

Response

We thank Dr. Cai for his very helpful comments that have improved the paper. We have addressed all the comments below (as blue, Arial 11 font in the supplemental file) and revised the paper accordingly. In the online version of our response, we have added the WLC to denote referee comment and NRB/JTM response.

We have added two new figures that we hope will aid the reader in following the discussion in Section 5. We have also added an Appendix in this response but not in the paper that illustrates the likely processes that influence the inorganic carbon system in the Barents Sea. The confidence levels for the impact of many processes on the Arctic CO₂ sink or source are uncertain in the near-term and are highly uncertain over the next century (Table 2) due to the limitations of data that exist in the Arctic at present.

WLC. Bates and Mathis have done an excellent job in synthesizing the current status of CO₂ air-sea exchange rates and biogeochemistry in the Arctic Ocean shelves and basins. They also went through the processes that determine the fluxes, and the effects of possible current/future changes including the ocean acidification will do to these processes. This paper will have a good impact on the future of the Arctic C cycling research. Overall the paper is well written and very informative, and I have enjoyed reading it. The Introduction section (Sect. 1) is straightforward and tells readers the structure of the paper (I like this style). Sect. 2 provides some useful physical and biogeochemical background for understanding various issues to be discussed later. In particular the three categories (inflow, interior, and outflow shelf) are very helpful in understanding the differences in CO₂ uptake flux in various shelves that will be discussed in Sect. 4. Sect. 3 discusses historical data (which is necessary) and CO₂ chemistry in general. While it is helpful, I find the textbook content not all that useful. See further comment in Specific comments 1. Sect. 4 provides a complete and excellent synthesis of the state of knowledge. I learned a lot by reading this part twice. I also enjoyed reading Sec. 5 very much as it provides an excellent synthesis on how the current climate changes in the Arctic may affect air-sea CO₂ flux. The authors have made some visionary statements. However, I do feel they, occasionally, speculated too much (see Specific comments 2). Ocean acidification is all new in the Arctic Ocean research, and the authors have laid a good foundation for this field in the paper. But again, I feel not all the words are needed (see Specific comments 1). In several places, I wish the discussion can be more quantitative (see Specific comments 3) and more balanced (see Specific comments 4 and 5). I feel the authors may have put a little bit too much confidence on the factors that would increase CO₂ uptake. But there are many factors that could go the other way and deserve the attention as well (see Specific comments 4). Overall, an excellent review paper!

Specific comments 1. The authors are gifted in introducing and explaining questions to readers (which are often very helpful), but, occasionally, they seem to have the tendency to be excessive in providing background and textbook information. None of the equations (R1-R5) is used in the paper, thus not all are necessary. Also, for example, p.6703, lines 6 to 11, repeat what has been already said in lines 3 to 5. I feel that, at least, lines 6 to 11 can be shorten to half (negative delta(pCO₂), CO₂ undersaturation, CO₂ uptake, and CO₂ sink really mean the same thing but they occurred in the same sentence). I appreciate the authors' intention to make the reading smooth and easy, but feel they could trust the readers a bit better. Another example

is on the ocean acidification, I feel the leading paragraph before 6.1 is all correct but not have much to do with the Arctic situation. It can easily be shortened to half.

NRB/JTM response.

We agree with Wei-Jun that the equations (R1-R5) on 6701 and 6702 are not directly referenced again in the paper. But we felt that it was useful for the reader (who might not be so familiar with the carbonic acid system) as a means of introduction. We have also referenced the Zeebe and Wolf-Gladrow, 2001 and Dickson et al., 2007 manual in an attempt to direct readers who want to know more about the carbonic acid system. Hopefully, we have struck a reasonable balance. We also discuss DIC, TA, $p\text{CO}_2$ and Ω throughout the paper and use DIC and TA data to calculate $p\text{CO}_2$ (as many others have done) in the paper and for the Appendix 1 where the Barents Sea is treated as a case study.

We also slightly reduced the paragraph on $\Delta p\text{CO}_2$, uptake/release, sink/source, etc. On careful consideration, we felt it was still important to define $\Delta p\text{CO}_2$, in terms of negative or positive, and flux as sink/source terms for those readers perhaps less familiar with CO_2 flux. Again, hopefully we struck a reasonable balance.

WLC. Specific comments 2. The authors have made some visionary statements, but occasionally, they speculated too much. One clear example is in p.6722, lines 10-14. The release of alkalinity could be important in shallow water environment (in the context of influence surface water $p\text{CO}_2$). But no evidence so far has suggested its importance in any deep water environment (again, in the context of influence surface water $p\text{CO}_2$). Speculation on this possibility in the Arctic Ocean Basin, I feel, is appropriate at community discussion but not in a published paper (although it is also part of the community discussion).

NRB/JTM response. We have clarified this statement to highlight the potential of this process to alter the inorganic carbon system of "shelf waters". We have added the caveat that no evidence has yet been forthcoming about the potential influence of this process for Arctic shelves. It may not be significant!

WLC. Specific comments 3. Discussion can be more quantitative. For example, in p. 6707, CO_2 input from the air is cited as one reason for $p\text{CO}_2$ increase in fall and winter (probably not winter as ice will block the inflow of CO_2). I agree. I suggest make this a bit more quantitative by estimating how much the air-sea flux would change $p\text{CO}_2$ in the surface (or mixed-layer) water (probably in one high flux and shallow water area, Chukchi, and in one low flux/deep water area). $p\text{CO}_2$ air-sea can be estimated by estimating $\Delta(\text{DIC})_{\text{air-sea}}$ (i.e., time integrated CO_2 flux) during the ice open period and assuming a constant TA. Or the Revelle equation can be used for this purpose.

NRB/JTM response.

It is with some hesitation that we expand on the discussion in a more quantitative manner. In the Barents Sea, Kaltin *et al.*, 2002 and Omar *et al.*, 2007 have assessed the relative contribution of gas exchange and new production to seasonal changes in $p\text{CO}_2$. We have revised the text in section 4.1 to reflect this previous work.

Ideally, we would need a very good physical-biological coupled model of the Arctic to quantify the relative importance of these terms for the inorganic carbon system. We feel it would be very difficult to be more quantitative in attributing seasonal changes in $p\text{CO}_2$ to different processes within the scope of this paper. It would highly speculative to do so for the Siberian, Beaufort and Canada archipelago shelves due to the limitation of data.

However, in response, we have constructed a simple carbon mass balance in a case study of the Barents Sea using data and framework of Kaltin *et al.*, 2002 and Omar *et al.*, 2007. We strongly felt that this would interrupt the flow of the paper and that we should not include the appendix below in the paper (better served in another paper, for example). There are lots of caveats and speculations in preparing such a mass balance and so quantitative attribution must

be used with extreme caution. Although different scenarios and outcomes can be demonstrated, for example, by changing rates or starting points, warming/cooling, new production and gas exchange appear to be dominant controls on the inorganic carbon cycle (as Kaltin *et al.*, 2002 and Omar *et al.*, 2007 suggested previously). There remains much uncertainty about the entrainment or horizontal transport terms, for example.

Appendix 1. Case study of Barents Sea seasonal changes in $p\text{CO}_2$.

As a framework for understanding the principal processes that influence the seasonal changes in $p\text{CO}_2$ and DIC, we provide a case study of the Barents Sea using data and interpretations of Kaltin *et al.* (2002) and Omar *et al.*, (2007) primarily. In the Barents Sea, the mixed layer depth shoals from ~150 m to 40 m, and temperatures increase from ~5°C to ~9°C between later winter/early spring and mid-summer (Kaltin *et al.*, 2002), rebounding to winter values through the fall period. Seasonally seawater $p\text{CO}_2$ decreases from ~350 μatm in early spring to ~250 μatm by mid-summer seasonally rebounding to ~350 μatm during fall and winter (Omar *et al.*, 2007). Using mean DIC, TA and salinity values (Kaltin *et al.*, 2002), and gas exchange and new production rates of 20 $\text{mmoles C m}^{-2} \text{d}^{-1}$ and 60 $\text{g C m}^{-2} \text{season}^{-1}$ (Kaltin *et al.*, 2002; Omar *et al.*, 2007; Macdonald *et al.*, 2009), carbon mass balance approaches can be used to estimate the impact of temperature changes, gas exchange, new production and other processes on mixed layer $p\text{CO}_2$ and DIC, for the spring-summer period ($\Delta_{\text{spring-summer}}$) and summer-fall period ($\Delta_{\text{summer-fall}}$) (Table A1). There are many caveats to this approach with the strong proviso that the rates for different processes must be interpreted with caution. During the spring-summer period, seawater $p\text{CO}_2$ appears primarily influenced by warming, CO_2 uptake via gas exchange and new production. To achieve mass balance, other processes have to be invoked to decrease $p\text{CO}_2$ with little change of DIC. We assumed a constant alkalinity in the mass balance and thus horizontal advection of seawater or input of freshwater with lower DIC to TA ratio and excess TA relative to springtime Barents Sea water could decrease $p\text{CO}_2$ without changing DIC. Calcification by coccolithophores in the Barents Sea (Signorini and McClain, 2009) would likely have an opposite effect. During the summer-fall period, seawater $p\text{CO}_2$ appears primarily influenced by cooling, and continued CO_2 uptake via gas exchange. Other processes include deepening of the mixed-layer that entrains DIC from subsurface waters and increase seawater $p\text{CO}_2$.

Table A1. Seasonal changes in mixed layer $p\text{CO}_2$ and DIC for the Barents Sea and, attribution to different physical and biological processes through a carbon mass balance approach.

	Units	Spring	Summer	Fall	$\Delta_{\text{spring-summer}}$	$\Delta_{\text{summer-fall}}$
Temperature	°C	~5	~9	~5	+4	-4
Mixed layer depth	m	~150	~40	~150	-110	+110
Salinity	n/a	35.0	35.0	35.0	0	0
Seawater $p\text{CO}_2$	μatm	~350	~250	~350	-100	+100
DIC	$\mu\text{moles kg}^{-1}$	2142		2048	2142	-94 +94
TA	$\mu\text{moles kg}^{-1}$	2309		2309	2309	0 0
Gas exchange	$\text{mmoles C m}^{-2} \text{d}^{-1}$	3	20	3	n/a	n/a
New Production	$\text{g C m}^{-2} \text{season}^{-1}$	n/a	60	n/a	60	0
a. Seasonal impacts on $p\text{CO}_2$					(units μatm)	
Temperature change					+75	-75
Gas exchange					+60	+60
New production					-175	?
Other processes					-70	+115
Net change					-100	+100
b. Seasonal impacts on DIC					(units $\mu\text{moles kg}^{-1}$)	
Temperature change					0	0

Gas exchange	+25	+25
New production	-125	?
Other processes	+6	+71
Net change	-94	+94

WLC. Specific comments 4. I feel the authors put more confidence on the factors that would increase CO₂ uptake and thus more discussion on them. But there are many factors that could go the other way. For example, stronger upwelling can bring more nutrient, but also high DIC and pCO₂ subsurface water. Same is true with the increased inflow of Pacific water after warming. The mixing with high pCO₂ water should be taken into account. Other factors such as the amount of nutrient input vs OC input from river will also provide a more balanced view on river influence.

NRB/JTM response.

Where possible, we put confidence indicators for the factors that could increase or decrease CO₂ uptake. In response to the comment by Referee Lisa Miller, we have added a few qualifier statements and added a new “caveat section”, 4.5.1. The new caveat section discusses the limited data, the potential for winter outgassing (particularly in the Laptev and east Siberian Seas) but also the potential for winter ingassing due to brine rejection during deep winter formation (as discussed by Omar et al., 2005). We simply do not know how the balance of wintertime polynya/lead outgassing and ingassing contributes to the overall annual air-sea CO₂ exchange.

In section 5, we use the two new figures as a framework for discussing the processes that could increase or decrease the CO₂ sink in the Arctic. We have added statements that provide consideration of the processes that could increase

In summary, at present the Arctic appears to be a CO₂ sink. In the near-term also a likely yes, but beyond the next decade? The Arctic is in rapid transition and the flux term could easily reverse.

WLC. Specific comments 5. Sect. 3.4 and p. 6704. I feel the references of Cai and Dai 2004) and Cai et al. (2006) together with those of Borges et al. (2005) and Chen and Borges (2009) should be used for two reasons. First, Cai and Dai (2004) was the first to point out the latitudinal distribution pattern of uptake CO₂ in the high-mid latitude shelves and release (or neutral) in lower latitudes. Second, the Cai et al. (2006) paper provides a difference approach, province-based method, to synthesis shelf CO₂ air-sea flux. 1.Cai, W.-J. and Dai, M. 2004. A Comment on “Enhanced open ocean storage of CO₂ from shelf sea pumping.” Science, 306, 1477c. 2.Cai, W.-J., M. Dai, and Y. Wang. 2006. Air-sea exchange of carbon dioxide in ocean margins: A province-based synthesis, Geophysical Research Letters, 33, L12603, doi:10.1029/2006GL026219.

NRB/JTM response. We agree that the references to Cai and Dai, 2004, Cai et al., 2006 and Chen and Borges, 2009 are useful in the brief discussion of net metabolism of continental shelves. Both papers suggested by the reviewer have been added to the paper.

WLC. Other minor issues: Abstract: Well written but concentrated too much on the CaCO₃ saturation issue. The summary is actually a better one (more balanced)

NRB/JTM response. We agree and have reduced this emphasis.

WLC. p.6700, L13, Tanhua et al., 2009 is not the refs.

NRB/JTM response. We have added the Tanhua et al., 2009 reference to the revised paper.

WLC. p.6701. R1, “-2” should “2-“

NRB/JTM response. Corrected in the revised text

WLC. p.6701. R3, there is no need to –[minor species] (or even use []), it is sufficient to say +minor species, which could be positive or negative.

NRB/JTM response. Corrected in the revised text

WLC. p.6701. L18, please do not use italic p in pH or pK (to be consistent with what is used in chemistry and to differential it from <italic-p>CO₂). While there is no rule that p or italic p must be used, I think it is good to recognize the difference between them (i.e., in pH and pCO₂).

NRB/JTM response. Corrected throughout in the revised text.

WLC. p. 6702, line 10, delete + in R4.

NRB/JTM response. Corrected in the revised text

WLC. p.6705, line 14, add (Fig.1) to the end (after freshwater inputs). Do the same for all subsections/shelves.

NRB/JTM response. We have added as suggested.

WLC. p.6712, line 21, delete one “principally”?

NRB/JTM response. Corrected in the revised text

WLC. p.6715, line 25 and 27. use of “-“ or “+” for flux is a bit confusion. Strictly to say, when a direction is given (such as influx or degassing), then it should be positive.

NRB/JTM response. The direction of gas exchange is given and the negative sign is deleted on page 6714 (line 1) as well as page 6715.

WLC. Lots of formatting issues and errors in the references.

NRB/JTM response. We have to apologize for the errors in the citation list and the irritation it will have caused. In response, we have carefully revised the references, including misspelling, wrong author order, issue and doi numbers etc. We have also added a few citations in response to comments from this and other reviewers. We have added the following papers by: Alonso-Saez et al., 2008; Cai and Dai, 2004; Cai et al., 2006; Dieckmann et al., 2008; Dmitrenko et al., 2005; Fransson et al, 2009; Garneau et al., 2008, Gow and Tucker, 1990; Tanhua et al., 2009; Rysgaard et al., 2008; Signorini and McClain, 2009; Trembley et al., 2009; Winsor and Bjork, 2000, Yamamoto-Kawai et al., 2009.