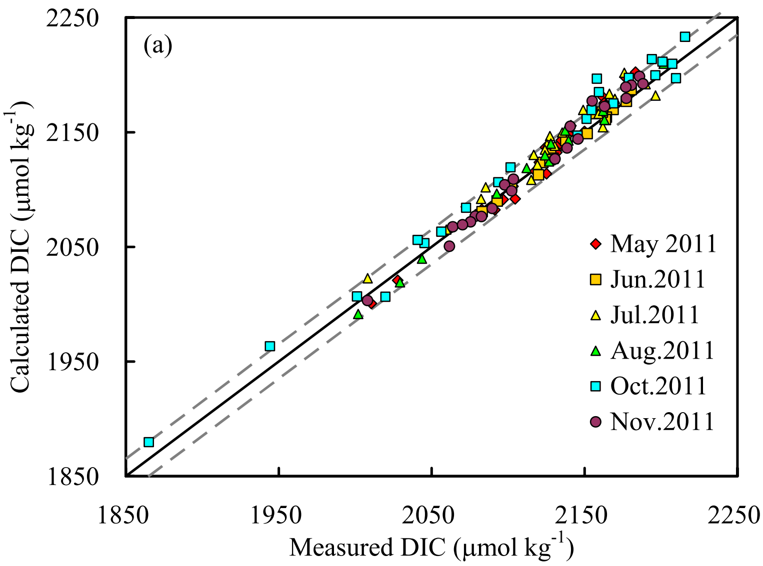
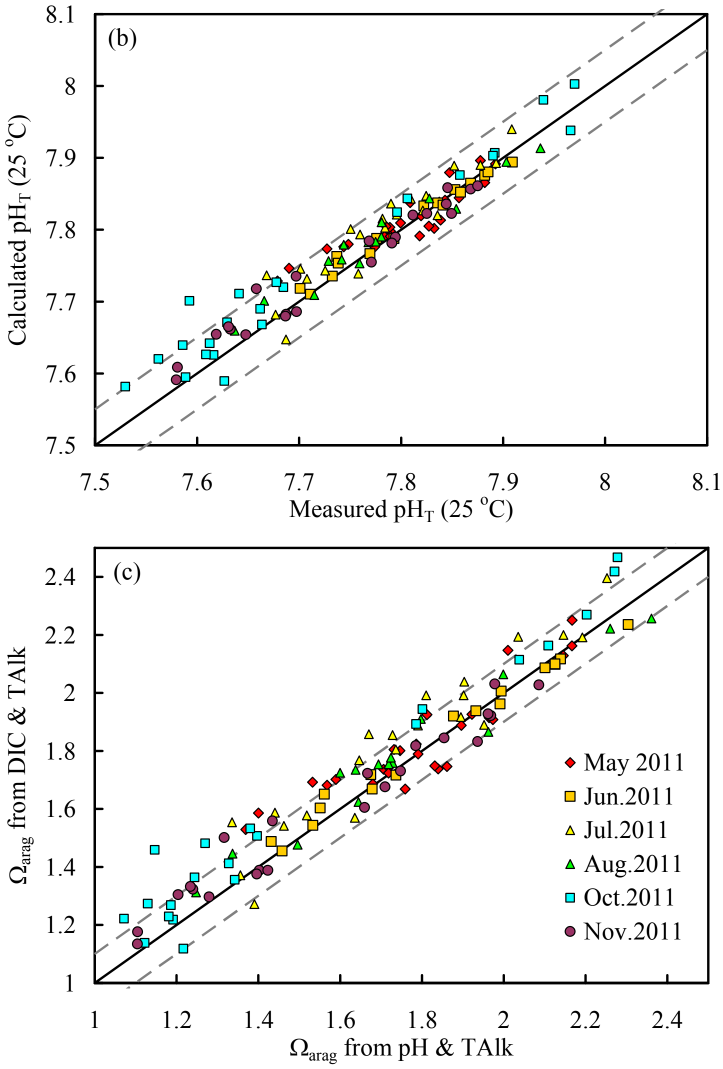
Response to anonymous Review #1 (C890–C894)

We thank the reviewer for his comments and constructive suggestion. Most of them are incorporated in the modified MS. Here all point-by-point responses are provided using blue color and after the mark "Response:", with original comments provided in plain text.

General comments: The manuscript present seven field measurements in TA and pH with related parameters e.g., dissolved oxygen, water temperature and salinity in the North Yellow Sea (NYS) between May 2011 and January 2012. It investigated present status on seasonal variations of pH and Ωarag (carbonate saturation state of aragonite) with related controlling mechanisms in the western and central parts of the NYS and then made future predictions of marine environment changes under the atmospheric CO2 rising scenarios. The author mentioned that the high quality dataset of Ωarag is the first reported for the NYS region. The Ωarag data with pCO2 levels in the NYS were, however, calculated from the pH and TAlk and might have large uncertainties or errors involved. For instance, the pHT (25oC) values in October 2011 listed in Table 1 may be problematic. The authors shall have double checks.

Response: In the modified MS, we re-calculate bottom water pH and Ω_{arag} using DIC and TAlk. To further assess the quality of this dataset, we compare the calculated DIC (from measured pH at 25 °C and TAlk) versus measured DIC, and the calculated pH (from DIC and TAlk) versus field-measured pH, and Ω_{arag} values from DIC and TAlk versus those from field-measured pH and TAlk. Most of them are consistent with each other at deviation levels of ± 15 μmol kg^{-1} (DIC), ± 0.05 (pH_T at $25^{o}C$) and ± 0.1 (Ω_{arag}). New Ω_{arag} data give similar patterns to the original MS, although some Ω_{arag} values from DIC and TAlk are slightly higher than those from field-measured pH and TAlk by 0.1 to 0.2. See below figures, which have also been presented in the modified MS as supporting materials.





The data are overall not typical and lack of new knowledge of the community since similar papers previously addressed the things what the authors proposed. The combined effects of global atmospheric CO2 increase, regional environmental changes and local biogeochemical processes were proposed but not de-convoluted i.e., the contributions from each effect on low pH and Ωarag. That caused the authors overestimated only based on one-year seasonal survey instead of decadal time-series and lost merits of the paper which data are.

Response: We have changed our title to "Seasonal variations of subsurface pH and carbonate saturation state of aragonite on China side of the North Yellow Sea". We agree that most of the controlling processes have been documented in literatures. However, in order to better understand their regional effects, more researches are still needed, especially for those coastal zones of economic importance. In the NYS, the similar spatial distribution pattern and low values of Ω_{arag} have been observed again in the 2012 autumn (See "Bulletin of Marine Environmental Status of China: 2012", http://www.soa.gov.cn/zwgk/hygb/zghyhjzlgb/hyhjzlgbml/2012nzghyhjzkgb/201303/t20130329_24712.html). That is, this phenomenon may occur every year in the NYS. Our MS is the first try to report this significant phenomenon in details. On the other hand, so far most researchers resolve seasonal variations of pH and Ω_{arag} only based on several snap-shot like surveys. We contend that this is insufficient since carbonate system in any marginal sea shall be subject to tremendous heterogeneities in both space and time. In this study, we try to resolve the seasonal variations and spatial distributions of subsurface pH and Ω_{arag} in the NYS, based on nearly monthly surveys conducted from May 2011 to January 2012 (plus another May 2012 dataset in the modified MS). This detailed baseline dataset shall benefit the scientific community for long.

The authors tried to do future predictions under the same season conditions in relation to the effects of the global and regional changes to address the changing of pCO2, pH and Ωarag status in the NYS. However, the data presented here may be only reflected to the one-year seasonal variation instead of the effects as mentioned above although the authors wanted to build up the case. Overall speaking, the manuscript needs to have a major revision if the paper could be accepted. The paper is also hard to read and to understand since the current presentation sometimes at 25 oC or sometimes in-situ is quite confusing and the data could be checked again. Finally, could it be possible to put all the hydrographic and carbonate data in the supplementary materials?

Response: According to the reviewer, the focus has been changed into seasonal variations of subsurface pH and carbonate saturation state of aragonite on China side of the North Yellow Sea. The impacts of mixing process are substantially discussed. And the future prediction is touched under a scenario of global atmospheric CO_2 increase. So, we have tried to appropriately utilize the high quality dataset and to adequately reveal its merits. We also change most pH_T (25 ^oC) into in situ. Finally, we put all the bottom water data of temperature, salinity, TAlk, DIC, DO, field-measured pH (25 ^oC), and calculated pH (in situ) and Ω_{arag} in the new Table 1.

Specific comments: P3080: Could the title: "Subsurface low pH and carbonate…: combined effects of global atmospheric CO2 increase, regional environmental changes, and local biogeochemical Processes" be changed to "Subsurface low pH and carbonate…: present status and future predictions"?

Response: The title has been changed into "Seasonal variations of subsurface pH and carbonate saturation state of aragonite on China side of the North Yellow Sea". Thank the reviewer for his constructive suggestion.

P3081: In abstract, please take a good care of significant digits such as Ωarag in one significant digit or two; pH, salinity, DO etc. Salinity is unitless without psu for the whole text.

Response: In the modified MS, we make significant digits consistent and delete "psu" from salinity values. Thank the reviewer for reminding us.

P3081, L5-6: Subsurface waters were nearly in equilibrium with air in May and June. Could it be added fCO2 and pH values in May behind and June, respectively? fCO2: 371±42 μatm, 392±35 μatm; pH (25 oC): 7.79±0.05, 7.79±0.07. pH shall be mentioned either at 25 oC or in-situ. Will pH presented in-situ be better than ones at 25 oC?

Response: All those unclear values have been identified. And most pH values have been changed into in situ.

P3081, L9-10: It is the same for adding the fCO2 and pH values in November behind and January, respectively

Response: Done.

P3081, L19-20: Please clearly define typical North Yellow Sea bottom water mass

Response: Done.

P3083, L166: " ..with an area of 71 300 km2 and a mean depth of 38 m". Please add a reference or define the range in Fig. 1

Response: We refer to Xue et al. (2012). And also, we clarify the difference between the NYS and our study area in the modified MS.

Refer to: Xue, L., Xue, M., Zhang, L.-J., Sun, T.-M.,Guo, Z., and Wang, J.-J.: Surface partial pressure of CO_2 and air–sea exchange in the northern Yellow Sea, J. Mar. Syst., 105, 194–206, 2012.

P3087, L11: Significant digits of temperature shall be consistent in the following text and table.

Response: Modified. Thank the reviewer for reminding us.

P3087, L11: Please make clearly the two northern stations in Fig.1.

Response: Marked by a purple-line ellipse in modified Fig. 1.

P3088, L02: The definitions of water masses could be moved to the section of the method.

Response: Modified accordingly.

P3088, L8-10: The comparison of chl-a in NYS to ones in the ECS will be good instead of the SCS. Overall, the data were so high, particularly in January. Please check!

Response: Several remote sensing researches have demonstrated the high chl-a / primary production levels in the NYS (Tan et al., 2011; He et al., 2013). We also dig out a set of field-measured data of chl-a in the NYS (Gao and Li, 2009), which was collected during 20 July - 07 August, 2006 (summer) and 31 December, 2006 - 16 January, 2007 (winter). See the below figure. During their winter cruise, both the spatial distribution pattern and magnitude of surface chl-a are similar to our results in January 2012. In their summer cruise, however, the sea surface chl-a are reported as 0.14-2.26 μg/L (Gao and Li, 2009; Yang et al., 2009), which is lower than our sea surface chl-a values of 0.75-8.17 μg/L in May 2011, 0.44-4.66 μg/L in July 2011, and 0.81-10.97 μg/L in October 2011. In 2011, 7 cases of red tide were recorded in the study area (http://www.soa.gov.cn/zwgk/hygb/zghyhjzlgb/hyhjzlgbml/2011ml/201212/t20121206_21275.html). We also saw broad algae blooms of *Noctiluca Scintillans* on the spot by eyes. Therefore, these high values of surface chl-a are reasonable.

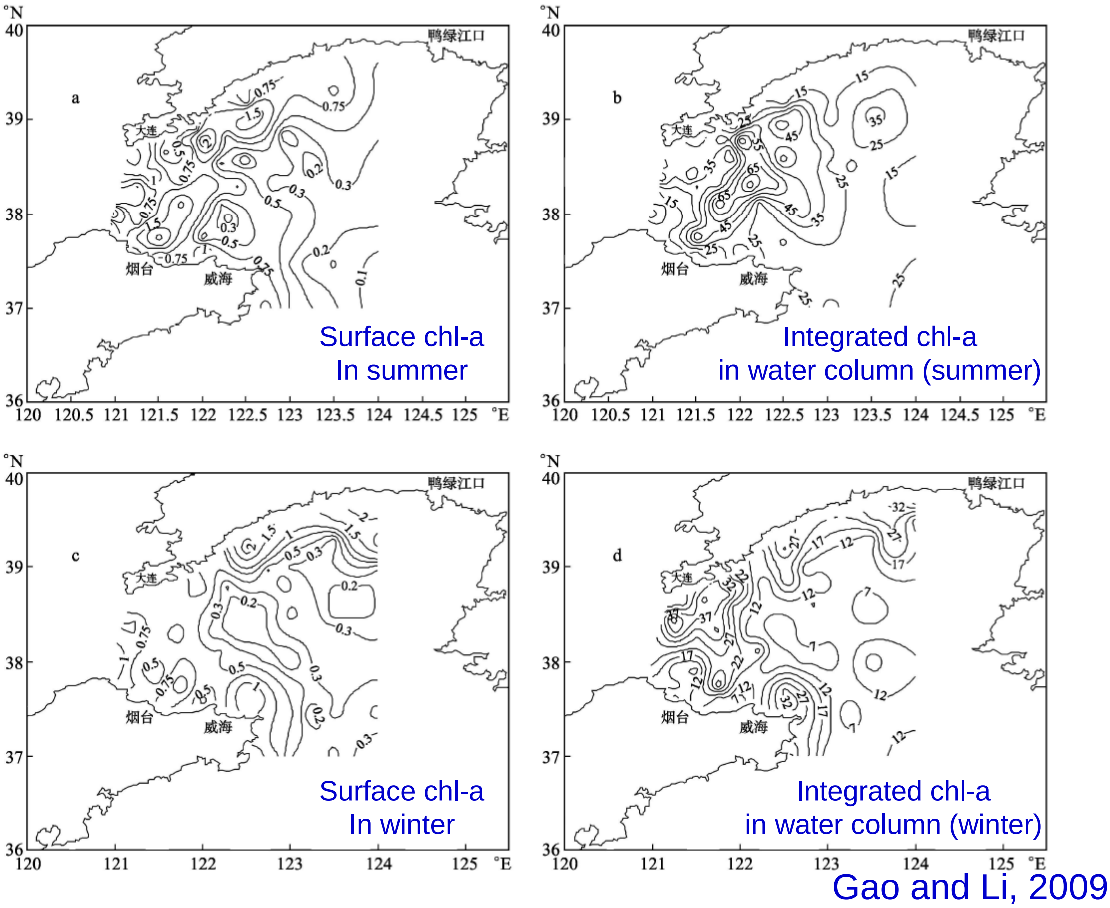


图 2 北黄海夏、冬季表层 Chl *a* 浓度/mg• m^{-3}和水柱 Chl *a* 含量/mg• m^{-2}分布图

Fig. 2 The spatial distribution of Chl *a* in summer and winter in surface water(mg• m^{-3}) and water column (mg• m^{-2}) in the Northern Yellow Sea

(a. 夏季表层 Chl *a* 浓度; b. 夏季水柱 Chl *a* 含量; c. 冬季表层 Chl *a* 浓度; d. 冬季水柱 Chl *a* 含量 a. Chl *a* in summer in surface water; b. Chl *a* in summer in water column; c. Chl *a* in winter in surface water; d. Chl *a* in winter in water column)

In the modified MS, this issue has been clarified. We also delete the comparison of chl-a between our study area and the SCS.

References:

Gao, Y. and Li, Z.-Y.: Spatial and seasonal variation of chlorophyll and primary productivity in summer and winter in the Northern Yellow Sea, P. Ocean Univ. China, 39, 604-610, 2009 (in Chinese).

He, X.-Q., Bai, Y., Pan, D.-L., Chen, C.-T. A., Chen, Q., Wang, D.-F., and Gong, F.: Satellite views of seasonal and inter-annual variability of phytoplankton blooms in the eastern China seas over the past 14 yr (1998-2011), Biogeosciences Discuss., 10, 111-155, 2013.

Tan, S.-C., Shi, G.-Y., Shi, J.-H., Gao, H.-W., and Yao, X.: Correlation of Asian dust with chlorophyll and primary productivity in the coastal seas of China during the period from 1998 to 2008, J. Geophys. Res., 116, G02029, doi:10.1029/2010JG001456, 2011.

Yang, G.-P., Zhang, H.-H., Su, L.-P., and Zhou, L.-M.: Biogenic emission of dimethylsulfide (DMS) from the North Yellow Sea, China and its contribution to sulfate in aerosol during summer, Atmospheric Environment, 43, 2196-2203, 2009.

P3088, L26-28: please add DO saturation numbers in after DO concentration.

Response: Added.

P3089, L13: Fig.8 shows the bottom water pHT (25 oC)… Could the plots be made from pHT (25 oC) to pHT (in-situ), please? Explain why the lowest pH<7.7 occurred? or data quality?

Response: In the modified MS, most pH data have been reported at pH_T (in situ), other than the plots of pH_T against DO. The lowest pH_T (in situ) is calculated at 7.79. If we measure pH at 25 ^oC, however, the lowest pH_T of <7.7 is really obtained. According to Gieskes (1969), a temperature increase of 10-20 ^oC (from bottom water temperature of 5-15 ^oC to 25 ^oC) shall lead to the pH drop of 0.11-0.23. Therefore, the lowest pH_T (25 ^oC) value of ~7.60 is correct.

Refer to: Gieskes, J. M.: Effect of temperature on the pH of seawater, Limnol. Oceanogr., 14, 679-685, 1969.

P3090, L4-26: Please check all the Ωarag data in bottom waters again at 25 oC or in-situ. Could all the data be uniformed as in-situ?

Response: All the Ω_{arag} data have been reported at in situ. The Ω_{arag} drop between August and October is still there.

P3091, L17: In the NYS, the apparent temperature coefficient of bottom water pHT in 2011 was estimated at -0.0144 pH units oC-1 (n=7, r =0.996), based on the survey averaged dataset summarized in Table 1. How do you get the slope: -0.0144 pH units oC-1 (n=7, r =0.996)? Could you plot the pH vs Temp ?

Response: Since Gieskes (1969) has reported a similar temperature coefficient of pH, we deleted this confusing statement. Thank the reviewer for reminding us.

P3091, L24: ..overall seasonal variation of 0.25 units in 2011.. Is 0.25 right?

Response: Based on calculated pH_T data from DIC and TAlk, the overall seasonal variation of pH_T (in situ) should be 0.22 units (8.09±0.03 in May minus 7.87±0.06 in October). The modified MS has been updated.

P3092, L23-25: You could make a plot shown in the supplementary material.

Response: We deleted this discussion in the modified MS.

P3092, L28: ..temperature normalized fCO2 (at 25 oC)? How to calculate? Please add a reference.

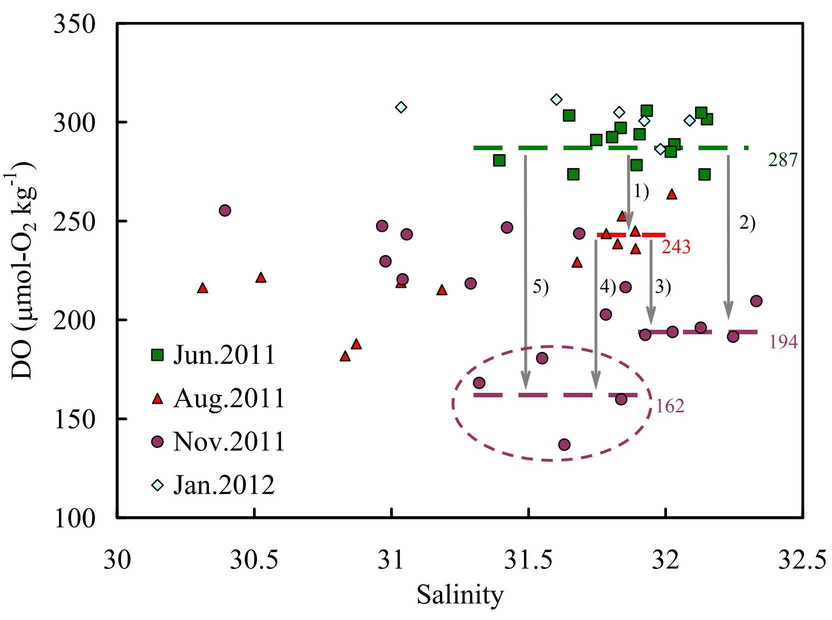
Response: We calculated all those carbonate system parameters including fCO_2 (at 25 ^{o}C), based on DIC, TAlk, seawater temperature, and salinity, using the calculation program CO2SYS.xls. In the modified MS, this issue has been clarified.

P3093, L22: pH-DO correlation could be reported in the supplementary material.

Response: We discussed the plot of pH against DO in the modified MS.

P3094, L15: 0.55–1.0 μmolO2 kg-1 d-1? How to get it? Just got 0.2 from Table1 between August and November.

Response: We are sorry for the unclear expression. Salinity distributions of DO are plotted blow. Our November 2011 was subject to cooling induced vertical mixing. Therefore the bottom water DO values were not uniform. The relatively high values indicated vertical mixing induced oxygen recovery. However, even under this bad condition, we still obtained two relatively uniform DO levels for the November 2011 cruise. One is 194±2 μmol kg^{-1} (n = 4, with the salinity range of 31.3 to 31.9), and another is 162±19 μmol kg^{-1} (n = 4, with the salinity range of 31.9 to 32.4).



From June to August, a DO depletion rate of (287-243)/60=0.73 μmol kg^{-1} d^{-1} (1) is obtained, while the DO depletion rate values in different salinity ranges are (287-194)/150=0.62 μmol kg^{-1} d^{-1} (2) from June to November, (243-194)/150=0.55 μmol kg^{-1} d^{-1} (3) from August to November, (243-162)/90=0.90 μmol kg^{-1} d^{-1} (4) from August to November, (287-162)/150=0.83 μmol kg^{-1} d^{-1} (5) from June to November. All of them are consistent in a range of 0.55-0.90 μmol kg^{-1} d^{-1}. The modified MS has been clarified.

P3095: In the 4.4 section ("Effects of Bohai Sea Water mass outflow") be suggested to add the theoretical mixing line of two end-members into the graphs/section to identify effect of the two distinctive sources by water masses. The same question in mixed uses of the pHT (25oC) and pHT (in-situ) occurred. Could you make all pH at in-situ, instead of at 25 oC since Ωarag data were derived from the pH (in-situ) and for comparison consistency, please?

Response: The theoretical mixing lines have been added. Thank the reviewer for his suggestion. Also we have changed most pH (25°C) to pH (in situ).

P3097, L7-12: The statements related to low Ωarag status, not only influenced by global atmospheric CO2 increase and local respiration/remineralization, but also by major environmental changes may be vaguely descriptive. However, do you have any data or direct evidence to prove especially for the issue of environmental changes?

Response: The discussion has been deleted, since the modified MS has been focused on seasonal variation and spatial distributions.

Could the author quantify the contributions on each effect on the low Ωarag status change? or make a preliminary first-order budget ?

Response: We thank the reviewer for his constructive suggestion. We have made a first-order budget in a table in the modified MS.

P3103: in Table 1, could the authors add more information such as pHT(in-situ), DO (%saturation), DIC, calculated fCO2(in-situ)? data range after average in each variable? Salinity is unitless. Please indicate the calculated Ωarag data derived by pHT (in-situ)

Response: In the modified MS, we mostly calculate Ω_{arag} and pH_T (in-situ) data from DIC and TAlk, other than the January 2012 cruise. In the January 2012 cruise, however, DIC data were not available. Therefore the fCO_2, pH_T (in situ) and Ω_{arag} values in this specific cruise were calculated based on pH_T (25°C) and TAlk. In the modified MS, the changes have been illuminated.

In the modified MS, we put all those primary data of bottom waters in the study area into the new Table 1, including T, S, water depth, pH_T (25°C), DIC, TAlk, DO, and calculated pH_T (in situ) and Ω_{arag} values. To avoid too large a table, we do not list other calculated parameters. However, we summarize them in the new Fig. 4.

And also we delete “psu” from salinity values.

P3106: suggest to combine Figures 3, 6, 7, and11 into one new figure 3? Horizontal arrangement is each variable; Vertical column is in the order of month from January to November. or vice versa. The variables could be Temp., Chl-a, %DO, pHT (in-situ), Ωarag etc.

Response: According to the review’s suggestion, we combined vertical profiles of Temperature, DO%, pH_T (in situ), and Ω_{arag} into one figure. Chl-a has no vertical profile data.

P3106: suggest to combine Figures 4, 8, 9, and10 into one new figure 4? same as described above.

Response: According to the review’s suggestion, we combined bottom water temperature/salinity, NTAlk, pH_T (in situ), and Ω_{arag} into one figure.

P3117-3118: suggest to combine Figures 14, and15 into one new figure? same as described above. Please add one more variable i.e., TA in.

Response: According to the review’s suggestion, we combined salinity distribution of TAlk, bottom water DO, bottom water pH_T (in situ), and bottom water Ω_{arag} into one figure.

P3119: Add 1:1 reference line into the graph and make both x and y scales the same.

Response: Modified accordingly.

P3120: why only compare three cruise data? Please add the DIC data and discuss it in the manuscript especially for the issue of the seasonal drops in subsurface pH and Ωarag.

Response: In the modified MS, we compare six cruise data in 2011. We also present the DIC data of the six cruises and fully discussed them. See our response to the reviewer’s general comments. During our January 2012 cruise, the DIC data are unavailable. During the supplementary May 2012 cruise in the modified MS, the field-measured pH data are unavailable.

Response to anonymous Review #2 (C995–C999)

We thank the reviewer for his comments and constructive suggestion. Most of them are incorporated in the modified MS. Here all point-by-point responses are provided using blue color and after the mark "Response:", with original comments provided in plain text.

This manuscript "Subsurface low pH and carbonate saturation state of aragonite on China side of the North Yellow Sea: combined effects of global atmospheric CO2 increase, regional environmental changes, and local biogeochemical processes" contains good dataset for an interesting research area in one of the important coast rejoin in the Eastern Asia. This area, North Yellow Sea, have experienced mixing processes and regional environmental change. The carbon dynamics also showed seasonal variation in the bottom water in this area. However, while the author emphasized the combination effects as listed in the title; the authors discuss each effect independently and make this manuscript hard to follow. There are also gaps between the dataset and the conclusion for most discussion sections. This study needs critical evidence to reach these main ideas. Finally, this nine-month data is more suitable to explain spatial variation and likely seasonal variation, and is less likely to explain long-term variations. Thus, the manuscript could be benefited by focusing on the seasonal variation and mixing process; and might be some discussion on those long-term variations. This manuscript needs a major revision before it can be considered for acceptance.

Response: We have substantially reorganized the MS. According to the reviewer, the focus has been changed into seasonal variations of subsurface pH and carbonate saturation state of aragonite on China side of the North Yellow Sea. The impacts of mixing process are substantially discussed. And the future prediction is also touched under a scenario of global atmospheric CO_2 increase. So, we have tried to appropriately utilize the high quality dataset and to adequately reveal its merits.

The major and minor comments are as follow:

Major comments:

1. The mixing process needs to be further quantified in this study. Please provide the method of the normalization of alkalinity at P3089 L21. Please clarify the assumption behind this calculation. If the mixing process between river and seawater were considered, this should be a two end-member mixing. Please also provide the TAlk value in the river end-member in this section to support this. In addition, please clarify the "long-term" mixing at L25-26 and how the evaporation and precipitation affect the TAlk values in Bohai Sea under such "long-term" mixing.

Response: In the modified MS, we have heavily strengthened the demonstration on the mixing process. Basically, we plot TAlk against salinity, including those TAlk data obtained in the Bohai Sea. We find that the relatively low-alkalinity Yalujiang River has effects on the NYS carbonate system, probably due to the winter cooling induced freshwater subduction and mixing with salty waters. Therefore, we obtain a three end-member mixing pattern, including a NYS water mass (salinity ~32), and a Bohai Sea water mass (at salinity ~31), and the low-alkalinity river plume end-member. However, we find that the summer-time Bohai Sea outflow current was mainly localized in the upper 25-m layer in warm/flooding seasons. On those deep stations (water depth > 25 m), the effects of summer-time Bohai Sea water mass outflow are limited, due to the strong stratification in summer in the study area. In the modified MS, we have changed the relevant statements and discussion.

As for the TAlk value in the Yalujiang River end-member, we have dug out it (~ 740 μmol kg^{-1}) from a book chapter (Zhang et al., 2007). This value is consistent with that the extrapolation value of our winter dataset. In the modified MS, we add the relevant discussion.

In the modified MS, we also clarified the normalization of alkalinity in the method section. "To eliminate dilution/concentration effects of precipitation and/or evaporation on the seawater carbonate system, we also calculate out salinity-normalized values of TAlk and DIC, via NTAlk = TAlk×35/Salinity and NDIC = DIC×35/Salinity respectively."

During the water mixing process, evaporation and precipitation not only affect the TAlk values, but also the salinity, while the NTAlk should be a constant. However, water discharge from the high-alkalinity river will increase both TAlk and NTAlk in sea waters. The Bohai Sea is just the case.

Finally, we add several references in the study area part, so as to clarify "long-term" water mixing in the Bohai Sea. "The semi-enclosed Bohai Sea has a very long half-life time of water exchange of 17 to 21 months (Wei et al., 2002). It is fed by more than a dozen rivers of moderate to very high alkalinity (1470 to 6300 μmol kg^{-1}) (Xia and Zhang, 2011), including major runoff contributions from the Yellow River." The water discharge and alkalinity fluxes circulate in the Bohai Sea for several years (Wei et al., 2002; Mao et al., 2008), supporting relatively homogeneous and low salinity and relatively high alkalinity in the Bohai Sea.

References:

Mao, X.-Y., Jiang, W.-S., Zhao, P., and Gao, H.-W.: A 3-D numerical study of salinity variations in the Bohai Sea during the recent years, Cont. Shelf Res., 28, 2689-2699, 2008.

Xia, B. and Zhang, L.-J.: Carbon distribution and fluxes of 16 rivers discharging into the Bohai Sea in summer, Acta Oceanol. Sin., 30(3), 43-54, doi:10.1007/s13131-011-0118-3, 2011.

Wei, H., Tian, T., Zhou, F., and Zhao, L.: Numerical studies on the water exchange of the Bohai Sea: simulation of the half-life time by dispersion model, J. Ocean Uni. Qingdao, 32, 519-525, 2002 (in Chinese).

Zhang, J.: Impact of drainage basin weathering upon riverine chemistry, in: Biogeochemical Studies of Major Chinese Estuaries - Element Transfer and Environment, edited by: Zhang, J., China Ocean Press, Beijing, 1-15, 1997 (in Chinese).

2. As comment #1, please clarify this statement in the last paragraph of Section 4.4, "The Bohai Sea water mass outflow had important impacts on the dynamics of subsurface pH (25C) and Ωarag in the NYS. … Bohai Sea water mass out flow effectively increased the NYS mean bottom water Ωarag". Please clearly point out how much proportion that conservative mixing affects these pH and Ωarag variations. Please also clarify the large variation of Ωarag among salinity 31.5 to 32.5. Furthermore, please also clarify the sentence in the abstract P3081, L21-23 "Out flow of the Bohai Sea water mass counteracted the subsurface Ωarag reduction in the North Yellow Sea." In addition to salinity, please also provide other signals, such as T-S diagram, to elucidate that the water was from Bohai Sea.

Response: As our response to the reviewer#2's comment #1, we have made several corrections on the effects of summer-time Bohai Sea water mass outflow in the study area. We also delete this misleading sentence from the abstract.

We add T-S diagrams to help analyses of water mass. Since temperature is not a conservative parameter in shallow waters, in this study we do the water mixing analysis mainly based on TAlk-salinity diagrams. We also add conservative mixing lines for most parameters against salinity. Based on the below figure of Ω_{arag}-salinity, the bottom water among salinity 31.5 to 32.5 accumulated the low Ω_{arag} signal from June to October-November in 2011. And the low Ω_{arag} begins to recover during November 2011 to January 2012. Note that we add another DIC/TAlk dataset obtained in May 2012, so that we can show the overall seasonal variations of subsurface pH and Ω_{arag} in the study area.

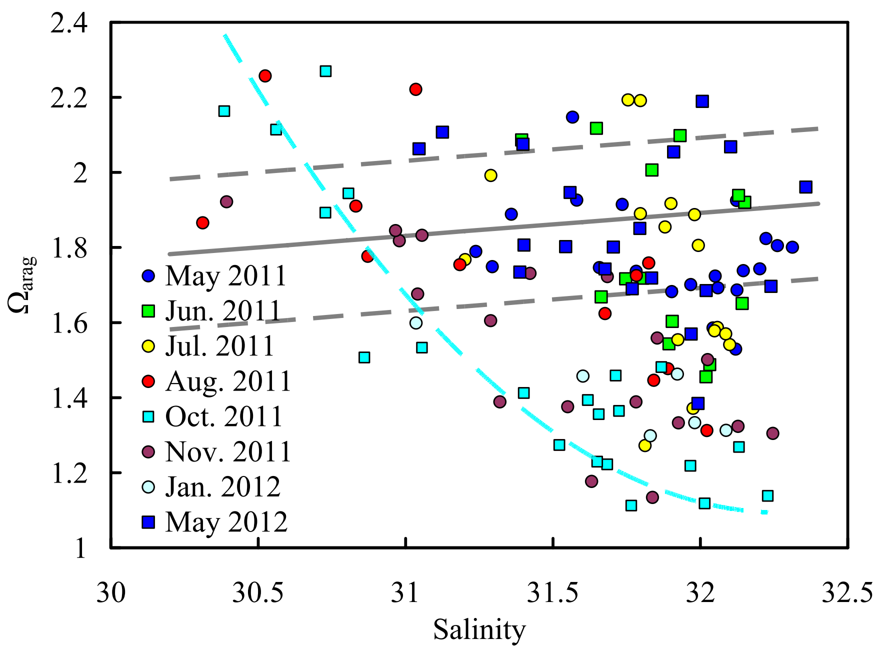


Figure Caption: Bottom water carbonate saturation state of aragonite (Ω_{arag}) against salinity at deep stations (water depth > 25 m). Grey lines show the conservative mixing line (the grey real line) between rain water and the NYS water mass (salinity ~32 and NTAlk ~2500 μmol kg^{-1}) and a variation range of ±0.2 (grey dashed lines). The sky-blue dashed line is the regression line fitted to our Oct. 2011 dataset.

3. Section 4.3, P3094, L2 to 4. Please also explain the effect of lateral transport as you also emphasize the outflow from Bohai Sea as mentioned in comment #1 and 2.

Response: In our deep stations, the effects of lateral transport are negligible. This is consistent with the fact that a typical bottom cold water mass of 5 to 11°C appears in spring and summer in the NYS (Miao et al., 1991; Chen, 2009), which is regarded as the remains of winter cooling and documented as the NYS Cold Water Mass (e.g. Miao et al., 1991). In the modified MS, this issue has been clarified.

References:

Chen, C.-T. A.: Chemical and physical fronts in the Bohai, Yellow and East China seas, J. Marine Systems, 78, 394–410, 2009.

Miao, J.-B., Liu, X.-Q., and Hsueh, Y.: Study of the formational mechanism of the Northern Yellow (Huanghai) Sea Cold Water Mass (I)—Solution of the model, Science in China (Ser. B), 34, 963-976, 1991.

P3094, L18-19, please clearly indicate what is included in “all regional/local processes”.

Response: The so-called “all regional/local processes” are water mixing and respiration / remineralization. In the modified MS, this issue has been clarified.

4. It is author’s choice to discuss the effect of varied atmospheric CO2 value on carbonate system as in Section 4.2. The authors said “Therefore, air-equilibrated pH levels in the NYS declined by 0.13 pH units since the pre-industrial period” in this section. However, as the authors only have nine-month data and an over-simplified model in this discussion, there is still a gap to reach the conclusion: “This study showed that the carbonate system in the North Yellow Sea was substantially influenced by global atmospheric CO2 increase” in the abstract. Such simplified model also conflicted with section 4.5 “Recent regional environmental changes”. Please consider such conclusion again.

Response: We have substantially reorganized the MS. According to the reviewer, the focus has been changed into seasonal variations of subsurface pH and carbonate saturation state of aragonite on China side of the North Yellow Sea. Also the impacts of mixing process are substantially discussed. And the future prediction has been made under a scenario of global atmospheric CO_2 increase.

Furthermore, the atmospheric CO2 in such coastal area might be affected by terrestrial sources and doesn’t fully match the global average. If possible, please provide local atmospheric CO2 value. Please also provide the reference for those preindustrial, present, and future air CO2 values in this study.

Response: Yes, we have local atmospheric CO_2 data. We have provided them in the modified MS. The future air CO_2 concentration refers to IPCC IS92a.

5. In Section 4.5, please provide the reference or measured data for P3097 L5-7 “In the hydrological settings during the 1950s and 1960s, more Bohai Sea water and TAlk were introduced into the NYS, and the above-mentioned seasonal acidification processes were mitigated much more than present.” Furthermore, please provide a better linkage between your data and the first paragraph in Section 4.5, P3096 L9-20.

Response: In the modified MS, we deleted this speculation.

6. The word “typical” in this study should be re-considered as this study only has nine month data, and another paper in Bohai (Zhai at. al., 2012, Chinese Sci. Bull.) also only has short-term data.

Response: In the modified MS, we changed this word. We also add a reference, i.e. Chen (2009). In this large-scale research based on climatological data, roughly salinity ranges of 30.5-31.5 in the Bohai Sea and 31.5-32.5 in the NYS were presented in his Fig. 2b and Fig. 5b.

Reference:

Chen, C.-T. A.: Chemical and physical fronts in the Bohai, Yellow and East China seas, J. Marine Systems, 78, 394–410, 2009.

7. Please improve the introduction. The title mentioned increasing air CO_2, regional environmental change, and local biogeochemical processes; but only increasing air CO_2 is emphasized in the abstract. Please balance these main ideas in the introduction.

Response: In the modified MS, we have substantially reorganized the introduction.

8. Please edit these two sentences in P3082, L24 to 27. They were very similar to the third paragraph in Page 1063 at Zhai et al., 2012, Chinese Sci. Bull.

Response: In the modified MS, we have substantially reorganized the introduction.

9. Please keep the writing consistent over this study. For example, the importance of increasing nutrient discharge and reduction of terrestrial alkalinity inputs were emphasized in the last sentence of the conclusion (P3098, Line 1↑ 3), but this statement was overlooked in the abstract. Major comment #2 also showed unclear/inconsistent between in the Section 4.4 and in the abstract. Another example is as minor comment #3.

Response: Done. We thank the reviewer for reminding us.

Minor comments:

1. The author mentioned “several stations” in Fig. 6c. Please clearly point out these stations on the profiles.

Response: Marked.

2. Please also point out where "those northern stations" in P3087, L19 are on Fig 3f.

Response: In the modified MS, we plot contours of bottom water temperature in the salinity distribution maps. We have referred those northern well-mixed stations to the relatively high-temperature (> 10 ^{o}C) bottom waters in the new contour maps.

3. "China side of the North Yellow Sea" was only mentioned in the title. It was changed to "Chinese side of the North Yellow Sea" in the abstract and conclusion. However, the authors only used "NYS" in the other places. In addition, previous studies (such as Xue et. al., 2012) used "northern Yellow Sea" for this study area. Please check if "North Yellow Sea" is an official name. Finally, please modify the words in P3097, L15-16, western North Pacific continental margin to NYS as the data were only in NYS.

Response: In the modified MS, we change "Chinese side of the North Yellow Sea" into "China side of the North Yellow Sea". We also clarify that our study area is located on China side of the NYS. And then we use "the study area" in any confusing occasion.

As for "northern Yellow Sea" and "North Yellow Sea", both of them are used in literatures. The later usage refers to Tan et al. (2011) and Yang et al. (2009). The similar usage of "South Yellow Sea" is also seen in literatures (e.g. Zhang et al., 2012). We would like to use the "North Yellow Sea" as it is semi-closed and its hydrography is relatively isolated from the South Yellow Sea, just like "North Pacific" versus "South Pacific", and "North Atlantic" versus "South Atlantic". It is also different from the case of "northern South China Sea".

In the summary part of the modified MS, we change "this western North Pacific continental margin" into "the NYS", so as to avoid the possibly misleading statement.

References:

Tan, S.-C., Shi, G.-Y., Shi, J.-H., Gao, H.-W., and Yao, X.: Correlation of Asian dust with chlorophyll and primary productivity in the coastal seas of China during the period from 1998 to 2008, J. Geophys. Res., 116, G02029, doi:10.1029/2010JG001456, 2011.

Yang, G.-P., Zhang, H.-H., Su, L.-P., and Zhou, L.-M.: Biogenic emission of dimethylsulfide (DMS) from the North Yellow Sea, China and its contribution to sulfate in aerosol during summer, Atmospheric Environment, 43, 2196-2203, 2009.

Zhang, J.-L., Xu, F.-S., Liu, R.-Y.: Community structure changes of macrobenthos in the South Yellow Sea, Chinese Journal of Oceanology and Limnology, 30, 248-255, 2012.

4. The author used several words to describe pH and , such as “very low”, “critically low”, and “extremely low”. Please point out biogenic meaning behind them; otherwise these words should be deleted or merely used.

Response: We change these words into quantificational numbers.

5. P3082, L27, please add a reference for the Redfield equation and delete “traditional”.

Response: Modified accordingly.

6. P3083, L21-22, please add reference to support this.

Response: In the modified MS, this sentence has been deleted. According to Fig. 1 of Chen (2009), the Bohai Sea water mass outflow along the southern shoreline is an ever-present phenomenon in a year.

Refer to: Chen, C.-T. A.: Chemical and physical fronts in the Bohai, Yellow and East China seas, J. Marine Systems, 78, 394–410, 2009.

7. P3085, L19, please check if this is a “pH analyzer” or a “pH meter”. Also in P3086 L11.

Response: Thank the reviewer for reminding us. This is a “pH meter”.

8. P3085, L18, please provide the volume of the HgCl2 that was used for each sample, and also provide the volume of the sampling bottles. Also in P3086, L9.

Response: Water samples for pH, DIC and TAlk analyses were stored in 140 mL brown borosilicate glass bottles (for pH), 60 mL borosilicate glass bottles (for DIC), and 140 mL high-density polyethylene bottles (for TAlk). They were all poisoned with 50 μL saturated $HgCl_2$ and sealed with screw caps and preserved at room temperature until determination. This issue has been clarified in the modified MS.

9. P3085, L21, please write down the pH value of each standard buffer.

Response: Added. They are pH = 4.01, 7.00, and 10.01 at 25.0 °C (Thermo Fisher Scientific Inc., USA).

10. P3086, L12, Reference Material is for reference, not calibration.

Response: The statement has been changed into “both DIC and TAlk determinations were referred to Certificated Reference Materials from Andrew G. Dickson's lab”.

11. P3087, L21, Instead of "small changes", please provide a number.

Response: Modified.

12. P3088, L2-4, please clarify which stations are affected. This sentence is more suitable in discussion.

Response: Thank the reviewer for reminding us. We transfer it into the discussion section.

13. P3088, L8-10, such comparison might be more appropriated for discussion.

Response: We delete the interregional comparison.

14. P3088, L13-16, please clarify the relationship between these carbonate data and the red tides.

Response: Thank the reviewer for reminding us. We add several explanations.

15. P3089, please add a topic sentence for section 3.2.

Response: Added.

16. P3091, L20-24, not clear. Please check.

Response: Since Gieskes (1969) has reported a similar temperature coefficient of pH, we deleted this confusing statement. Thank the reviewer for reminding us.

Refer to: Gieskes, J. M.: Effect of temperature on the pH of seawater, Limnol. Oceanogr., 14, 679-685, 1969.

17. Please revise the following paragraphs if they were not edited after the major revision: P3088 L17 to 28; P3090 L4 to L13; P3096 L9-L21.

Response: Done.

18. Please consider to be revised by professional English editing before the submission.

Response: Yes. We will do it before submitting the modified MS.