and Bates 2011), feedbacks between oceanic DMS emissions and regional climate do appear to be significant,

especially in the Arctic, where warming is amplified. Peaks in DMS emissions and nucleation events were recorded during the ArcticNet/Arctic SOLAS program in Lancaster Sound (Chang et al., 2011, Rempillo et al., 2011; Motard-Côté et al., 2011). Seasonal sea ice itself is seen as a biogeochemical medium favorable to the production of DMS/P/O (Carnat et al. 2014a,b; Galindo et al., 2014), and significant DMS fluxes from sea ice have been detected in the Antarctic (Zemmelink et al., 2006; Nomura et al., 2012). Those same ice algal blooms responsible for heightened DMS production may potentially also produce N_2O (Randall et al., 2012).

Untangling likely responses to climate change of the system's carbonate system, and emission characteristics of climate-active gases forms the basis of this projects underlying objective.

Activities

1. Six team members took part in the shortened 2013 science cruise of the CCGS Amundsen. The project objectives were (i) to examine hydrographic and biological drivers of the carbonate system variability, by determining the involved water masses, and the biological modulation of the hydrographic signal, (ii) the effects of said drivers on air-sea CO_2 exchange, (iii) document how fresh water (ice melt and river water) influence surface water pCO_2 and air-sea CO_2 exchange in the Labrador fjords, and across waterways of the Canadian Arctic Archipelago (CAA), (iv) explore the dynamics, distribution and production of DMS and distribution N_2O in the ArcticNet domain, in relation with the complex oceanic circulation and sea ice retreat, and (v) to study the spectral irradiance dependence of sunlight effects (UV/PAR) and photo-acclimation response of phytoplankton cells on dimethylsulfide (DMS) production. The Baffin Bay data (objective 1) from the 2013 expedition will be compared to data gathered during the 2007/2008 CFL/ARCTINET expeditions, and the earlier 1998 North water Polyna study, in order to gain first insights in temporal variability of the Baffin Bay system. Monitoring and on-site experiments consisted of basic ship- and boat (zodiac and barge) based sampling of water for carbonate chemistry analysis and oxygen-18 isotopic ratio. The dissolved CO_2 concentration in water was measured using a portable automated flow through air-water CO_2 monitoring system. Additional measurements included water analysis for DMSP and DMS concentrations in the water column. Labeled sulfur (35S) experiments were also conducted and analyzed on board of the CCGS Amundsen. N_2O and DMS profiles were produced at all full stations and some basic stations. Cruise participants included B. Else, T. Burgers, M. Pind (UofM), J. Mol (Dalhousie), M. Gourdal (Laval), and S-J Royer (Institut de Ciències del Mar, Barcelone, Spain).

2. Team members (B. Else. N. Geilfus, T. Papakyriakou, S. Rysgaard, J. Sievers, and L.L. Sørenson) participated in the 2013 Sea Ice Experimental Research Facility (SERF) experiment at the University of Manitoba. Research was undertaken in collaboration with the University of Manitoba's CERC program. Objectives were to quantify the inorganic carbon flows and budget associated in a closed sea ice / water column system. Overlapping SERF objectives with this ArcticNet project were to (i) investigate controls on ikaite formation/dissolution and (ii) derive carbon mass balances through a complete sea ice growth/decay cycle. Investigators (Geilfus, Papakyriakou, Sørenson and Sievers) contributed to complete the carbon mass balance by measuring the surface flux of CO_2 over small pond footprints using flux chambers and application of the eddy covariance approach with a hot-wire anemometer system.
3. One team member participated in the Arctic-ICE springtime sea ice study on Allen Bay (northwest of Resolute Bay: 74° 43 N; 95° 09 W) during the month of May. The ice camp provides a unique opportunity for multidisciplinary study into sea ice and upper ocean biology, biogeochemistry and geophysics over the spring to summer transition. This year's focus was a spatio-temporal analysis into the production of DMS and its precursors among ice environments typical of the Canadian

Arctic Archipelago. N_2O measurements were also conducted in the brines, the bottom of the ice, and in the water column.

4. Five team members (B. Else, N. Geilfus, T. Papakyriakou, J. Sievers and L.L. Sørenson) participated on the Nuuk sea ice campaign in southwest Greenland. Research was undertaken in collaboration with the University of Manitoba's CERC program. Our team's objectives were to investigate the dynamics of greenhouse gases within natural sea ice environments, strongly differing from those commonly observed within the CAA. Snow cover is typically deeper and sea ice thinner in the fjords relative to our CAA. Research revolved around two flux stations, one temporarily deployed within an ice-covered fjord (Papakyriakou), and the other long-term facility positions on land at the terminus of the fjord, and operated by Sørensen (Arctic Research Centre, Aarhus University, Denmark). The latter provided a unique opportunity to sample air-sea CO_2 exchange open water in the presence of forming sea ice – something not accomplished since the 2007-08 CFL overwintering campaign. Data provide the opportunity to confirm the presence of enhanced CO_2 uptake in freezing seawater previously reported by Else et al. (2011).
5. One team collaborator (C.J. Mundy) participated on a research cruise onboard the R/V Martin Bergman in proximity to Cambridge Bay. The cruise provided the opportunity for exploratory investigations into the distribution of seawater pCO_2 as influenced by freshwater and primary production. Research was conducted in collaboration with Mundy's ArcticIce project, and the University of Manitoba's CERC program, each with their own complementary research objectives.

   Our ship and ice-camp measurement suite also provides fundamental information on site microclimate in support of several ArcticNet projects. This dataset supported several publications through collaborations with other ArcticNet projects.

   In addition to field activities, we analyzed data from previous ship- and ice-camp experiments, and government databases and forwarded our modelling initiative.
6. Work has continued with the Global Ocean Turbulence Model (GOTM) (now including sea ice) with the aim of implementing an additional biological component representing ice algae. Work over the last year has involved basic development of GOTM, e.g. introducing iron limitation, testing parameter sensitivities, and calculating biogeochemical forcing of dissolved inorganic carbon in the system. Work has continued on model parameterisations of light penetration and attenuation in the GOTM sea-ice module to improve the representation of sub-ice and within-ice photosynthetic active radiation (PAR). The basis of the applied snow distribution module has been tested and improved. Progress has also been made in adapting the new Framework for Aquatic Biogeochemical Models (FABM) linked to GOTM. This includes transferring the existing biogeochemical model into the FABM structure and testing if the new structure is suitable for the combined sea ice - ice algae modelling system. This system will allow more flexibility with the exchange and combination of different ecosystem modules.
7. In parallel, we are continuing the the evaluation of the Canadian Earth System Model in the Arctic in collaboration with CCCma.

Results

Sea Ice Carbon Chemistry and Sea Ice - Ocean Carbon Transport

Ikaite is a calcium carbonate mineral thought to underpin a sea ice-driven carbon pump (Delille et al., 2007; Rysgaard et al., 2007, 2011, Papadimitriou et al., 2012). Its occurrence has been notoriously difficult to quantify, which has hindered our understanding on which environments support its precipitation and dissolution. Rysgaard et al. (2013a) report, for the first time on the vertical distribution of ikaite in winter sea ice in the Young Sound area of NE Greenland. The concentration of ikaite was 4-10 times higher than those reported in previous studies, and decreased with depth from surface to bottom layers of the ice. The crystals were located between the interstices of the

sea ice crystals. Solutes and gases (CO_2) were visible under microscope and were observed to move between the interstices of ice crystals. These observations confirm previous hypotheses (Rysgaard et al., 2007, 2009, 2011) that ikaite crystals are trapped within the sea ice matrix, whereas CO_2 released through ikaite production and dissolved within the brine can be lost from the sea ice. As a result, total alkalinity (TA) increases relative to dissolved inorganic carbon (DIC) in sea ice in spring and summer. When ikaite crystals dissolve during sea ice melt, surface water salinity and pCO_2 will decrease, promoting the uptake of CO_2.

The dynamic nature of ikaite was further examined by Rysgaard et al (2013b) in artificial sea ice at the SERF facility. During the experiment, ikaite precipitated in sea ice with temperatures below -3°C in distinct zones, in decreasing concentration from the surface brine skim (and frost flowers) to the ice base. Snow was observed to affect ikaite distribution through its affect on ice temperature. That is, snow accumulation caused the sea ice to warm, dissolving ikaite crystal, while the (artificial) removal of the snow cover allowed the sea ice to cool, allowing brine salinities to increase, resulting in rapid ikaite precipitation. Ikaite production appeared a strong function of temperature, and its occurrence was well model using the chemical model FREZCHEM. The model simulates and predicts the behaviour of substances at cold temperatures.

Fransson et al., (2013) examined the seasonal evolution of the sea ice carbonate system, and established links between sea ice processes and the carbonate system of the under-ice water. Observations were made during the 2007-08 CFL/ArcticNet IPY expedition. Changes in sea ice carbon system parameters (dissolved inorganic carbon – DIC, total alkalinity – TA, pH, CO_2, aragonite solubility - Ω) were associated with processes that transport and transform carbon, including: brine rejection, precipitation/dissolution of ikaite, CO_2 gas flux and biological processes of respiration and photosynthesis. Results indicated that ikaite and the CO_2 gas flux had a large impact on DIC across the entire ice core in the early spring, whereas the effect of biological processes was most evident near to the ice base. The effect of bacterial respiration on DIC was evident during all months, while the signal associated with primary production was most evident in May. Sea ice processes had a large impact on under-ice water. Observations suggest the CO_2 enrichment of sea ice brine associated were a strong increase in under ice DIC and concomitant reduction in aragonite solubility (Ω). On sea ice melt aragonite solubility increased, likely due to the combined effects of ikaite dissolution and primary production.

Brown et al. (in prep) carried out a six-week time series of carbonate system and stable isotope measurements to investigate the effects of sea-ice on air-sea CO_2 exchange during the early melt period in the Canadian Arctic Archipelago. They observed significant changes in bulk sea ice and sack-hole brine carbonate system parameters associated with increasing temperatures and the build up of chlorophyll a concentrations in bottom ice. In the bottom ice, biological carbon-uptake maintained undersaturated pCO_2 conditions throughout the time series, whereas pCO_2 was supersaturated in the upper parts of the ice. Low CO_2 permeability in the sea ice matrix and snow cover at the air-sea ice interface effectively impeded CO_2 efflux from the upper ice surface to the atmosphere, despite the strong pCO_2 gradient. Throughout the middle ice column, brine pCO_2 decreased significantly with time and was tightly controlled by sea ice temperature and in-situ melt dilution. Once the influence of melt dilution was accounted for, however, both ikaite dissolution and seawater mixing were found to contribute alkalinity and inorganic carbon to brines, with the ikaite contribution driving brine pCO_2 to values lower than predicted from melt-water dilution alone.

Sea Ice DMS Production

The production of DMS in the ocean is likely to be affected by changes in sea ice cover and thickness (through affects on biological production), while their concentration and fate in the atmosphere are likely to be affected by changing emissions from the ocean and by changing atmospheric dynamics in a warming

Arctic. Ice algal blooms that take place at the bottom of the ice in spring represent large source of DMS. Further, brine inclusions in sea ice host important algal communities and many potential producers of DMS and its precursor DMSP.

Carnat et al. (2014a) conducted a year-round survey of the DMS cycle in sea ice in Amundsen Gulf using data from the 2007-08 combined CFL/ArcticNet IPY cruises of the CCGS. The results contained therein are the first combined measurement of DMS, DMSP and dimethylsulfoxide (DMSO). DMSO is a third organosulfur compound acting as both source and sink for DMS through photochemical and bacterial processes. The contribution of each of the three dimethylated sulfur compound in the total pool of DMS was largely dominated (>50%) by DMSO in most samples, a pattern which contrast strongly with oceanic waters and Antarctic sea ice (Figure 1). These high percentages showed a linear decrease with depth

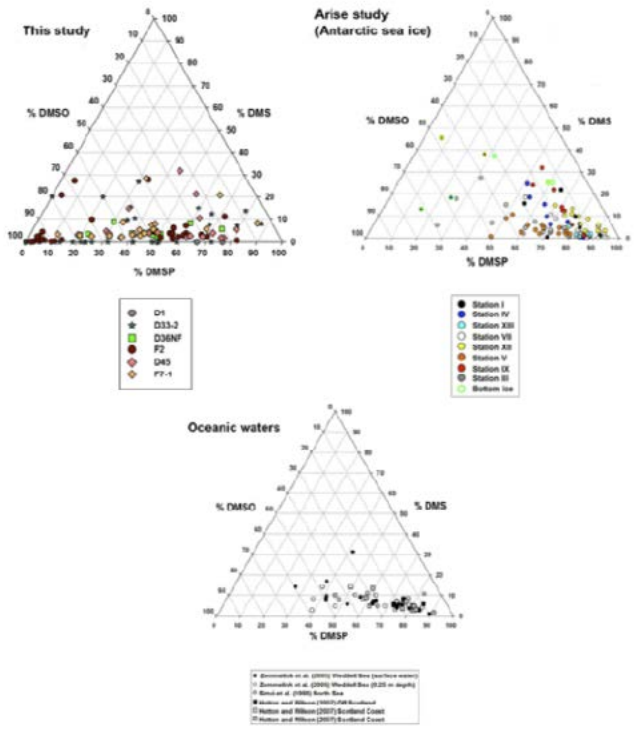

Figure 1. Percent contribution of organosulfur compounds DMS, DMSO and DMSP. (Carnat et al., 2014a).

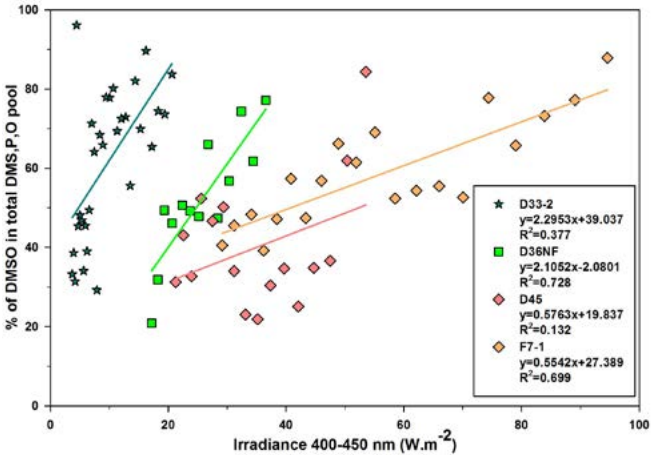

Figure 2. Percent DMSO in total DMS, P, O pool. (Carnat et al. 2014a).

and were well correlated to irradiance values (Figure 2) when the snow cover was low, suggesting that photo-oxidation of DMS into DMSO might be an important process in surface Arctic sea ice.

Measurements of DMSP (particulate and dissolved) in sea ice of the Canadian Arctic Archipelago (CAA) (Gourdal et al., in prep) confirmed our previous observations, showing very high values (up to 9,314 nmol l^{-1} and 17,500 nmol l^{-1}, respectively) found in the bottom 10 cm of the ice (see Galindo et al., 2014). In contrast with our previous sampling years, DMS measurements were made (in addition to DMSP) during the 2012 spring season (Figure 3). DMS concentrations were always highest at the bottom of sea ice (lower 10 cm), ranging between 71 and 2,000 nmol l^{-1}. In contrast with DMSPp and DMSPd concentrations, which tended to decline, DMS concentrations in the bottom sea ice increase during the sampling period. High DMS concentrations (up to 140 nmol l^{-1}) were also measured in brines.

A unique temporal series of DMSP and DMS was conducted in two melt ponds that developed toward the end of our sampling period (Figure 4). The melt ponds formed on the 10th of June 2012 and were sampled until the 23rd of June 2012. On the last day of sampling, melt ponds were several meters across

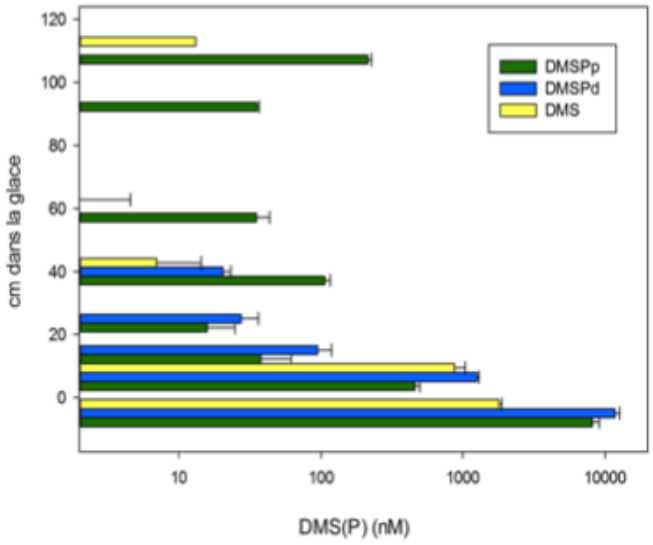

Figure 3. Vertical distribution of particulate DMSP, dissolved DMSP, and DMS concentrations measured in sea ice during the spring of 2013 near to Resolute, NT.(Gourdal et al., in prep.).

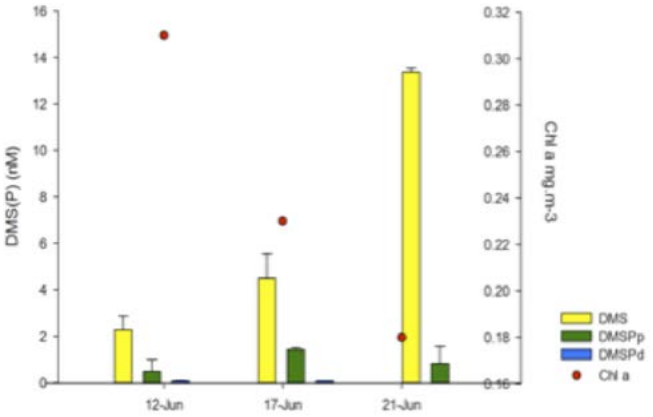

Figure 4. Temporal variations of DMSPp, DMSPd, DMS and Chl a concentrations in one melt ponds formed above sea ice on the 10th of June 2012 offshore Resolute. (Gourdal et al., in prep.).

and some of them were interconnected. The two melt ponds monitored remained independent. The melt ponds salinity was low, and increased from 0.1 to 3.2 during the sampling period. Chl a concentrations in the melt ponds ranged from 0.18 and 0.41 mg.m^{-3}. In both melt ponds, DMSPp and DMSPd concentrations were low but DMS concentrations increased progressively, reaching 14 nmol l^{-1} at the end of the sampling period.

Carnat et al. (2013b) measured the seasonal and vertical variations of DMS and DMSP in fast ice at Cape Evans, McMurdo Sound (Antarctica) during the spring-summer transition of 2011 and winter-spring transition of 2012. Measurements were motivated by our desire to broadly sample DMS production acroass a wide range in sea ice environments. Unlike Arctic profiles reported to date, isolated DMS and DMSP maxima were found in interior ice and corresponded to the occurrence of platelet crystals in the ice texture. They suggested that platelet ice formation strongly modified the production of DMSP in the ice cover by favoring the incorporation of potential strong DMSP producers and increasing the environmental stresses these producers were exposed to. From October to November (austral summer), the DMS and DMSP profiles were strongly influenced by the development and decline of a diatom-dominated bloom in bottom ice, with DMSP variations remarkably mirroring chl a variations (Figure 5), consistent with observations from the Arctic. From November to December, the increase in brine volume fraction (i.e. ice permeability) on warming was shown to trigger (1) an important release of DMS to the under-ice water through brine convection (2) a vertical redistribution of DMSP across the ice. Similar features were reported in Galindo et al. (2014).

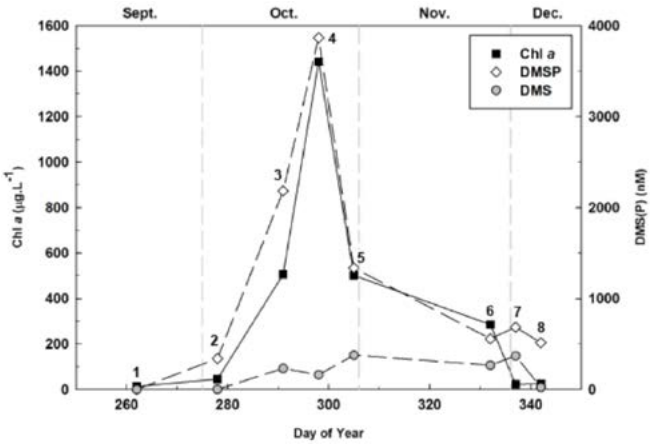

Figure 5. Seasonal evolution of DMS (nM), DMSP (nM) and chl a (ug L^{-1}) concentrations in bottom ice (5 cm). Each set of dots corresponds to a sampling station. (Carnat et al., 2013b).

Ocean Carbonate System

Results reported here are attributed to recent measurements (i.e., post IPY) and from data archived as part of government monitoring programs that pre-date the ArcticNet NCE. In regard to the former, previous work by this group has shown both the seawater pCO_2 and resulting air-sea flux show tremendous variability on short space and time scales within the ArcticNet domain. Much of our attention has, in recent years, focused on the southeastern Beaurfort Sea and Amundsen Gulf, leaving the CAA, Hudson Bay, and other coastal environments under-reported in the literature. The 2010, 2011 and 2013 ArcticNet datasets provide the opportunity to examine the response of pCO_2sw (sw corresponds to surface water), and the water carbonate system to unique regional environments. In particular the 2011 cruise (through the CAA) provided the opportunity to examine conditions associated with respectively summer and fall seasons. Several works are in progress associated with these data sets and preliminary results shown here. First however a detailed examination into the relationships between food web dynamics, CO_2 fluxes and water mass mixing in the southeastern Beaufort Sea will be reviewed.

Using an integrated data set from the 2009 Malina/ ArcticNet cruise of the CCGS Amundsen, Forest et al. (2013) developed a carbon budget across the Mackenzie shelf break in the southeastern Beaufort Sea to identify key indices of ecosystem response to climate change. Key questions included: (1) To what extent net ecosystem metabolism was coupled with air-sea CO_2 fluxes? (2) What are the roles of cross-shelf exchange, river inputs and sea ice in driving carbon fluxes? (3) How did he planktonic food web respond and feedback to organic and inorganic carbon cycling?

Results indicated that the system was over-all net heterotrophic, but none-the-less acted as a weak sink for atmospheric CO_2. Uptake estimates were comparable to published rates in earlier studies (e.g., Else et al., 2012a; Mucci et al., 2010). This apparent contradiction was attributed elevated primary production over the shelf prior to the study, and through the response of the inorganic carbon system to freshwater dilution by river runoff and sea ice melt and the presence of cold surface waters offshore. Key results of the study are that: climate change is exacerbating an existing biological gradient across the shelf-basin system, the Mackenzie Shelf acts as a weak sink for atmospheric CO_2, the effect of CO_2 transfer to the atmosphere associated with shelf break upwelling can be mitigated in the presence of enhanced diatom production associated with the concomitant introduction of nutrients to the surface layer.

An examination of the comparatives roles of the biological and physical processes in shaping the biogeochemical properties of Hudson Bay was undertaken by Pengelly (2013). They observed that the characteristics of the surface waters in Hudson Bay were dominated by river input. The river signal was observed until approximately 50 m of depth. Data indicate that the intermediate waters of Bay are of Pacific origin from the CAA. Brine formation, as evidenced by salinity concentrations greater than 33, altered the waters in Foxe Basin and in the deepest waters of Hudson Bay (Figure 6). In the Bay, the brine formation signal was weaker and appeared overwhelmed by biological processes. The δ13CDIC values became isotopically lighter while the DIC increased as the water moved from west, counter-clockwise to east around the Bay. The DIC was observed to be greater and the δ13CDIC more depleted on the east side of Hudson Bay, compared to the west side (Figure 5). First thoughts are that the observation is attributed to differences in river input and primary production, as δ13CDIC is only affected by biological production and the invasion of CO_2. In previous work Else et al. (2008) showed the Bay to be only a weak CO_2 sink, therefore it is assumed that the difference in distribution of carbon species is largely related to biological production.

Pind (2013) observed the waters within the CAA to be undersaturated in CO_2 relative to atmospheric levels, and hence the waterway acted as a sink for atmospheric CO_2, absorbing on average -17.6 mmol

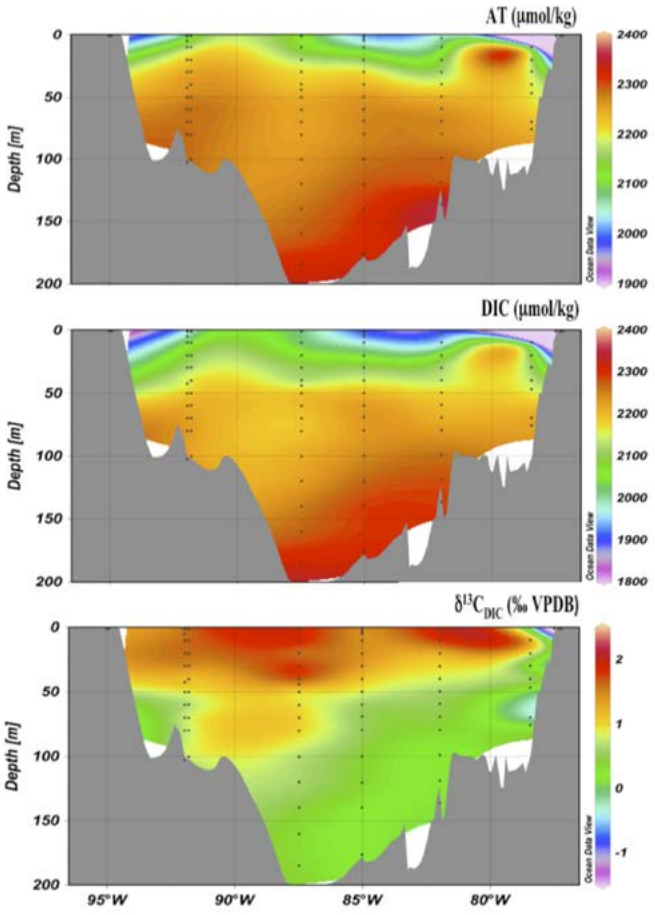

Figure 6. Depth transects moving across Hudson Bay from west to east for; AT, DIC, and 13CDIC. (Pengelly, 2013).

CO_2 m^{-2} day^{-1}. The observation is consistent with studies across Arctic shelves. Although higher mean pCO_2sw was observed during the fall season, a larger range in values occurred during the summer. The general pattern observed in pCO_2sw was that of increasing values from east to west during the summer and fall periods (Figure 7), consistent with trends in temperature and salinity. Variation in pCO_2 across the cruise track is dramatic, where considerable change in pCO_2sw is observed across very short spatial scales. Variation is interpreted in Figure 8. Much of the variability was associated with changing temperature and availability of fresh water from sources including river discharge and ice melt from both sea ice and icebergs. The highest pCO_2sw was associated with

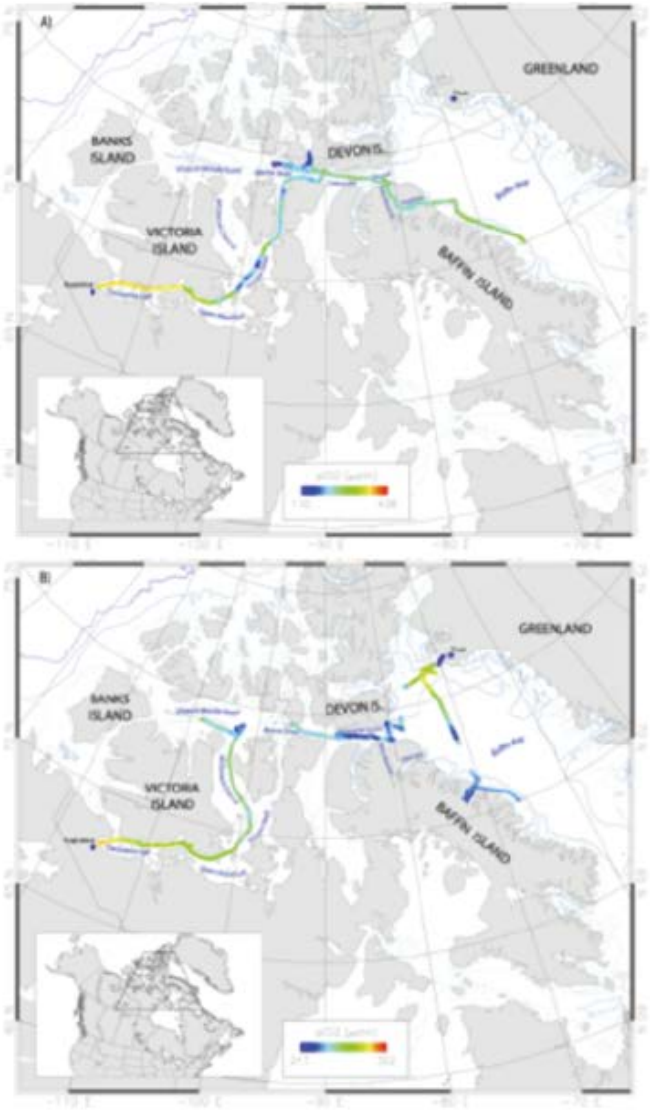

Figure 7. Cruise track of pCO2 across the CAA during the summer (A) and fall (B) season. (Pind, 2013).

the influx of freshwater from the Coppermine River (i.e., the westernmost reach of Coronation Gulf). In contrast, the lowest pCO_2sw was located just west of Thule, Greenland. The waters at this locale were slightly warmer and less saline than the waters to its west, but of particular note was that many icebergs littered the waters towards Greenland. In general lowest pCO_2 was co-located with melting ice (sea ice and icebergs). Alternatively, high pCO_2 corresponded to regions of river inflow and ice formation.

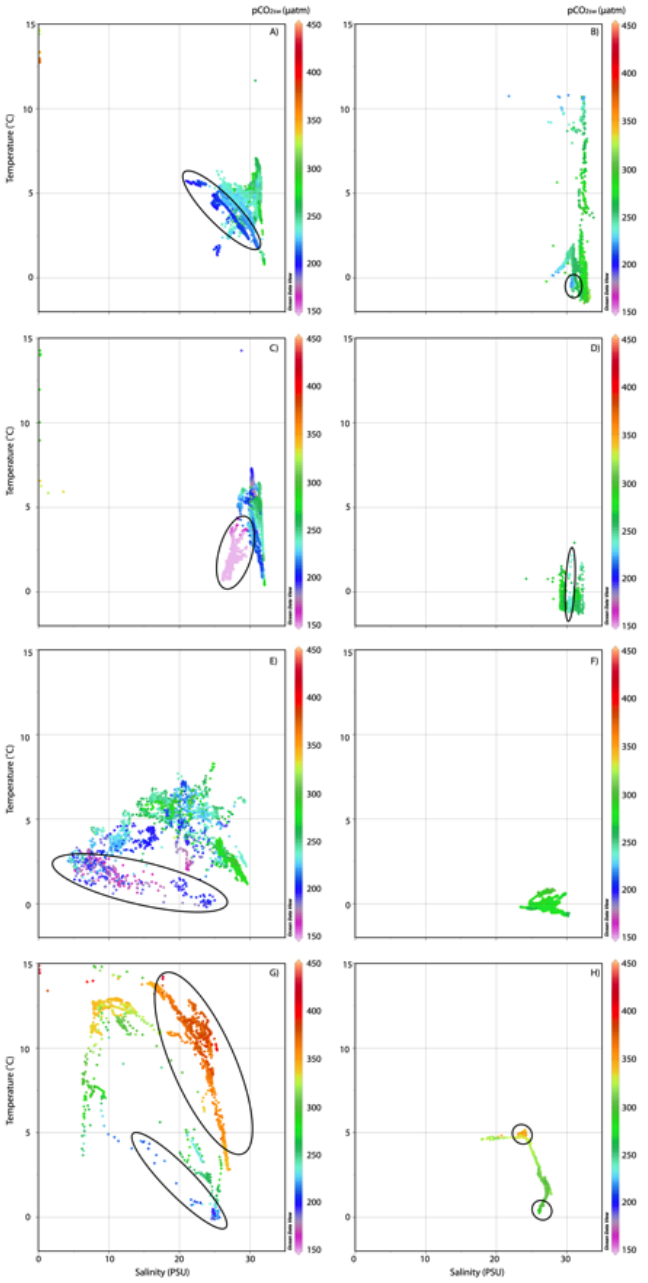

Figure 8. pCO_2 is presented in temperature-salinity space in Baffin Bay during summer (A) and fall (B), Lancaster Sound during summer (C) and fall (D), Victoria Strait during the summer (E) and fall (F), and in Coronation Gulf during the summer (G) and fall (H) cruises. Circled areas represent regions of lowered pCO_2sw from melted ice (A, C, E, lower circled area in G), melting icebergs (B), river influence (upper circles in G and H), and waters cooling prior to ice forming (H). Y axis has been trucated (Pind, 2013).

Results from the 2013 cruise of the CCGS Amundsen showed interesting relationships between pCO_2, freshwater, and primary production in the Labrador fjords (Else et al., in prep.). In the Okak Fjord, we recorded some of the lowest pCO_2 measurements we have ever observed (<100 uatm, Figure 8). These particularly low values were associated with fresh water, which suggests that river discharge may be playing an important role in CO_2 cycling in this fjord. In Nachvak Fjord, we observed low pCO_2 values near the fjord entrance (225 uatm, Figure 9), which were associated with extremely strong primary production (M. Gosselin, pers. comm., 2013). Analyses to better understand these pCO_2 distributions is ongoing, and should yield interesting results in the near future.

Although ocean carbonate system research has accelerated over the last two decades in response to public and political concern about climate change and ocean acidification, chemical oceanographers have been studying the marine inorganic carbon cycle for much longer. The Institute of Ocean Sciences began measuring inorganic carbon (total inorganic carbon, DIC; total alkalinity, TA) in the Arctic Ocean in 1974. Logistics, protocols, and analytical methods have obviously changed since that time, hopefully improving data quality, but nonetheless, the early data

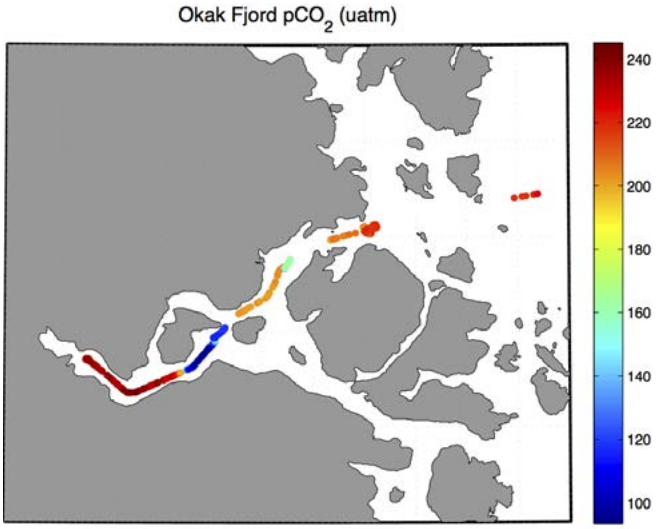

Figure 9. Observed pCO_2 in Okak Fjord.

are a valuable resource contributing insight into how the system has evolved.

Miller et al. (submitted) examined changes in the marine carbonate system of the western Arctic between 1970 and 2010. The characteristic profiles carbon system parameters for the Beaufort Sea and southern Canada Basin show substantial variability within the upper halocline, with DIC concentrations intermittently varying by up to 100 µmol/kg (Figure 10a). The change from high values in 1990-1992 to lower values in 1995 is coincident with observations of large-scale water-mass changes, with an increasing Atlantic Water volume and shoaling of the upper halocline in the Canada Basin, as described by McLaughlin et al. (2002). They attributed those changes to atmospheric circulation patterns associated with the Arctic Oscillation. While we see another DIC peak in the upper halocline following the high AO index at the turn of the millennium (Figure 10a), there does not appear to be a relationship between upper halocline DIC and the AO index after that time. A plot showing DIC at the core of the Pacific Water layer (Figure 10b) reveals that although DIC is generally increasing, there is no clear geographical pattern in that increase.

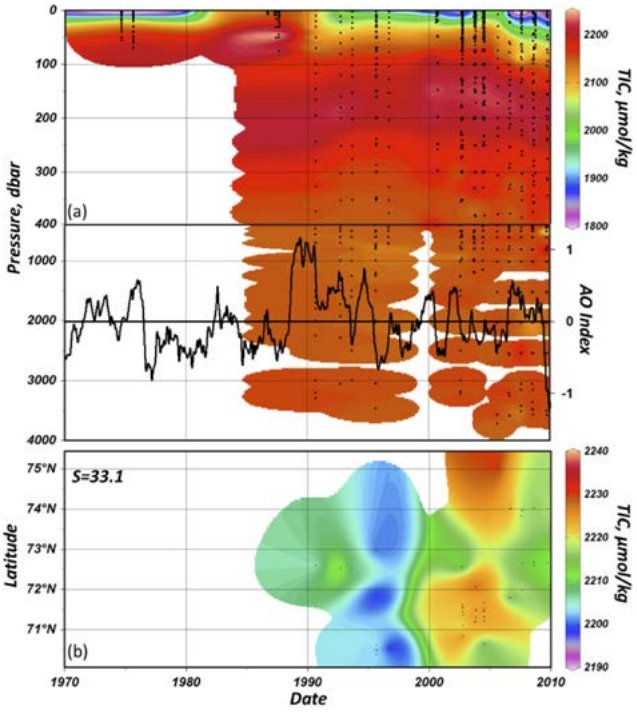

Figure 10. (a) Vertical DIC (TIC in the plot) distributions in the Beaufort Sea and Canada Basin through time overlaid with the Arctic Oscillation index, defined as the difference in sea level atmospheric pressure anomalies of opposite sign between the Arctic and 37-45 °N (data from the NOAA Climate Prediction Center). (b) DIC on the 33.1 salinity surface, corresponding to the Pacific water core and defined as salinities between 32.9 and 33.3. Note the different DIC color scales between (a) and (b).

The Beaufort Sea/Canada Basin area appears particularly inclined towards low $CaCO_3$ saturation states (Figure 11) because of sea-ice melt and the high-pCO_2SW Pacific water inflow (Yamamoto-Kawai et al., 2011; Chierici and Fransson, 2009). Our time series (Figure 12) shows that the aragonite saturation state (ΩAr) is highly variable but has generally been decreasing in both the upper halocline and deep waters.

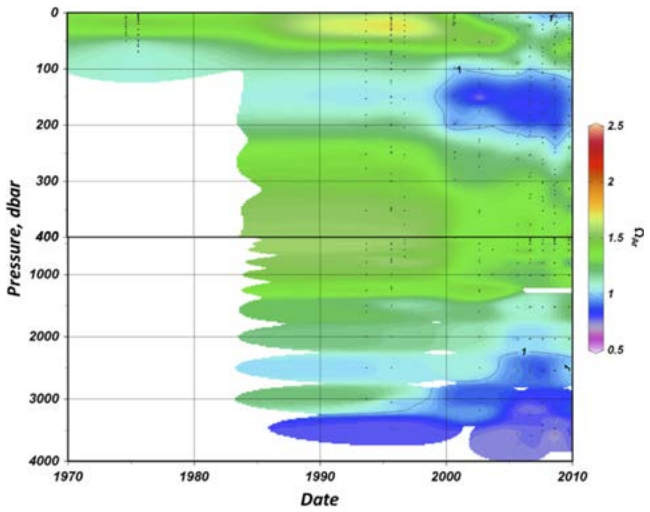

Figure 11. Vertical distributions of aragonite saturation state (ΩAr) in the Beaufort Sea and Canada Basin through time. ΩAr = 1 contour shown, indicating undersaturated zones in dark blue and purple. ΩAr calculated from measured TIC and total alkalinity using the CO2SYS MatLab version (van Heuven et al., 2011), with the carbonate system constants of Mehrbach et al. (1973), refit by Dickson and Millero (1987), and the KSO4 constant of Dickson (1990).

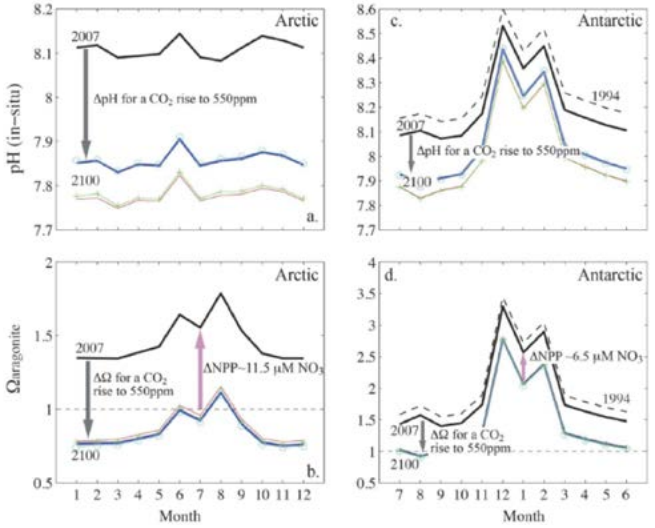

Figure 12. Estimated seasonal cycles in pH (top) and aragonite solubility (Ω) between the years 2007 and 2100 in Amundsen Gulf (left) and Prydz Bay Antarctica (right). The 2007 values (black lines) are compared to 2100 with (i) rising anthropogenic CO2 (solid blue), rising CO2 and warming (red), CO2 increase with freshening (pale blue with open circle), and rising CO2, warming, and freshening (green line with + symbols). (Shadwick et al. 2013).

Shadwick et al. (2013), using CFL/ArcticNet data, determined that Arctic waters are more vulnerable to acidification relative to Antarctic waters. The Arctic waters generally experienced greater freshening, lower nutrient loading, and a lower over-all alkalinity relative to Antarctic counterparts (Figure 12d). Data sets indicate that the Arctic system has a smaller capacity to buffer against decreases in pH. In addition to having higher seawater alkalinity, Antarctic seas may allow the mitigation of acidification through the removal of CO_2 by enhanced summer primary production.

Water Column DMS and N2O

An objective of the 2013 CCGS Amundsen cruise was to expand our understanding on the dynamics, distribution and production of DMS and distribution N_2O in the ArcticNet domain, in relation to sea ice cover and complex oceanic circulation of the region. Related, spectral irradiance dependence of sunlight effects (UV/PAR) and photo-acclimation response of phytoplankton cells on dimethylsulfide (DMS)

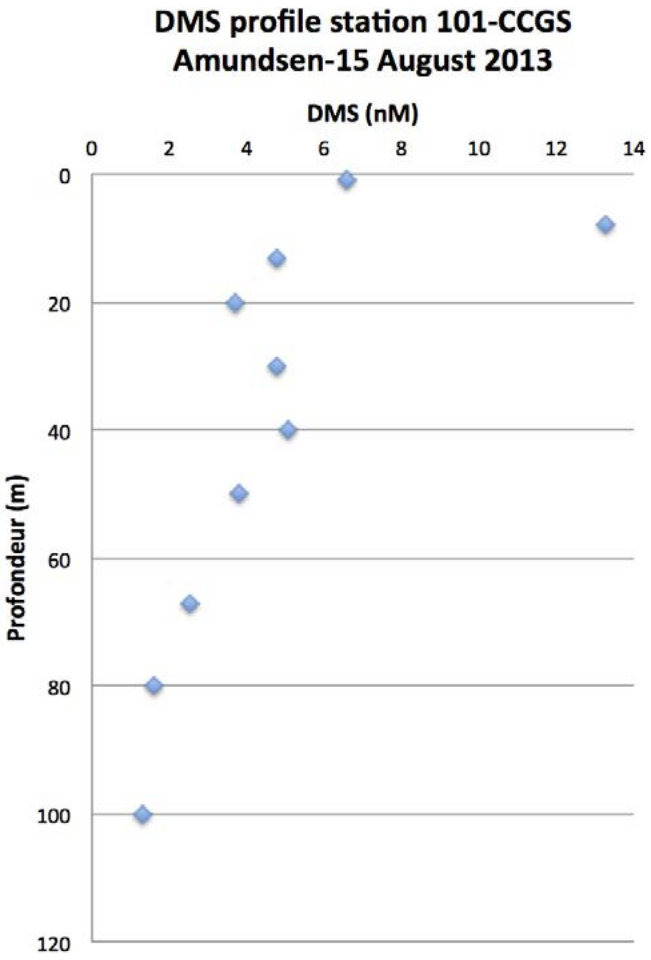

Figure 13. Vertical distribution of DMS concentrations at station 101 measured during the 2013 ArcticNet cruise.

production were examined. Sample analysis is underway. An example of the vertical distribution of DMS is shown as Figure 13.

Modelling of the Ocean Carbonate System

Changes in carbonate chemistry and acification under a changing climate have been evaluated using CanESM2 and 5 additional CMIP5 Earth System models. Steiner et al. (2013), discusses a sensitivity study with CanESM2 looking at enhanced CO_2 exchange in sea-ice areas in the Arctic Ocean (preliminary results were reported in last year's progress report).

The study has been expanded to analyze Arctic Ocean acidification. Steiner et al. (2014) summarize that projections under Representative Concentration Pathways (RCPs) 8.5 and 4.5 consistently show reductions in the bidecadal mean surface pH from about 8.1 in 1986-2005 to 7.7/7.9 by 2066-2085 in the Canada Basin, closely linked to reductions in the calcium carbonate saturation state Ω from about 1.4 (2.0) to 0.7 (1.0) for aragonite (calcite) for RCP8.5. The large but opposite effects of dilution and biological drawdown of DIC and dilution of alkalinity lead to a small seasonal amplitude change in Ω, as well as intermodel differences in the timing and sign of the summer minimum. These CMIP5 model results strengthen earlier findings, although large intermodel differences remain: differences of projected acidification changes are primarily related to sea ice retreat and responses of wind mixing and stratification.

Discussion

Sea Ice Carbon Chemistry and Sea Ice - Ocean Carbon Transport

Ikaite concentrations reported by Rysgaard et al. (2013a) are far higher (up to 10 times) than previously reported in Antarctica (Dieckmann et al., 2008) and four times higher than reported from Fram Strait (Rysgaard et al., 2012). The differences may reflect real differences in ikaite distribution between sites and seasons, or may be attributed to the quantification procedure. Undertaking analysis in the field makes our study unique. Both the concentration range and distribution agree with the FREZCHEM, supporting our supposition that ikaite concentration increases with decreasing temperature. The result that varying snow conditions may play a role in ikaite distribution may have implications for the air-ice CO_2 exchange, given modelled projections for increasing snow in a warmed Arctic.

Observations over the winter-spring transition by Brown et al. (2013) illustrate the influence of sea ice on the seasonal air-sea CO_2 exchange during this dynamic season. High brine pCO_2 at the beginning of the sampling program in early spring appeared to be associated with relatively small CO_2 efflux from the ice. As warming progressed, the gradual decrease in brine pCO_2 over the sampling period culminated with uptake of CO_2 into the melting ice, when brine was undersaturated with respect to the atmosphere. The decrease in pCO_2 throughout the sea ice column was associated with processes that influence TA and/or DIC in the brine, or processes which act directly on CO_2 itself. These processes include biological production of organic C, CO_2 degassing, brine dilution, and calcium carbonate (ikaite) precipitation.

Fransson et al. (2013) developed a conceptual model (Figure 14) of sea ice processes associated with the carbonate system, and their impacts on the under ice water. Processes are described for two periods fall to winter (Nov. to Dec.), early spring (March to May) and late spring (May to June). CO_2 outgassing and downward transport via brine rejection dominate

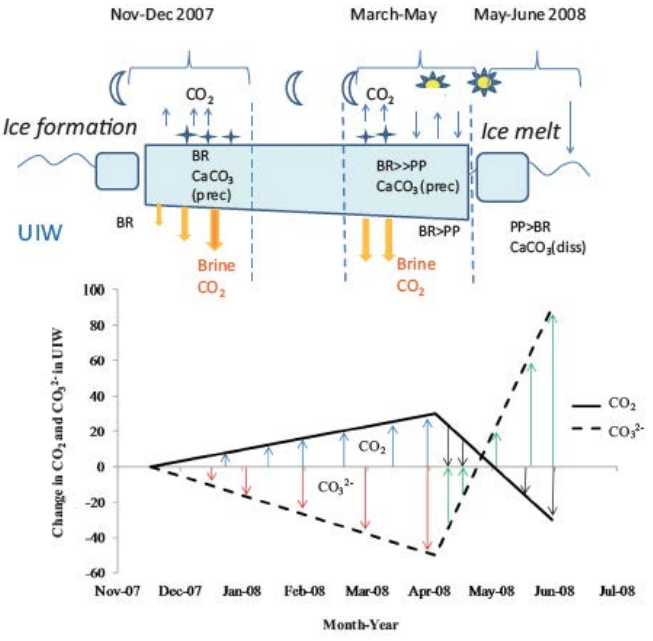

Figure 14. Conceptual model of sea ice processes and their impacts on properties in the under ice water. (Fransson et al., 2013).

the cold season – the latter giving rise to enhanced CO_2 (and DIC) in the under-ice water. Production of ikaite and bacterial respiration are key to high sea ice pCO2 over this period. The combination of primary production and dissolution of ikaite elevate CO_3-concentration, with a concomitant lowering of pCO_2 in the under-ice water. The later drives CO_2 uptake, as predicted by the sea ice carbon pump model.

Sea Ice DMS Production

High concentrations of DMS are produced within the seasonal ice environments studied. The variable distribution of DMS/O/P across sites suggests variable formation processes of these materials. The fraction of the DMS that eventually reaches the atmosphere is ice environments is still not known with any certainty. Preliminary results indicate that melt ponds contain DMS, although the processes dictating its occurrence are unclear. The implications are that melt ponds are also a source of primary and secondary aerosols. Recent observations have revealed the presence of extended under-ice phytoplankton blooms in the springtime Arctic (Arrigo et al., 2012; Mundy et al., 2013). Given an apparent relationship between under-ice blooms and DMSP (Galindo et al., 2014), uncertainties in the transformation mechanisms of DMSP and DMS in Arctic waters need to be resolved, and their contribution to upward emissions of DMS needs to be quantified. Collectively these uncertainties severely limit our ability to model climate-ocean feedbacks.

We observed significant differences in the vertical distribution of dimethylated suflur compounds in sea ice between the Arctic and the Antarctic. For instance, strong production of DMS and DMSP was observed in interior ice in ice-shelf influenced fast ice in the McMurdo Sound (Antarctica) due to the formation of platelet crystals, a process occurring in the Arctic. In the Arctic the DMS and DMSP production remains essentially restricted to the bottom ice. Platelet ice formation was shown to (1) favour the incorporation of potential strong DMSP producers such as dinoflagellates, (2) increase the environmental stresses these producers are exposed to. Also, a lower snow cover at the Arctic sampling sites relative to the Antarctic sampling sites, was shown to favour the photo-oxydation of DMS into DMSO in surface ice, leading to a generally greater proportion of DMSO in the Arctic sea ice cores. These two processes should have a direct impact on the contribution of sea ice to oceanic DMS emissions in both hemispheres.

Ocean Carbonate System

Observations reported here, and in previous reports, over a large range in ocean environments, indicates pCO_2sw and other CO_2 system parameters are subject to significant spatial and temporal variability. Variability in the carbon system parameters is in response to strong variability in environmental drivers. For example, variability in pCO_2sw within the CAA could be interpreted based on observed variation in variables including temperature, salinity, sea ice and river discharge (Pind, 2013). In the presence of sharp observed gradients in the variables that underpin air-sea exchange, it is imperative that the scale (space and time) of observation is commensurate with environmental variability. This is noted by Anderson et al. (2010) and demonstrated through the recent work of Forest et al. (2014). It is therefore arguable whether we will be able to effectively detect change without first developing the capacity to effectively retrieve those variables that underpin seawater pCO_2, and air-sea CO_2 exchange using satellite technology. Satellite-based remote sensing offers a means of regionally monitoring the drivers of the air-sea exchange at reasonable temporal frequency. Attempts to apply remote sensing to estimating air-sea CO_2 fluxes and budgets in Arctic seas have been frustrated by sea ice and the 'universality' of empirically-derived sea water pCO_2 relationships involving sea water temperature and water colour. The development of an Arctic-optimized pCO_2sw relationships and transfer velocity is a priority and will direct team research in the coming year.

Forest et al.'s (2014) carbon balance study in the southeastern Beaufort Sea demonstrates the utility of

integrating both inorganic and organic carbon pools and flows. Using a holistic approach, the authors were able articulate dependencies between the living and non-living systems, thereby connecting the atmospheric carbon pool to food web dynamics and water mixing. The study, along with that of Pind (2013) illustrates that the consequences of environmental change on CO_2 system parameters and carbon cycling need not be geographically uniform. This is evidenced, for example, by sharp discontinuities in pCO_2sw delineating river plume and ice melt in Pind (2013). Across the shelf break, upwelling, sea ice, river plume exert considerable influence on carbon cycling over comparatively small spatial (and temporal) footprints. Further, the study enforces the importance of lateral transport between coupled systems.

As shown in Figure 11 (Miller et al., submitted), the Beaufort Sea/Canada Basin area has low $CaCO_3$ saturation states. Therefore, even small direct anthropogenic acidification effects could tip the area into a biologically stressful state. Bottom water saturation states, which are also depressed by high hydrostatic pressure, are of particular concern, because benthic communities are generally less mobile than their pelagic neighbors, and organic matter remineralization in oxic surface sediments releases additional CO_2 to the bottom waters (Anderson et al., 2011).

Conclusion

Sea Ice Carbon Chemistry and Sea Ice - Ocean Carbon Transport

Ikaite is prevalent in seasonal sea ice and is found in the interstices between ice crystals. Concentration increases with decreasing temperature and hence is observed to following strong reduction in concentration with depth during the cold season. Because of this temperature dependency, changes in the timing and distribution of snow could have strong implications on air-sea ice carbon cycling. Field studies indicate that ikaite formation is a key process (along with bacterial respiration) in the vertical transport and ultimately export of CO_2 via brine, strongly impacting the inorganic carbon stocks in the under ice water. The effect of sea ice on the inorganic carbon system of the upper water column varies with seasonal progressions from winter to summer.

Sea Ice DMS Production

Overall, our results from 2013 field season confirm the diversity of sources of DMS in the Arctic and highlight the extremely high concentrations observed at the sea ice base during the spring seasons, and within the under-ice water column. The presence of appreciable concentration of DMS in melt pounds heralds their possible role as an important DMS source for the Arctic atmosphere during the melt season. Factors controlling the production of DMS in melt ponds are still not understood, but may be linked to the low salinity and high light environment experienced by the microalgae trapped in this peculiar environment.

Ocean Carbonate System

The collection of inorganic carbon data from the Canadian Arctic is increasing our understanding of the carbon fluxes in this climate-sensitive area. Both long-term time series and seasonal data underscore the fact that this area is currently undergoing dramatic changes and that the processes contributing to those changes are also in transition. Striking in the data sets are the high level of variability of surface water pCO_2, and other components of the CO_2 system.

Key results of a carbon balance the study are that: climate change is exacerbating an existing biological gradient across the shelf-basin system, the Mackenzie Shelf acts as a weak sink for atmospheric CO_2, the effect of CO_2 transfer to the atmosphere associated with shelf break upwelling can be mitigated in the presence of enhanced diatom production associated with the concomitant introduction of nutrients to the surface layer. In studying the regional marine carbon balance it is important that biological and physical systems be considered.

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