

ANSWERS TO REVIEWERS

We thank the Referees for their interest in our work and for helpful comments that will greatly improve the manuscript and we have tried to do our best to respond to the points raised. The Referees have brought up some good points and we appreciate the opportunity to clarify our research objectives and results.

As indicated below, we have checked all the general and specific comments provided by the Referees and have made necessary changes accordingly to their indications.

Anonymous Referee #1

General comment:

C1: I would recommend to structure the paper according to the observed confronting water masses ("Siberian Coastal Current" and "open marine water masses") instead of discussing first the West-Siberian, then the East-Siberian and finally the interannual variability within these basins.

A1: Based on our multi-years investigations on the Siberian Shelf, which results have been published in (Semiletov et al., 2005, 2007; Pipko et al., 2008; Anderson et al., 2009 etc.) and literature data (Antonov, 1957; Gorbunov, 1957; Shpaikher et al., 1972; Nikiforov and Shpaikher, 1980; Münchow et al., 1999; Weingartner et al., 1999; Steel and Ermold, 2004; Dmitrenko et al., 2005, etc.) we have considered the shelf of the East Siberian Sea (ESS) as a transition zone between Siberian shelf water and Pacific-derived water. A number of investigators noted that Siberian Coastal Current (SCC) in the ESS was not found (Gorbunov, 1957; Shpaikher et al., 1972; Münchow et al., 1999; Weingartner et al., 1999). For example, drifter observations in the anticyclonic summer of 1995 showed distinct water transport from the ESS toward the west and absence of the SCC. Our data of summer 2008 confirmed this finding. Hence SCC can be considered as one of the possible (temporal) water structures but not a specific water mass of the ESS. For this reason, we have considered a more common situation when freshened Siberian Shelf waters interplay with transformed Pacific Origin waters.

C2: Moreover, the main results of the manuscript, i.e., the presentation of year to year changes in air-sea CO₂ fluxes of the entire ESS during the two atmospheric regimes appear rather short and more emphasize on how these calculations have been done (spatial averaging) and the related uncertainties should be presented.

A2: Thank you for suggesting. We have expanded the discussion of CO₂ fluxes.

Specific comments:

C3: Page 1228: Line 14: high productive compared to what?

A3: Eastern part of ESS is characterized by higher rates of primary production compared to western part of the ESS (about 7 folds), because it's influenced by water of the Chukchi Sea, which is among the most productive Arctic seas (Walsh et al., 1989; Springer and McRoy, 1993; Vedernikov et al., 2000). Note, that productivity of the eastern part of the ESS ($0.4 \text{ g C m}^{-2} \text{ day}^{-1}$) is closed to productivity of the Chukchi Sea in September ($0.53 \text{ g C m}^{-2} \text{ day}^{-1}$, Springer and McRoy, 1993; Vetrov and Romankevich, 2004) and production rate, measured in the northwest Chukchi Sea during August of 1993 ($0.30 \text{ g C m}^{-2} \text{ day}^{-1}$, Cota et al., 1996).

C4: Page 1228: Line 20: how can that be: a sink of CO₂ and simultaneously a net flux of CO₂ into the atmosphere?

A4: The comment is true, but our intention was to state that the whole ESS acts as a weak source. We have now clarified this in the text.

C5: *Page 1229: Line 6: “marine hydrology”: I would use hydrography for marine waters and hydrology for fresh waters (this should be used consistently throughout the manuscript).*

A5: Thank you. We have followed this recommendation.

C6: *Line 24: there were a bunch of recent publications that should be given (also by the co-authors from this manuscript)*

A6: OK, it has been done.

C7: *Pages 1230-1231: I would suggest having a “study area” paragraph in the M&M section;*

A7: Thank you, we'll consider this suggestion.

C8: *Page 1231: Line 8 and following: the two atmospheric regimes are central in the discussion of the data; I would suggest mentioning already here the position of the Siberian coastal current and introducing the structure of the paper according to the position and extend of the two confronting water masses as a result of the two atmospheric regimes.*

A8: Thank you for suggestion. We have added information concerning with interplay of water masses (see A1) as a result of the two atmospheric regimes. However, we have not changed the structure of the paper as explained under A1.

C9: *Page 1232, second paragraph: please indicate what is new in this paper. Is this purely a review of three surveys or does it add something new; the overall budget I guess??*

A9: This study aims at producing overall budget of CO₂ for the studied area based on multi-year measurements of carbonate system (CS) parameters and highlights the importance of different factors governing the CO₂ fluxes dynamics in the ESS. To date, this is first such study.

C10: *Page 1235: Line 24: what is a significant fraction; you mention at several occasions in the manuscript the effect of sea ice and melting, but you do not constrain this in any way; thus I would take it out.*

A10: We made conclusion about significant fraction of melt water in the ESS freshwater budget based on the literary data. We compared volume of the direct river discharge in the East Siberian Sea (about 150 km³ per year), the Lena River annual discharge (535 km³ per year) (<http://rims.unh.edu>) and long-term summer melt water influx into the ESS – 529 km³ (Dmitrenko et al., 2008). We have transferred this information into the description of study area.

C11: *Page 1237: Line 11 and following: this is the main and new finding in the manuscript and is rather shortly presented. As I understand are these data the basis for the “first direct air-sea CO2 flux multi-year considerations” presented at page 1247. The reader cannot reproduce how the spatial averaging has been done leading to the conclusion presented at page 1247; is this a stable front that we see in 2008 or is this only a snapshot view. How certain are these estimates, how variable is the coastal current also within the discussed years. The reader needs simply more information on this and more room should be provided in this paper (both in the result and discussion section) on this central point of the presented paper.*

A11: Thank you for the important comment. Following this recommendation, we expanded presentation of the results and devoted more space to discussing the central point of the paper.

C12: *Page 1238: General: I would suggest to present the key implications of this paper in the introductory part of the discussions (they are presented far later at page 1247). Later on you can discuss the major processes and uncertainties involved (but only those you have really data for, the others such as melting sea ice or resuspension should be only mentioned shortly); moreover you should discuss the uncertainty in spatial averaging of the two water masses and their related air-sea fluxes*

A12: Thank you for suggestion. We have modified the paper structure and data presentation.

C13: Line 17 and following: a consumption of 1 mol C m⁻² is 12 g m⁻² and if we assume an f-ratio of 0.1 to 0.3 the overall PP can be between 50-100 gC m^{-s} which is not too low. However, the measured consumption of CO₂ is net, i.e, the PP may be much higher since respiration adds continuously CO₂ to the water column

A13: We have clarified which data are new PP and which are total. However, we do not make any conclusions if the EES is a high or low productive sea, only refer to other studies. But we conclude that very few measurements have been performed here making the PP estimates uncertain.

C14: Page 1239: the data presented in this paper do not help to explain coastal erosion patterns; I would therefore suggest to cut down this discussion significantly

A14: Data presented in this paper has not been aimed to explain coastal erosion patterns but rather suggest that CO₂ fluxes should be considered one biogeochemical consequence of coastal erosion. We have cut down significant part of this discussion, but left what we think important to support our point of view.

C15: Page 1240: Line 6 and following; this is redundant information that should be given in the result section

A15: Accepted.

C16: Line 19 and following section in 1241: your data do not shed new light on melting, brine formation and so forth, i.e. cut this discussion down significantly; Alling et al 2010 says that ice does not play a major role!

A16: Thanks, we'll cut down significant part of this discussion.

C17: Page 1242: Line 19: normally the euphotic zone is given as the 1% light level

A17: Following your recommendations, we now present PAR data relative to the 1% light level – it's really more useful for the interpretation.

C18: Page 1243: In what respect does your data set presented help to explain benthic boundary layer phenomena? In case not cut down the discussion on this issue.

A18: The purpose of the discussion is to present possible explanations of the observed data. Thus processes at the benthic boundary layer contribute to high pCO₂ values in the shallow western part of the ESS. Because of its shallowness, it is highly affected by water mixing down to the bottom during storm event, which occur there more often than in deeper parts of the ESS.

C19: Page 1245-1246: Instead of discussing interannual variability as a sub-chapter I would use the two water masses "Siberian Coastal Current" and "surrounding water masses" or "open marine water masses" to present and discuss the major processes governing the CS and air-sea fluxes

A19: Please, see A1.

C20: Page 1247: The main finding presented here need better estimates of uncertainties (spatial averaging) as well as the potential of processes unaccounted for (short discussion!) as the effect of CH₄ oxidation in the water column and a potentially huge degassing event after ice-break up as observed in lakes

A20: We have discussed uncertainties of estimates as well as some unaccounted processes (effects of fall convection, ice-freezing and ice-break up etc.) in the revised version.

C21: Page 1249: Line 14: maybe add here some info on global patterns of coastal seas as a carbon pump

A21: Thank you for suggestion. It has been considered.

Anonymous Referee #2

C1: When I first read this manuscript, I thought it was nice to see a new data set in a marginal sea of high latitude (particularly a multiple years' data set), where becomes more and more important on estimating the global oceanic CO₂ uptake, due to the impact of climate change. Data of September 2003 and 2004, which have been published in Semiletov et al. (2007), are used for this manuscript again. Figures 9 (a) and (b) in this manuscript are almost the same as Figures 11(A) and (B) in Semiletov et al. (2007). Data of 2003 and 2004 in Table 2 of this manuscript are very similar to Table 4 in Semiletov et al. (2007). Most importantly, after carefully went through this manuscript again, I still get an impression on that the authors present the data in a manner as they are all new and have never been published.

A1: We are a bit confused about Referee's statements made in this paragraph. One of the purposes of this paper is to analyze the inter-annual variability of CO₂ fluxes – how could that be done without using multi-year data sets? Would there be a point to present only data of 2008 and discussing its difference from other data sets by referring the reader to the mentioned paper (Semiletov et al., 2007), where 2003 and 2004 were shortly described? We are also confused about Referee's opinion that nothing new is presented in the paper. We believe that we clearly distinguished novelty of this paper by presenting the overall budget of CO₂ over the studied area (based on multi-year direct measurements of CS parameters together with CDOM, PAR and SPM data) and highlighting importance of different factors governing carbonate system and CO₂ fluxes dynamics in the ESS. Thus, we totally disagree with the comment.

Anonymous Referee #3

C1: The manuscript presents hydrographic and carbonate system measurements from three field studies that took place in the East Siberian Sea during August 2003, 2004 and 2008. The primary conclusions are that the carbonate system (magnitude and distribution) is strongly influenced by organic matter degradation (the organic matter primarily originating from river runoff and coastal erosion), river inflow and meteorological forcings. The authors support these conclusions by examining correlations between, for example, pCO₂ and suspended particulate matter and by qualitative descriptive associations. The data are interesting but have apparently already been reported in a previous paper – this needs to be clarified or the paper withdrawn.

A1: We believe that we clearly distinguished novelty of this paper by presenting the overall budget of CO₂ over the studied area (based on multi-year direct measurements of CS parameters together with CDOM, PAR and SPM data) and highlighting importance of different factors governing carbonate system and CO₂ fluxes dynamics in the ESS.

C2: My other primary concern with the paper is that there is no reason to believe the calculated pCO₂, upon which most of the conclusions are based. This arises because the pCO₂ was calculated from pH measured with glass electrodes calibrated using NBS buffers. These buffers are not appropriate for electrode calibrations for seawater measurements (see DOE 1994) and can lead to large systematic errors.

There are no uncertainties (accuracy or precision) quoted for the pH.

The only way the accuracy can be rigorously established is by independent measurement of another carbonate parameter such as dissolved inorganic carbon, by pH measurement of an appropriate seawater buffer (tris) or by direct comparison with spectrophotometric pH measurements. A thorough discussion of the accuracy of the calculated pCO₂ must be provided.

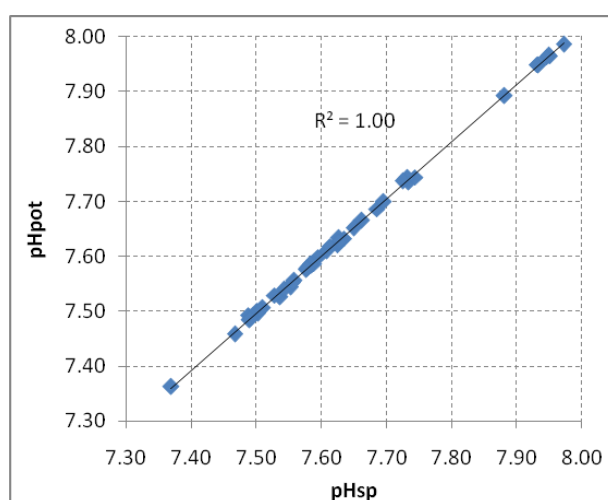
A2: We are grateful to this deep critical analysis of our manuscript. As many other colleagues worldwide, we used tris buffer prepared according to DOE (1994) and Dickson et al. (2007) for

sea water analysis. pH measurements were made in “total” scale in the closed thermostated cell under temperature 20°C. We wanted emphasize that electrodes were calibrated in the NBS scale also for the work at low salinities (for example, near the river mouth). So, we strongly believe that there is no reason not to believe the calculated pCO₂, used in manuscript.

The precision of pH measurements was ±0.004 pH units, as is specified in the manuscript.

Direct comparison between potentiometric (pH_{pot}) and spectrophotometric (pH_{sp}) measurements (both in “total” scale) was carried out in September 2008. Results of this comparison are presented in the Figure below and demonstrate a good coincidence between two methods. Potentiometric analyses was made at temperature 20°C and the spectrophotometric data were recalculated to 20°C. Furthermore in 2008 the CO₂-system was over determined, dissolved inorganic carbon (C_T), total alkalinity (A_T) and pH, and computations using the different constituents showed good pH accuracy as C_T and A_T were calibrated versus CRMs, supplied by Andrew Dickson, Scripps Institution of Oceanography.

Figure. Plot of pH_{pot} vs pH_{sp}



The partial pressure of CO₂ (pCO₂) was computed from pH and total alkalinity using the software CO2SYS (Lewis and Wallace, 1998). The carbonic acid dissociation constants (K₁ and K₂) of Mehrbach et al. (1973) as refit by Dickson and Millero (1987) were used. The uncertainty in computed pCO₂ was about 10 µatm.

Reliability of our results is confirmed by direct measurements of seawater pCO₂ by SAMI-sensor also (Semiletov and Pipko, 2007).

C3: I also agree with the other reviewer that overall the paper is poorly organized and needs to be rethought and corrected for grammatical errors. There are paragraphs that have no logical order and many redundant sentences.

A3: We do not agree that the paper is poorly organized, but have looked over the grammar and taken into account all the constructive suggestions for improving this paper.

Other specific comments are as follows:

C4: P. 1231, paragraph 10: refer to Figure 3 to make this more clear.

A4: We discuss in the Introduction the general atmospheric situation (two predominant large-scale centers of atmospheric circulation over the Arctic Ocean) and guess that reference and discussion of Figure 3 are more relevant in the Result section.

C5: p. 1233, 15-20: Ex=, Em = ? this is wavelength but it is not obvious.

A5: Fluorometer used to assess the in situ CDOM concentration have a single excitation ($E_x=370$ nm)/emission ($E_m=460$ nm) wavelength pair. This has now explicitly been stated in the manuscript.

C6: p. 1234, 5: why is Schmidt number mentioned here?

A6: Thank you for remark; we have added the equation for the gas transfer velocity (k) where Schmidt number was used.

C7: Figure 4: the legend has fine print that is not easy to see. Simplify these figures

A7: Done.

C8: Plot and compare Alkalinity versus salinity curves for the three years

A8: Done.

C9: p. 1238, 15: low light levels are frequently mentioned but PAR data are not shown. Show profiles of PAR from the 3 years, in different areas, to support the arguments

A9: Done.

C10: p.1240 (top): report the actual change in pCO_2 , not the temperature coefficient (also, it's Wallace, not Wallase).

A10: Done and it's really Wallace. Thank you!

C11: p. 1240 (bottom): here Alk-S is mentioned with a non-zero offset on the following page? Show these correlations.

A11: Done.

C12: p. 1241, 20: the riverine DIC and Alk endmembers are less than the seawater levels, but that is not what is said here. Perhaps you mean the salinity-normalized values? However, there is still much less inorganic carbon in the freshwater source.

A12: This sentence has now been rewritten to: "Runoff impacts the seawater composition not only in lowering salinity, but also by increasing the dissolved organic carbon (DOC) as well as the salinity-normalized A_T and DIC, the latter as the runoff is characterized by low pH and high pCO_2 (Figs. 7 and 8)."

C13: Heterotrophy of allocthonous organic matter is cited as the source of the high pCO_2 yet is it possible to have high rates of remineralization in these cold waters? These statements need to be supported by literature measurements of respiration.

A13: The pioneering studies on the activity of the microbiota in the Arctic were performed by famous Russian microbiologist B.L. Isachenko. He published a number of papers concerning bacterial activity in the Arctic (for example, "Investigation of the bacteria in the Arctic Ocean" (1914)). His work is summarized in "Selected Proceeding" (1951). He first established that microbiota in the Arctic seas are very efficient.

Saliot et al. (1996) also pointed to the significant adaptation of arctic bacterial communities to the a priori unfavourable environmental conditions such as temperature.

A comprehensive investigation of carbon and nutrient cycling in Arctic marine sediments was presented in Rysgaard et al. (1998). It showed that benthic mineralization rates in the permanently cold environment are comparable with rates from much warmer locations. It was established that mineralization of organic matter in high-Arctic sediment was highly efficient and regulated by the availability of organic matter and not by temperature.

The fact that the organic matter buried in the permafrost is bio-available has been confirmed earlier (Semiletov, 1999; Guo et al., 2004; van Dongen et al., 2008; Anderson et al., 2009; Vonk et al., 2010; Sánchez-García et al., 2011).

C14: Are there other possible sources of high pCO₂ (e.g. loss of alkalinity relative to DIC, or when you mix the river and seawater endmembers conservatively, can this result in high pCO₂?). Please address these other possibilities.

A14: OK, we have done it.

C15: p. 1246, 25: The atmospheric pressure pattern needs to be more clearly connected to the observed biogeochemical distributions. It leaves a lot up to the reader to make these connections.

A15: We have tried to illustrate these connections more clearly.

C16: In this same paragraph SPM in 2008 is mentioned but no SPM data from 2008 are presented.

A16: This information was kindly provided by O. Dudarev (personal communication). Maximal surface concentration of SPM in the western part of the ESS during September 2008 was about 6.5 mg l⁻¹.

C17: p. 1248 (top): It's helpful if gas fluxes are also reported with units of mol/m²/yr for comparison to annual rates in other areas, even if there is ice cover for most of the year.

A17: We'll try to do it.

C18: There are a number of papers that describe the carbonate system of the Chukchi Sea (Bates et al. papers, etc) that provide important end member information for the eastern ESS. These should be cited and discussed.

A18: Thank you for remark. Of course, we all know the nice works of Nicolas Bates group and some other (Murata and Takizawa, 2003; Kaltin and Anderson, 2005, for example) that describe the carbonate system of the Chukchi Sea. All these papers have analyzed data for the eastern part of the Chukchi Sea, influenced by the low saline Alaskan Coastal Water (Walsh et al., 1989; Springer and McRoy, 1993). On the contrary, western part of the sea is strongly influenced by more saline Anadyr Water. Working in the eastern (Pipko et al., 2002) and western (Pipko et al., 2006) part of the Chukchi Sea, we found that carbonate characteristics of the eastern and western parts are very different and the "eastern" Chukchi Sea data could not be used as C_T or A_T end-member in the eastern ESS.