



*Supplement of*

## **Underestimation of oceanic carbon uptake in the Arctic Ocean: ice melt as predictor of the sea ice carbon pump**

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# S1 Theoretical Derivation

How does storage of carbon in sea ice impacts the partial pressure of  $\text{CO}_2$ ? To answer this question, one can consider the upper layer of the ocean, partially covered by sea ice forming and melting. The atmospheric  $p\text{CO}_2$  and wind speed at 10m height are known, as are the ice concentration and melting-freezing rate and the ocean surface temperature and salinity. The surface ocean  $p\text{CO}_2$  (in  $\mu\text{atm}$ ) would vary along the seasonal cycle due to changes in temperature, salinity, air-sea gas exchange, dilution and concentration related to surface freshwater fluxes (either from precipitation-evaporation, or from ice melt and formation). Biology would also impact carbonate properties due to production and respiration. Finally, advection and mixing could also modify surface properties.

Two scenarios emerge, with for sole difference the storage and release of alkalinity and dissolved inorganic carbon (DIC) by sea ice.

- The first scenario, called CTRL (for control), corresponding to current Earth System Models, does not have any carbon in sea ice. In this case, sea ice melt and formation only dilutes and concentrate carbonate properties in underlying seawater, decreasing or increasing  $p\text{CO}_2$  accordingly.
- The second scenario, called EXP (for experiment) corresponding to a more realistic approach, stores some alkalinity and DIC in sea ice. In this case, when ice melts or freezes, on top of diluting or concentrating carbonate properties in underlying waters, it also creates a flux of alkalinity and DIC between ice and ocean. This will further modifies  $p\text{CO}_2$  due to chemical equilibration processes.

The difference in  $p\text{CO}_2$  between those two scenarios will in turn modify the magnitude of the air-sea gas exchange.

## S1.1 Differential Equation

From a mathematical perspective, the difference in  $p\text{CO}_2$  between both scenarios would translate as follow. Initial conditions are identical in both scenarios, so  $p\text{CO}_2^{\text{ICE}}(t=0) = p\text{CO}_2^{\text{CTRL}}(t=0)$ . If  $f$  is the difference in  $p\text{CO}_2$  between both runs at each time  $t$ , Then:

$$\forall t, p\text{CO}_2^{\text{ICE}}(t) = p\text{CO}_2^{\text{CTRL}}(t) + \int_0^t f(s)ds$$

If non-linearities are neglected, temperature, salinity and biology changes would impact  $p\text{CO}_2$  similarly in both runs. DIC and Alk changes due to dilution and concentration or mixing would have the same impact on  $p\text{CO}_2$  in both cases as well. So all those processes can be excluded from  $f$ .

The impact of the carbon flux between ice and ocean on  $p\text{CO}_2$  needs be taken into account in  $f$ .  $[\text{Alk}]_{\text{ice}}$  and  $[\text{DIC}]_{\text{ice}}$  are the alkalinity and DIC concentration inside the sea ice, respectively, in  $\text{mmol m}^{-3}$  and are assumed to be spatially constant. The flux of alkalinity and DIC between the ice and the ocean is assumed to be directly proportional to the freshwater flux between ice and ocean,  $\mathcal{F}_{\text{FW}}^{\text{ice-sea}}$ , in  $\text{m s}^{-1}$ . In this case, the ice-ocean flux of alkalinity would be equal to  $[\text{Alk}]_{\text{ice}} \mathcal{F}_{\text{FW}}^{\text{ice-sea}}$ , and that of DIC would be  $[\text{DIC}]_{\text{ice}} \mathcal{F}_{\text{FW}}^{\text{ice-sea}}$ .

Moreover, this carbon flux, by modifying the surface water  $p\text{CO}_2$ , would induce a difference in the air-sea gas exchange between both scenarios, written as:

$$\begin{aligned} \Delta \mathcal{F}_{\text{CO}_2}^{\text{air-sea}} &= \mathcal{F}_{\text{CO}_2}^{\text{air-sea,ICE}} - \mathcal{F}_{\text{CO}_2}^{\text{air-sea,CTRL}} \\ \Delta \mathcal{F}_{\text{CO}_2}^{\text{air-sea}} &= -k_g(t) S_{\text{CO}_2}(t) \lambda(t) (p\text{CO}_2^{\text{ICE}}(t) - p\text{CO}_2^{\text{CTRL}}(t)) \end{aligned}$$

where  $k_g$  is the gas piston velocity ( $\text{m s}^{-1}$ ),  $S_{\text{CO}_2}$  is the  $\text{CO}_2$  solubility ( $\text{kg m}^{-3} \text{Pa}^{-1}$ ) and  $\lambda = 1 - [\text{Ice}]$  the lead fraction (unitless). The air-sea carbon flux  $\Delta \mathcal{F}_{\text{CO}_2}^{\text{air-sea}}$  and the ice-ocean carbon flux are the only two non-negligible processes that need to be accounted for in  $f$ .

Since  $f$  corresponds to the change in  $p\text{CO}_2$  due to those fluxes, we need to multiply the two different fluxes by the slope of  $p\text{CO}_2$  in the alkalinity or DIC domain,  $\frac{\partial p\text{CO}_2}{\partial \text{Alk}}$  and  $\frac{\partial p\text{CO}_2}{\partial \text{DIC}}$ . Note that those two terms are fluxes at the surface and therefore have units of  $\mu\text{atm m}^{-2} \text{s}^{-1}$ . To match the  $p\text{CO}_2$  which has units of  $\mu\text{atm}$ , we can focus on surface layer of thickness  $H_0$  (m).

Then

$$pCO_2^{ICE}(t) = pCO_2^{CTRL}(t) + \int_0^t \frac{\partial pCO_2}{\partial DIC} \frac{1}{H_0} \Delta \mathcal{F}_{CO_2}^{air-sea}(s) ds + \int_0^t \left( \frac{\partial pCO_2}{\partial Alk}(s) \frac{1}{H_0} [Alk]_{ice} \mathcal{F}_{FW}^{ice-sea}(s) + \frac{\partial pCO_2}{\partial DIC}(s) \frac{1}{H_0} [DIC]_{ice} \mathcal{F}_{FW}^{ice-sea}(s) \right) ds$$

Then, with the notation  $g(s) = \frac{\partial pCO_2}{\partial Alk}(s)[Alk]_{ice} + \frac{\partial pCO_2}{\partial DIC}(s)[DIC]_{ice}$  and  $\Delta pCO_2^{i-c} = pCO_2^{ICE} - pCO_2^{CTRL}$ , we get:

$$\Delta pCO_2^{i-c}(t) = \int_0^t \frac{\partial pCO_2}{\partial DIC} \frac{1}{H_0} \Delta \mathcal{F}_{CO_2}^{air-sea}(s) ds + \int_0^t \frac{1}{H_0} g(s) \mathcal{F}_{FW}^{ice-sea}(s) ds \quad (1)$$

So, using the gas transfer formulation:

$$\Delta pCO_2^{i-c}(t) = \int_0^t -\frac{\partial pCO_2}{\partial DIC} \frac{1}{H_0} k_g(s) S_{CO_2}(s) \lambda(s) \Delta pCO_2^{i-c}(s) ds + \int_0^t \frac{1}{H_0} g(s) \mathcal{F}_{FW}^{ice-sea}(s) ds$$

While proving the differentiability of all those variables is beyond the scope of this study, it can be noted that they are continuous and unlikely to present any break or cusp. We will therefore assume that we can safely differentiate the previous expression to obtain the following linear differential equation of the first order:

$$\frac{\partial \Delta pCO_2^{i-c}(t)}{\partial t} = -\frac{\partial pCO_2}{\partial DIC} \frac{1}{H_0} k_g(t) S_{CO_2}(t) \lambda(t) \Delta pCO_2^{i-c}(t) + \frac{1}{H_0} g(t) \mathcal{F}_{FW}^{ice-sea}(t) \quad (2)$$

## S1.2 Solving an EDL1

According to the Cauchy-Lipschitz theorem, the Ordinary Differential Equation of order 1  $y' + ay = b$  has a unique general solution (Cauchy, 1844, Chapter 26) of the form:

$$y(t) = e^{-A(t)} \left( K + \int_{t_0}^t b(s) e^{A(s)} ds \right)$$

with  $A(t)$  a primitive of  $a(t)$  over the proper interval and  $K$  a constant that depends on initial conditions (at  $t_0$ ).

So for equation 2, a general solution would be of the form:

$$\Delta pCO_2^{i-c}(t) = e^{-A(t)} \left( K + \int_0^t \frac{1}{H_0} g(s) \mathcal{F}_{FW}^{ice-sea}(s) e^{A(s)} ds \right)$$

with  $A(t)$  a primitive of  $\frac{\partial pCO_2}{\partial DIC} \frac{1}{H_0} k_g(t) S_{CO_2}(t) \lambda(t)$ , written as  $A(t) = \int_0^t \frac{\partial pCO_2}{\partial DIC} \frac{1}{H_0} k_g(s) S_{CO_2}(s) \lambda(s) ds + \alpha$ .

Since the initial conditions for the control and sensitivity runs are identical,  $\Delta pCO_2^{i-c}(t=0) = 0 \Rightarrow K = 0$ .

So  $\forall t, \Delta pCO_2^{i-c}(t) = e^{-A(t)} \int_0^t \frac{1}{H_0} g(s) \mathcal{F}_{FW}^{ice-sea}(s) e^{A(s)} ds$

## S1.3 Uptake difference

Inserting this solution in equation 1 and rearranging, we get:

$$\int_0^t \frac{1}{H_0} \frac{\partial pCO_2}{\partial DIC} \Delta \mathcal{F}_{CO_2}^{air-sea}(s) ds = e^{-A(t)} \int_0^t \frac{1}{H_0} g(s) \mathcal{F}_{FW}^{ice-sea}(s) e^{A(s)} ds - \int_0^t \frac{1}{H_0} g(s) \mathcal{F}_{FW}^{ice-sea}(s) ds$$

If we consider  $H_0$  to be constant, we can move it out of the integrals and make it disappear. Similarly, if  $\frac{\partial pCO_2}{\partial DIC}$  is constant and non-null, we can move it outside the integral and divide both side by it.

We can then rearrange as follow:

$$\int_0^t \Delta \mathcal{F}_{CO_2}^{air-sea}(s) ds = \Delta \mathcal{F} = \frac{1}{\frac{\partial pCO_2}{\partial DIC}} \int_0^t g(s) \mathcal{F}_{FW}^{ice-sea}(s) \left( e^{A(s)-A(t)} - 1 \right) ds$$

Note that we can show that if  $\lambda(t) = 0$ , then  $\Delta \mathcal{F}_{CO_2}^{air-sea}(t) = 0$ . So we find again the result that the difference in uptake only depends on melting flux (with the assumption that if  $[Ice] \neq 1$ , ice is melting).

Using realistic alkalinity, DIC and  $pCO_2$  values for the Arctic Ocean ( $[Alk]_{sw} = 2300 \text{ mmol m}^{-3}$ ,  $[DIC]_{sw} = 2100 \text{ mmol m}^{-3}$ ,  $[Alk]_{ice} = 540 \text{ mmol m}^{-3}$ ,  $[DIC]_{ice} = 300 \text{ mmol m}^{-3}$  and  $pCO_2 = 280 \text{ } \mu\text{atm}$  and Revelle and alkalinity factors of 14 and -13.3 respectively), it yields  $g(t) \approx -314$  which is negative. All the terms inside the integrand of  $A(t) = \int_0^t \frac{\partial pCO_2}{\partial DIC} \frac{1}{H_0} k_g(s) S_{CO_2}(s) \lambda(s) ds + \alpha$  are positive, meaning  $A(t)$  is monotonously increasing with  $t$ . So  $t > s \iff A(t) > A(s)$ . This means that  $e^{A(t)-A(s)} < 1$ . Therefore, the sign of the integrand is determined by the sign of  $\mathcal{F}_{FW}^{ice-sea}$ .

## S1.4 Idealized case

If we make the assumption that wind speed and solubility are constant for the time period when ice is not fully covered the water (i.e.  $\forall t, \frac{\partial pCO_2}{\partial DIC} \frac{1}{H_0} k_g(t) S_{CO_2}(t) \equiv \kappa$ ), we can rewrite:

$$\begin{aligned} A(t) &= \int_0^t \frac{\partial pCO_2}{\partial DIC} \frac{1}{H_0} k_g(s) S_{CO_2}(s) \lambda(s) ds = \kappa \int_0^t \lambda(s) ds \\ &= \kappa \Lambda(t) \end{aligned}$$

with  $\Lambda(t) = \int_0^t \lambda(s) ds$ . Similarly, we can consider that  $\forall t, g(t) \equiv \gamma$ . Then:

$$\int_0^t \Delta \mathcal{F}_{CO_2}^{air-sea}(s) ds = \frac{1}{\frac{\partial pCO_2}{\partial DIC}} \gamma \int_0^t \mathcal{F}_{FW}^{ice-sea}(s) \left( e^{\kappa(\Lambda(s)-\Lambda(t))} - 1 \right) ds \quad (3)$$

This expression can be evaluated with the ice concentration and the freezing-melting flux used to force the 1D model, described in Section 3.a. To do so, values for  $\gamma$  and  $\kappa$  need to be determined. Using values from Section S1.3, we can set  $\gamma = -314 \text{ } \mu\text{atm}$ . Similarly, relying on values from Takahashi et al. [1993],  $\frac{\partial pCO_2}{\partial DIC} = 1.9 \text{ } \mu\text{atm mol}^{-1} \text{ m}^3$ . Following Wanninkhof [2014], their eq. 6, and using an average squared wind speed of  $50 \text{ m}^2 \text{ s}^{-2}$  and a surface layer of 1m, we can set  $\kappa = 7.7 \times 10^{-4} < U^2 > \frac{\partial pCO_2}{\partial DIC} \frac{1}{H_0} = 7.3 \times 10^{-2}$ .

## S2 Supplementary Figures

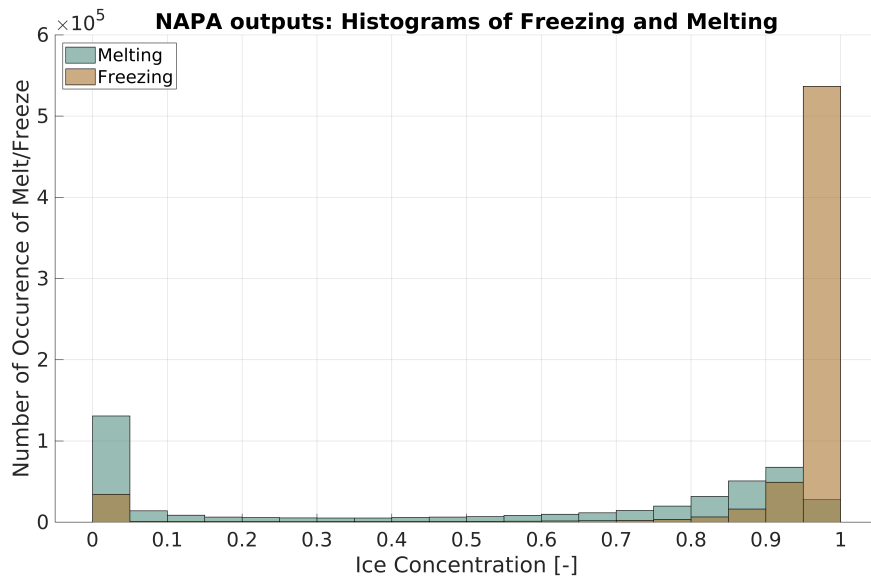


Figure S1: Histograms of freezing and melting as a function of ice concentration. calculated with daily outputs for the NAPA model, from 2014 to 2019, over the 732 grid cells used for the 1D simulation ensemble.

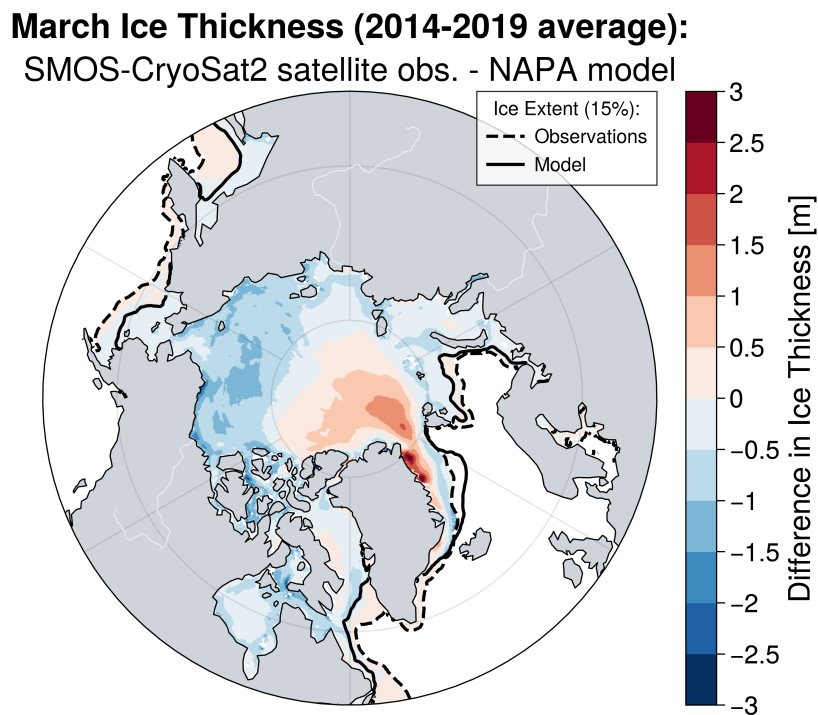


Figure S2: Validation of sea ice concentration: comparison between merged CryoSat2-SMOS satellite observations and NAPA model. Colours show observations minus model ice thickness during the month of maximum extent (March), averaged over 2014-2019. The contours show the ice extent, calculated as the 15% isoline for ice concentration. For details about satellite product, refer to Ricker et al. (2017).

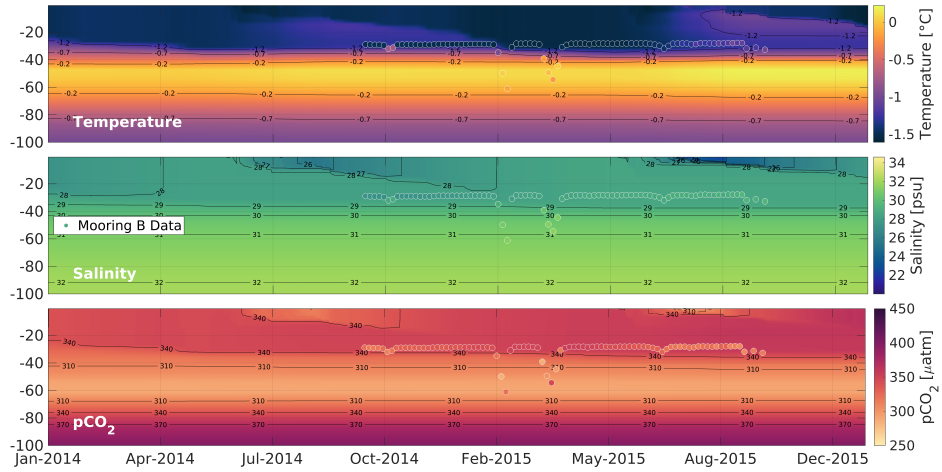


Figure S3: Comparison of 1D numerical model outputs with mooring observations from DeGrandpre et al. (2019) in the Beaufort Gyre station location (78°N, 150°W).

## Bibliography

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